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RADIATION RESPONSE ON THE MERCURY IODIDE CRYSTALS GROWN BY THE PHYSICAL VAPOR TRANSPORT AND THE BRIDGMAN TECHNIQUES

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ABSTRACT

This paper describes the establishment of techniques for preparing HgI_2 crystals to be used as HgI_2 semiconductor detectors, operating at room temperature. Two methods were developed for the HgI_2 growth: (1) the physical vapor transport technique, using an oil bath furnace and (2) the Bridgman method. The obtained crystals for both methods were characterized and the results obtained were analyzed considering the following properties: plan of the crystal orientation, resistivity and response to the radiation.
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1. INTRODUCTION

There have been attempts to develop room-temperature X- and gamma ray semiconductor detectors for various applications. Over the past decade, compound semiconductors have attracted considerable attention as possible alternatives to Si and Ge, for charged particle and photon detection. The application of these detectors has been somewhat restricted, since Ge detectors provide high-resolution capabilities only at cryogenic temperatures (-204 °), because of their low resistivity at room temperature and Si detectors exhibit less than 20% detection efficiency for gamma rays for energies above 35 keV because of their low photon absorption coefficient from these values [1]. Thus, there has been a strong interest in developing semiconductors that have high photon stopping power and which may operate at room temperature, without sacrificing the advantages of Ge and Si detectors.

The main physical semiconductor properties required for fabrication of room temperature semiconductor detectors are: (1) high atomic number; (2) high density; (3) high absorption coefficient; (4) a band gap large enough to keep leakage currents low, at room temperature and (5) large electron and hole mobility-lifetime products, for an efficient charge collection [2, 3].

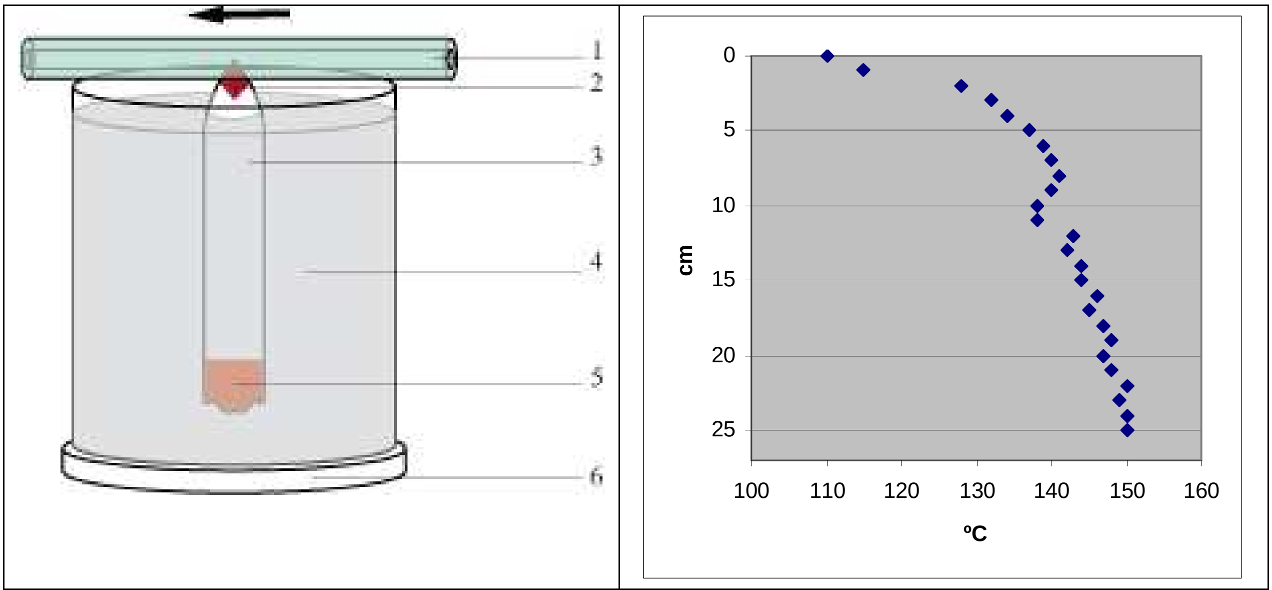
High-Z compound semiconductors, such as CdTe, $Cd_{1-x}Zn_xTe$ (CZT), HgI_2, PbI_2 and TlBr have been investigated as materials for nuclear radiation detectors that can operate at room temperature. Among these types of detectors, HgI_2 has emerged as a particularly interesting material in view of its wide band gap (2.13 eV) and its large density (7.5 g/cm^3). HgI_2 crystals are composed of high atomic number elements (Z_{Hg}=80 and Z_I=53) and with high resistivity (>10[14] Ωcm). These are important factors in applications where compact and small thickness detectors are necessary for X- and gamma rays measurements. However, the applications of HgI_2 are limited by the difficulty in obtaining high-quality single crystals and the long-term reliability problems in devices made from the crystals [3].

