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GAMMA-RAY SPECTROSCOPY FOR TlBr DETECTORS AT TWO DIFFERENT TEMPERATURES

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ABSTRACT

In this paper, the results of the development of the TlBr poly-crystals for application as gamma radiation detectors are presented. TlBr detectors were fabricated from the crystals purified by the multipass zone refining method and grown by the Bridgman method. 0.3mm thick detectors were prepared and systematic measurements of the current-voltage and height pulse spectra were carried out. The spectrometric performance of the TlBr detector was evaluated by ^{241}Am (59.5keV), ^{133}Ba (80keV) and ^{57}Co (122keV) gamma-ray excitation sources at room and -20°C temperatures. No significant difference was observed in the energy resolution between measurements performed in these two different temperatures. The stability of TlBr radiation detectors at reduced temperatures was slightly better than at room temperature.

1. INTRODUCTION

Thallium bromide has been subject of many investigations due to its specific technological features, for instance, its large applicability, at room temperature, as a gamma-ray semiconductor detector and photodetector coupled to scintillators crystals. The development of these devices requires good quality substrates [1]. However, the TlBr detectors development has been somewhat hampered by the problems for controlling residual impurities, stoichiometry and physical defects in the crystals [2].

The TlBr is considered a promising material to be used as gamma-ray spectroscopy due to high density (7.5g/cm^3) and high atomic numbers (Tl=81 and Br=35). It exhibits photon stopping power higher than other semiconductor materials used for radiation detector fabrication, such as CdTe, CZT and HgI_2. Additionally, it has a large bandgap (2,68eV), which minimize the thermal generation of charge carriers and thus low noise performance is obtained [3,4].

The low melting point (480ºC) and unique solid phase of TlBr are advantageous for melt purification and crystal growth [5]. TlBr crystals are relatively soft, with a knoop hardness number of 12 [1,6]. This parameter is important to be considered from the viewpoint of device processing, because a soft semiconductor requires more carefull handling techniques.

Several studies [3,4,7] have been carried out about the preparation of the TlBr semiconductor and progresses have been made to improve of the purification and crystal growth techniques. Although the specific underlying causes of this detector failures are not yet completely understood, it is believed that impurities are still one of the primary limiting factors in thallium bromide. Hitomi et al. [5,7] have demonstrated that TlBr crystals properly purified by zone refining and subsequently grown from the melt present spectrometry with good energetic resolution for X and gamma-rays.

In the recent literature [1,3,8] it is observed that, despite considerable performance improvement in the TlBr detectors, several problems remain limiting progress. Besides the impurities, another main problem [9] is the low time stability of detector parameters obtained for the most crystals. Also in CdTe and HgI_2 detectors, the time instability causes degradation of the spectroscopic characteristics (energy resolution and efficiency) during the measurements. The reason for this time instability is not clearly understood and can vary among materials.

The latest studies with TlBr detector [1,3,8,10] have been accomplished at an optimal temperature range from 0ºC to –30ºC. Cooling the detectors is one of the most efficient ways to reduce the detector leakage current [10]. The authors [1,3,8,10] suggest that reduction of the detector leakage current would improve the energy resolution of the TlBr detectors.

In this paper, the TlBr crystal results grown by the Bridgman method from purified materials by zone refining technique are presented. To evaluate the crystal as a radiation semiconductor detector, measurements of the resistivity, radiation response of gamma rays and current characteristics on time at -20ºC were carried out. The spectrometric performance of the TlBr detectors was evaluated by the gamma-ray excitation, from ^{241}Am (59.5keV), ^{133}Ba (80keV) and ^{57}Co (122keV) sources, at room and -20ºC temperatures.

2. DETECTOR FABRICATION

To produce pure TlBr crystals, commercial TlBr material (99,0%) was purified by zone refining process. 30 zone refining passes were carried out in a furnace at the speed of 5 cm/hr. The refined material has shown to be very pure, with impurities concentrations of <10ppm, which were evaluated by Inductively Coupled Plasma Mass Spectroscopy technique. The purest sections of TlBr purified material were used for Bridgman crystal growth. TlBr crystal with a 1cm diameter and 3cm long was obtained for detector characterization.

The crystal was cut using a diamond saw in low speed, in order to have less damage and smaller depths in the resulting layer. The transversal (110) oriented wafers with dimensions 1 x 1cm^2 were mechanically polished and etched with Br-methanol solution (10%) to an optically smooth and transparent surface. Thermal annealing, at 100ºC, was also performed to improve the crystallinity of TlBr slices.

