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# Optical characteristics of radiated multifunctional optical materials

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## ABSTRACT

Heavy metal halides have been used in variety of optical devices and systems including lasers, acousto-optic devices, wide transparency windows and doped materials for radiation sensing. The performance of optical and electronic materials and devices are affected significantly in space environment especially due to exposure of high energy radiations. We have investigated the effect of  $\gamma$ -ray radiations on multifunctional binary and ternary halide crystals in a laboratory environment. In this paper, the characteristics of mercurous halides ( $\text{Hg}_2\text{X}_2$ ,  $\text{X} = \text{Cl}, \text{Br}$ ) before and after radiation exposure are discussed. These crystals, synthesized with high purity source materials, were exposed with a  $^{137}\text{Cs}$   $\gamma$ -ray source for a period of more than 70 hours. It was observed that a  $^{137}\text{Cs}$   $\gamma$ -ray source with 5  $\mu$  curie source did not affect the optical characteristics of these materials which suggests their potential use in space-based components.

**Keywords:** Optical, Radiation, Binary Halides, Ternary Halides,  $\gamma$ -ray, Heavy Metal, High-Z

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## 1. INTRODUCTION

Crystals with high-Z materials such as thallium, mercury and lead based compounds have great potential for variety of devices and components suitable for space applications. These materials have shown excellent performance for the wavelength conversion in MWIR and LWIR regions, acousto-optic and great potential for radiation detection. The heavy metal halides especially have been studied since past several decades for variety of optical and radiological detector applications. Mercury, lead, and thallium based binary and ternary materials have unique properties [1-5]. Binary halides have been developed for acousto-optic filters, delay lines, Bragg Cells, acousto-optic imagers, and technologies have been transitioned. The mercurous halides,  $\text{Hg}_2\text{Cl}_2$ ,  $\text{PbCl}_2$ ,  $\text{Hg}_2\text{Br}_2$ ,  $\text{PbBr}_2$  and  $\text{Hg}_2\text{I}_2$ ,  $\text{PbI}_2$  are materials with high anisotropic and outstanding acousto-optic properties [1-5]. These binaries and their ternaries such as  $\text{Tl}_2\text{HgI}_4$  and  $\text{TlPbI}_2$  class have also shown excellent characteristics for radiation detection also. The crystal growth of these materials has been published extensively [6-10]. These materials transmit from the blue through the far infrared wavelength region without any absorption band. This large transparency along with very high refractive indices and large birefringence make them very attractive compounds for their applications in acousto-optic and opto-electronic devices. In summary, the properties that make this class of materials important can be summarized as (1) the transparency range, (2) Figure-of-merit due to high refractive indices, (3) the photo elastic coefficient, (4) the acousto-optic figure of merit compared to oxides, (5) acoustic velocity, and (6) high-Z, and (7) very high density. Binary halides belong to the tetragonal ( $D_{4h}$ ) and ternary to orthogonal symmetry. M. Gottlieb et al. have demonstrated that the acousto-optic figure of merit of these materials range from 700 to 3200 times compared to fused quartz. In addition, a single material can cover the operation from short wavelength (SWIR) to very long wavelength (VLWIR) region which is not possible with commercial materials such as tellurium oxide. In addition, due to extremely low acoustic velocity small crystals achieve very long delay line and very high efficiency. The overall objective of this study was to determine the effect of radiation on the optical characteristics of these materials. For this reason, materials were exposed with  $\gamma$ -ray radiations and optical performance was determined. In this paper we report the results of exposure on mercurous chloride ( $\text{Hg}_2\text{Cl}_2$ ) and mercurous bromide ( $\text{Hg}_2\text{Br}_2$ ) crystals. Figure 1 shows the crystals used for our experiments.



Figure 1. Mercurous chloride crystal is shown on the left. The right two pictures show mercurous bromide crystal.

## 2. EXPERIMENTAL METHODS AND RESULTS

The experimental arrangement for the optical studies is shown in Figure 2. The measurement set up involves a monochromator, a filter, a lens, and photomultiplier tube. The specular reflection of each sample was directed through the collection optics and subsequently removed using an appropriate colored glass filter. Figure 3 Shows the sample holder arrangement that was used to place the sample and rotate in desired orientation.