This work aims to develop and establish a methodology for preparing HgI_2 crystals to be used as a semiconductor detector, operating at room temperature. Two techniques were developed to the HgI_2 growth, which are the Physical Vapor Transport (PVT) technique [2,3] and the Bridgman technique [4,5].

2. EXPERIMENTAL PROCEDURE

The commercially available HgI_2 powder (Fluka Chemika), with nominal purity of 99.0%, was used as the starting material for growing crystals intended for detector applications.

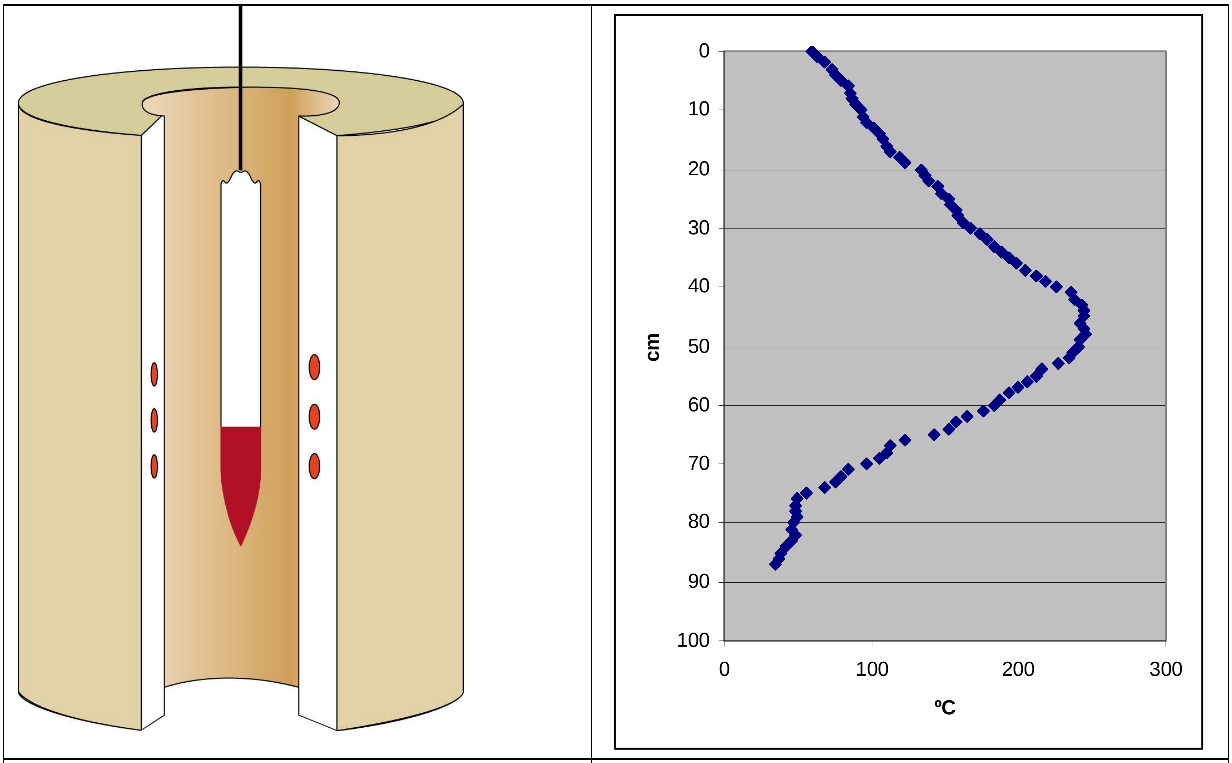
The schematic diagram of the oil-bath furnace, used for HgI2 crystal growth from PVT, and the typical temperature profile are shown in Fig. 1. The system consists of a lard oil bath, controlled by a controllable heating plate; an external heating piece was placed on the top of the growth ampoule, made of Pyrex glass. The growth was carried out in an inverted gradient, from source material residing at the ampoule bottom, using as seed of the initial nucleation obtained in the ampoule conic bottom. The maximum temperature used was 150 ºC and the crystal growth temperature was 110 ºC.