Radiation detectors were fabricated by Au vacuum deposition on both sides of the wafers. 0.11mm diameter Cu wires were attached to Au electrodes using a colloidal graphite suspension. The detectors were assembled inside of an aluminum box coupled to the Amptek A250 charge sensitive preamplifier. The radiation response of the detectors was studied using the conventional electronic setup including the Ortec 556 voltage power supply, Amptek A250 charge sensitive preamplifier, Ortec 450 amplifier and an oscilloscope. The detectors were glued to a Peltier cooler (8 X 8mm^2), capable of cooling the device to about -20ºC. The pulse height spectra were analyzed using an EG&G Ortec model 918A multichannel analyzer, and ^{241}Am (59.5keV), ^{133}Ba (80.0keV) and ^{57}Co (122keV) gamma radiation sources. Each source was placed outside the Al box, about 1cm from the detector. The set up for the detector performance measurements was kept constant (i.e., acquisition time of 1000 counts, bias of 100V, shaping time of 3μs, amplifier gain of 200 and collecting holes).

3. RESULTS AND DISCUSSIONS

Fig. 1 shows the current x voltage curves obtained from the detector with and without the Peltier cooler. This measurement was performed with the purpose of evaluating the effectiveness of the Peltier cooler on reduction of the detector leakage current. From the I-V characteristics, typical resistivities were found to be about 4 x $10^{10}\Omega$ cm. At biases of a 100V, the average recorded leakage currents were ~26nA at room temperature and ~20nA at -20ºC.

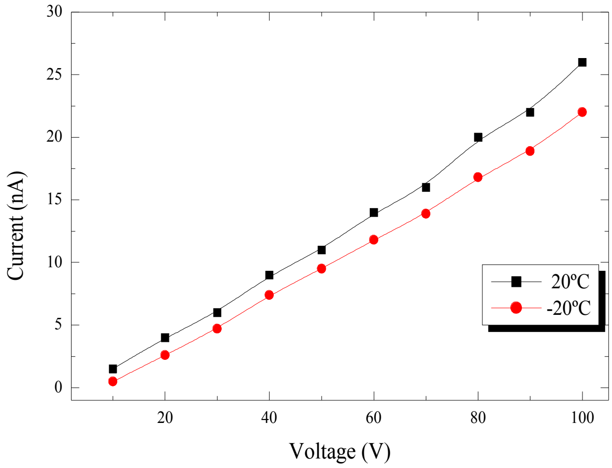

Figure 1. The current-voltage characteristics obtained from the same TlBr detector with and without Peltier coller.

Fig. 2 and 3 show the ^{241}Am, ^{133}Ba and ^{57}Co gamma-ray spectra obtained from the same detector. In the Fig. 2, the detector was operated at room temperature and the Fig. 3 at -20ºC. Energy resolutions of 14, 17 and 23keV at room temperature and of 13, 15 and 24keV at reduced temperature were obtained at input energies of 59.5, 80 and 122keV, respectively (Table 1).

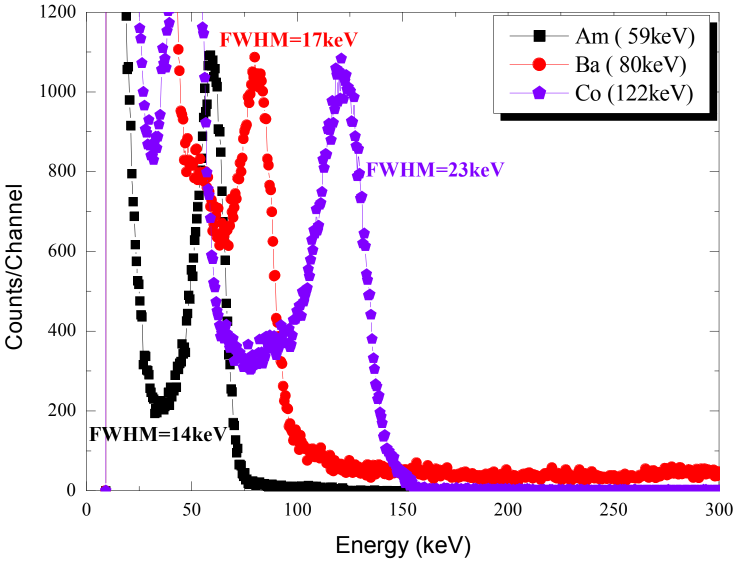

Figure 2. ^{241}Am, ^{133}Ba and ^{57}Co spectra obtained from the 300µm thick TlBr detector at room temperature.