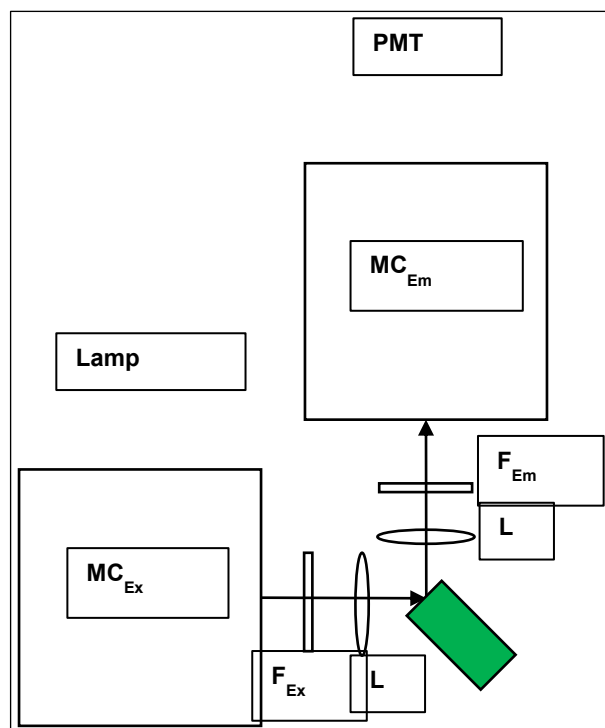


Figure 2. Schematic of collection optics



Figure 3. Custom sample holder

For each sample, data was collected at room temperature before exposing to radiation source. After measurement each sample was exposed to  $\gamma$ -ray sensor. The samples were irradiated using a Cs-137  $\gamma$ -ray source with a degradation rate of 5 curie. The total exposure time of each crystal to the  $\gamma$ -ray source was approximately 70 hours. The typical position of crystals compared to the source are shown in Figure 3. The left located crystal is mercurous chloride and the top positioned crystal is mercurous bromide. Optical density measurements were obtained using a Beckman DU 650 UV-VIS spectrometer. Spectra were acquired between 200-1100 nm. Fluorescence measurements were obtained using an Edinburgh FLS920 spectrometer equipped with a xenon arc lamp and cadmium selenide photomultiplier tube capable of NIR detection. Slits were set at 2 mm for all samples. Samples were scanned out to 1500 nm. The actual position of the mercurous chloride crystal inside spectrometers is shown in Figure 4 (a) and (b).