(a)	(b)

Figure 1 – Schematic diagram of the oil-bath furnace used HgI_2 crystal growth from PVT technique: 1 Cold finger, 2 crystal, 3 ampoule, 4 oil bath, 5 HgI_2 powder and 6 heater. (a) and typical temperature profile (b).

Fig. 2 shows the schematic diagram of the Bridgman furnace and the typical temperature profile used to grow the HgI_2. The crystal was grown by the vertical Bridgman technique, using a quartz crucible in vacuum. Initially, the crucibles were submitted to a rigorous chemical treatment and subsequent thermal treatment, to avoid the crystals adherence in the tube walls used in the melting. Afterward, the HgI_2 powder was introduced into a treated 20 cm long quartz tube, with 10 mm of diameter, evacuated to 10^{-6} Torr and sealed off. The crucible with HgI_2 was mounted into the vertical Bridgman furnace and the HgI_2 was melted at a temperature of 259 ºC. A crystal, around 10 mm diameter and 30 mm long, was obtained with a growth rate of 1 mm/h. The crystal was cleaved perpendicularly to direction (001), using a razor.



(a) (b)

Figure 2 – Schematic diagram of the Bridgman furnace used to HgI_2 crystal growth: 1 ampoule, 2 "hot zone", 3 crystal and 4 cold zone (a) and its temperature profile (b).

The crystalline quality and structural characterization of the HgI2 crystal were analyzed by X-ray diffraction (XRD); X-ray diffraction patterns were obtained in a Siemens (D5005) Diffactrometer, using CuKα radiation (2θ ranging from 5° to 70º).

In order to be prepared as a radiation detector, the electric contact was applied with conductive graphite painting on both sides of the wafers. Fig. 3 shows a schematic diagram of the detector and its connection to the preamplifier.

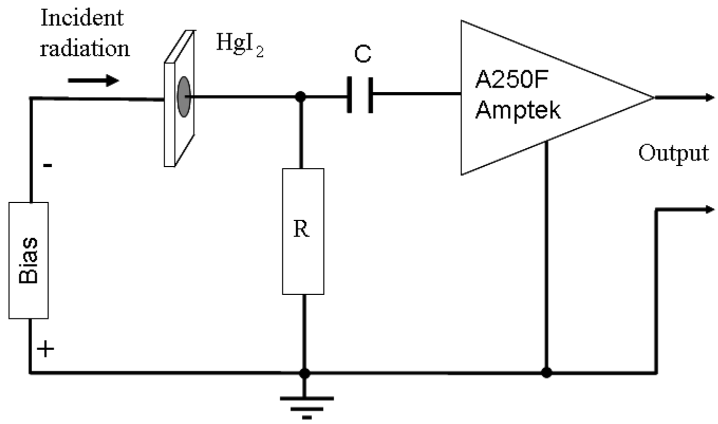


Fig. 3 – HgI_2 detector and preamplifier connection.

Fig. 4 shows the measurement schematic diagram. The experiments were carried out with the detector set inside in a box with silica gel, at room temperature. The silica gel was used to prevent the humidity at the detector surface [6]. The output from A250F charge sensitive preamplifier was connected to a 450 EG&G Ortec Research Amplifier and to a 918A EG&G Ortec Multichannel Analyzer, to obtain the pulse height spectra. The bias was supplied from 459 EG&G Ortec Bias Supply and the current measurements, for obtaining the crystals resistivity, with the 619 Keithley Electrometer. A ^{241}Am (59.5 keV, 401 kBq) gamma radioactive source was used for the measurements.

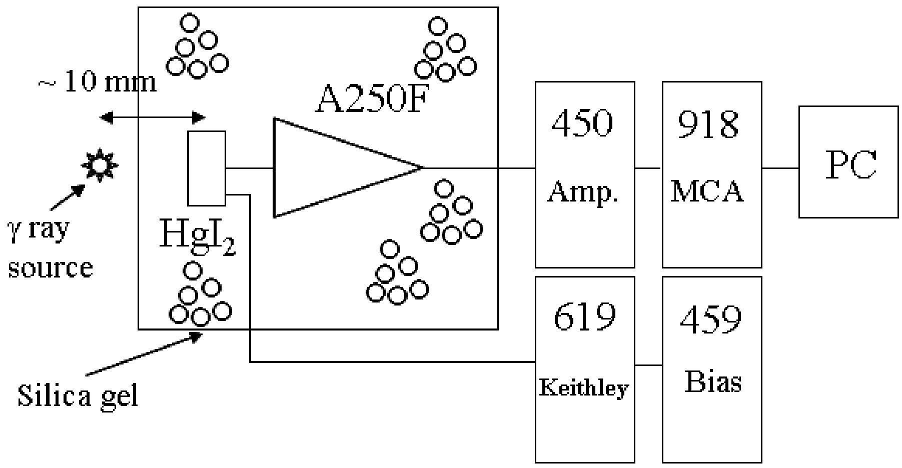


Fig. 4 – Schematic diagram for HgI_2 detector measurements.

3. RESULTS AND DISCUSSION

Fig. 5 shows the HgI_2 crystals grown by the PVT and Bridgman methods. As it can be observed in this figure, the crystal grown by PVT method (5a) presented a brilliant darker color, being more accentuated in the upper section. The crystal grown by the Bridgman method presented an opaque color along the whole length (5b). It can be, also, observed that the crystal grown by the PVT method was visually more transparent

(a)	(b)

Figure 5 - HgI_2 crystal grown by PVT method (a) and Bridgman method (b).

Fig. 6 shows the X-ray diffraction pattern of HgI_2 crystals grown by PVT method and Bridgman technique, respectively, exhibiting a complete set of reflections,. No significant difference was observed in the diffractograms between the crystals grown by both methods. The results show that the crystals have a similar structure to the tetragonal crystalline pattern of the HgI_2. The diffractogram indicates that the crystal is, preferentially, oriented in the (001) direction.