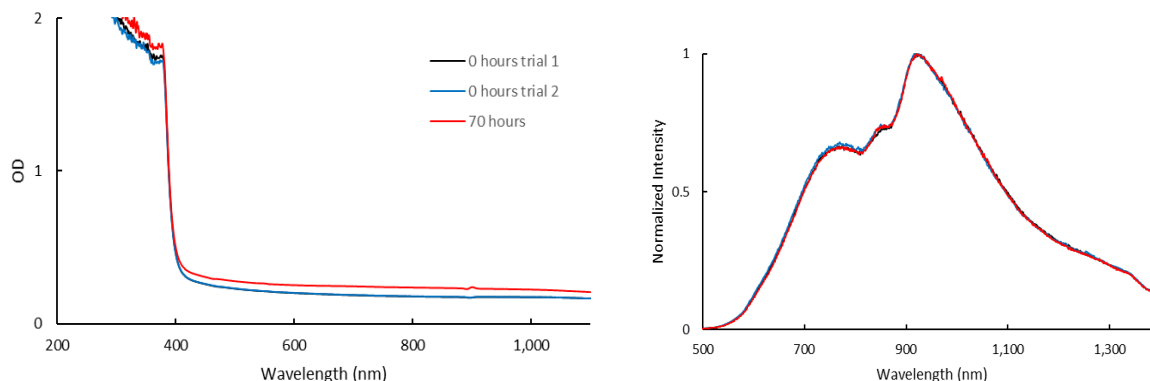


**Figure 3.** Position of crystals compared to the source. Crystals were within 2 cm of the radiation source.



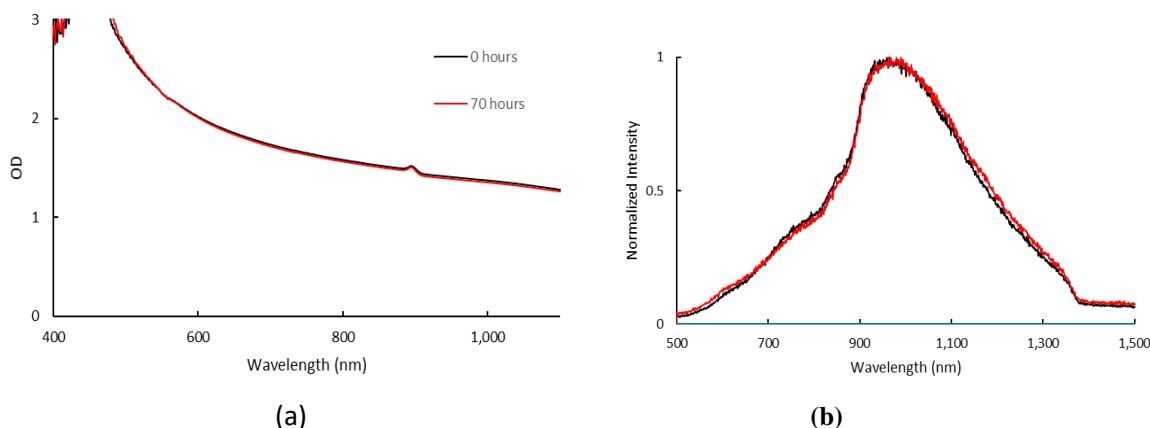
**Figure 4.** Position of mercurous chloride crystal in the measurement systems. (Left) Beckman DU 650 UV-VIS spectrometer and (Right) Edinburgh FLS920 spectrometer. Mercurous bromide crystals occupied similar positions.

The results of optical density studies for the mercurous chloride crystal before and after exposure is shown in Figure 5 (a) for the crystals without and after  $\gamma$ -ray radiation. The data for unirradiated crystals are very similar to those previously reported crystals prepared by using extremely pure source materials by Singh et al [8]. The emission data are shown in Figure 5(b) for crystal before and after radiation treatment. For this study we used excitation wavelength of 400 nm, a filter operating at 420 nm and slit width was 2 mm. We observed that the optical performance of material was totally unaffected even after extended exposure of 70 hours.



**Figure 5 (a)** Optical density (left) and emission characteristics (right) for the mercurous chloride crystals before (blue) and after (red) 70 hours of radiation.

Like the case of mercurous chloride, we placed the mercurous bromide slab in the measurement systems. The samples were irradiated using a Cs-137  $\gamma$ -ray source with a degradation rate of 5 curie for a period of 70 hours. The results of optical density studies for the mercurous bromide for the unirradiated and radiated crystal are shown in Figure 6 (a). We observed that properties for unirradiated crystals are again very similar to those previously reported crystals prepared by using extremely pure source materials by Singh et al [8] for bromide crystals. The results of the present studies for emission are shown in Figure 6(b) for crystal before and after radiation treatment. For this study also, like that used for mercurous chloride, we used the excitation wavelength of 400 nm, a filter operating at 420 nm and slit width was 2 mm. It was observed that the optical performance of material was totally unaffected by gamma ray radiation after an exposure of 70 hours.



**Figure 6 (a)** Optical density and emission data for the mercurous bromide crystals before (blue) and after (red) 70 hours of irradiation.

### 3. SUMMARY

It is well known that radiation hardened materials are needed for space components. Several high-Z and high-density crystals were chosen for the present study. Mercurous halides ( $\text{Hg}_2\text{X}_2$ ,  $\text{X} = \text{Cl}, \text{Br}$ ) have been proven for variety of optical applications including acousto-optic devices, lasers, and long delay lines. We have investigated the effect of long exposure of  $^{137}\text{Cs}$   $\gamma$ -ray source with 5  $\mu$  curie source on heavy metal halides for up to 70 hours. Optical studies were carried out in the visible and near-IR regions of the electromagnetic spectrum. It was observed that radiation did not affect the optical characteristics such as transmission, emission, and stability of materials. Small differences in intensity/counts were due to crystal inhomogeneities, normalization, and crystal alignment/mounting errors. This study is continuing for binary and ternary selenides also. Effect of longer than 70 hours exposure is not known. Further study in this direction is needed.

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