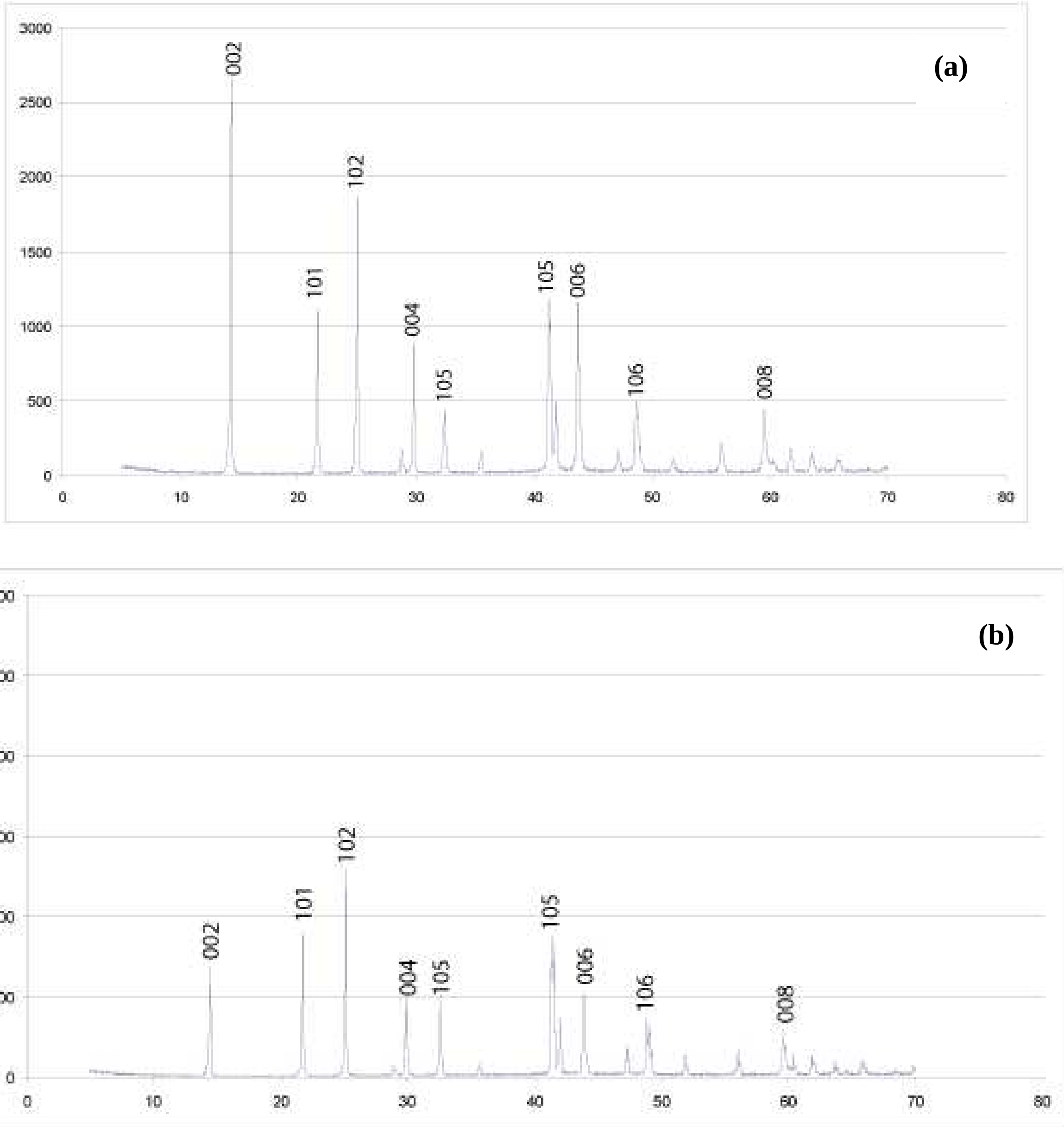
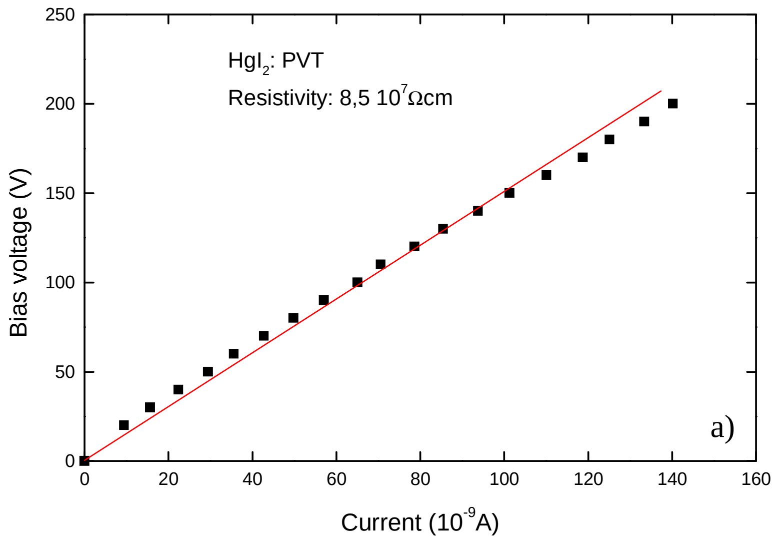


Figure 6 - X-ray diffractogram of HgI_2 grown by: PVT method (a), and Bridgman (b).

Fig. 7 shows the results of the leakage current, as a function of the applied bias. From these results, the resistivity value was inferred for each crystal. The worst resistivity result ($8.5x10^7$ Ωcm) was obtained for the crystal grown by the PVT method, compared to that of the crystal grown by Bridgman ($6.4x10^8$ Ωcm). The best charge carriers produced by radiation excitation, in the sample grown by PVT method, allowed to observe a photopeak of 60 keV gamma rays from ^{241}Am.



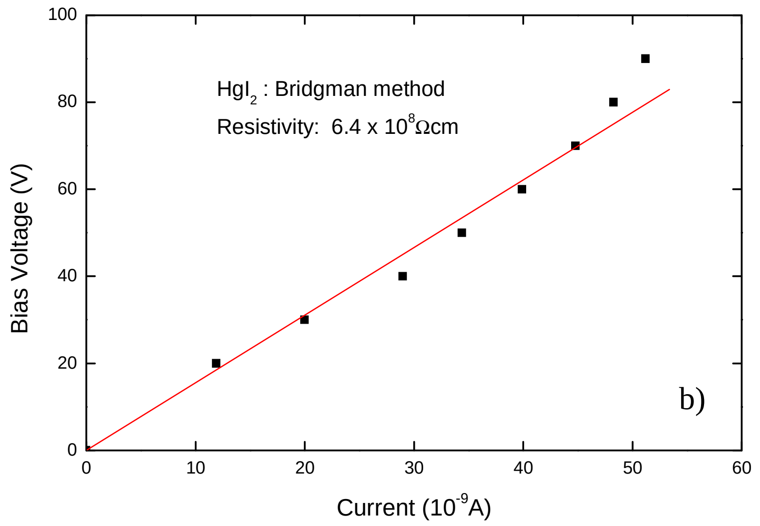


Figure 7 - Current versus voltage of HgI_2 detectors, prepared with crystal grown by PVT method (a) and Bridgman (b).

Figs.8 shows the pulse height spectrum obtained for the crystals grown by the PVT method using a gamma ^{241}Am source for the crystal excitation. The bias used was 200 V and the spectrum counted for 3000 s.

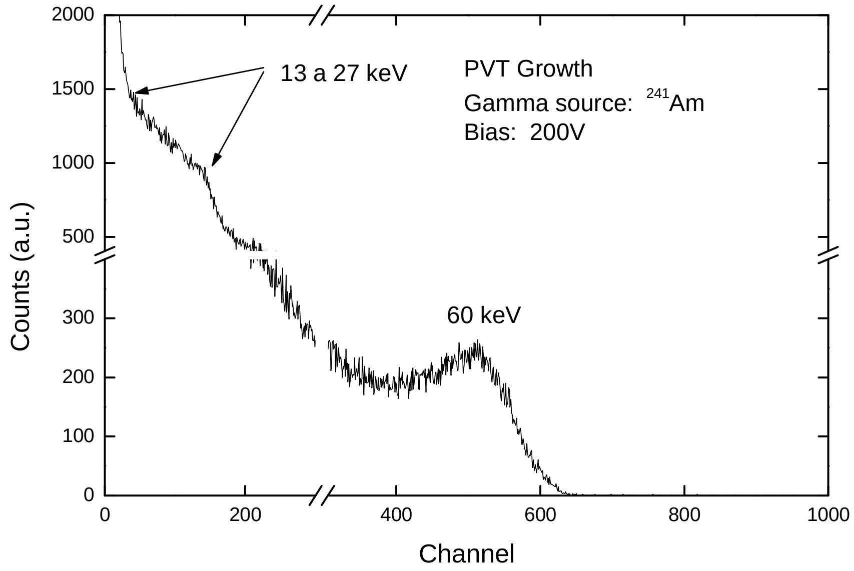


Figure 8 - HgI_2 detector energy spectrum with gamma rays from ^{241}Am excitation of the crystal, grown by the PVT method.

For the HgI_2 crystal grown by Bridgman method, it was possible to observe the radiation response only in the current mode. For these detectors, the pulse mode was not observed due to a low radiation response and a high noise signal. On the other hand, as it can be observed from the Figure 8, the detector prepared with the HgI_2 crystal grown by PVT presented pulse height with recognizable features under gamma ray excitation, although the resolution was poor. The better radiation response of this crystal, despite its worse resistivity compared to that the crystal grown by Bridgman may be attributed to its better crystallinity, which permitted to collect enough charge to identify the photopeak.

To obtain a better resolution is necessary to improve the charge collection [7]; to aim this goal, purification should be carried out in order to reduce the impurities and reduce the charge traps. Further studies should be made to evaluate the influence of the crystals impurities and cristallinity in the detector performance.

4. CONCLUSION

Preliminary results indicated that the PVT and Bridgman techniques are suitable to HgI_2 crystal growth. The HgI_2 crystal grown by PVT presented pulse height with recognizable features under gamma ray excitation, although the resolution was poor. On the other hand, for the HgI_2 crystal grown by Bridgman method, it was possible to observe the radiation response only in the current mode. For these detectors, the pulse mode was not observed due to a low radiation response and a high noise signal.

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