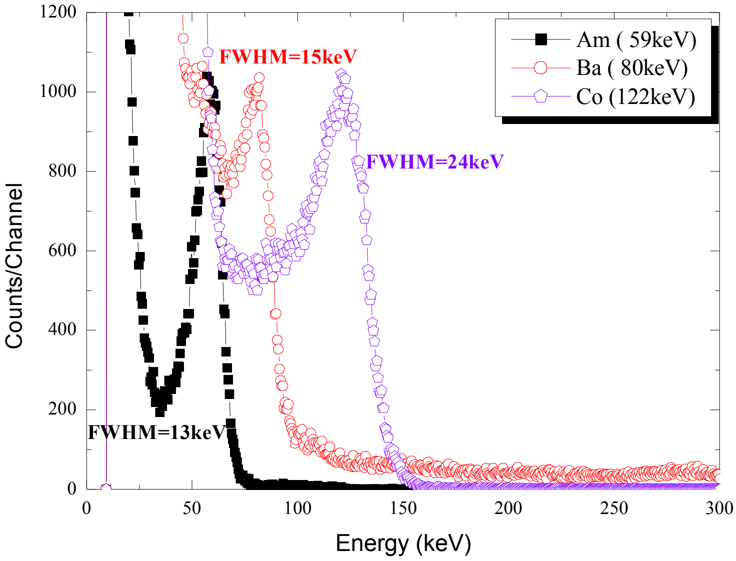

Figure 3. ^{241}Am, ^{133}Ba and ^{57}Co spectra obtained from the 300µm thick TlBr detector at reduced temperature (-20ºC).

Table 1. Energy resolutions of the TlBr detectors at 20ºC and - 20ºC.

Temperature and Source	FWHM (keV)	Resolution (%)
20ºC		
^{241}Am (59.5keV)	14	24
^{133}Ba (80keV)	17	21
^{57}Co (122keV)	23	19
-20ºC		
^{241}Am (59.5keV)	13	22
^{133}Ba (80keV)	15	18
^{57}Co (122keV)	24	19

No significant difference was observed in the energy resolution between measurements performed at room temperature and -20°C. Onodera et al. [11], also cooled the detector at -20ºC and they obtained detector with resolution of 3.3keV (5.5%) at 59.5keV energy, while for the room temperature the resolution was of 2.6keV (4.4%). The resolution results presented by the authors were better than that obtained in this work. However, cooling the detector, no significant improvement in the resolution was also observed by those authors. In spite of not having achieved an optimum spectroscopic resolution, for applications where a radioisotope with well-known energy is used, like in surgical probes for nuclear medicine, the resolution obtained for the our developed TlBr detector is suitable.

Despite the reduction of the leakage current was not so significant as expected either (Fig.1), it was possible to observe experimentally the stability improvement of this detector at reduced temperature. The stability is an important parameter to be evaluated for its application as a radiation detector. The decrease in the leakage current in function of time was observed along the measurements (-20ºC), as shown in Fig. 4. This behavior may be attributed to the polarization effect [1,8,12]. This term is commonly used to refer to any change in the performance of a radiation detector along the time, not correlated with changes in operating parameters [1].

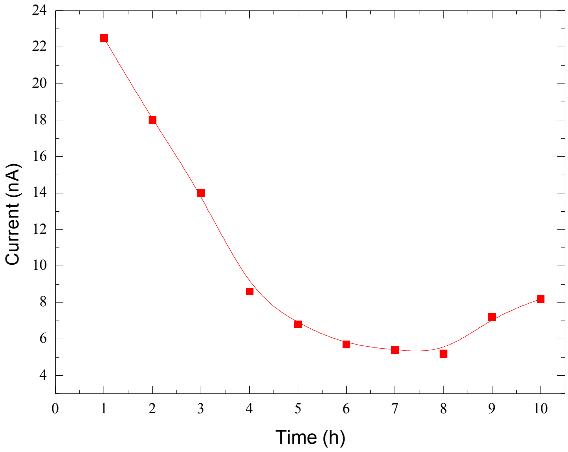

Figure 4. The current characteristics on time obtained from the TlBr cooled detector.

At room temperature, the detector presented the polarization effect on time scales ranging from minutes to hours, while at reduced temperature it was observed ranging from hours to days. After the bias removal some detectors have not recovered the initial characteristics, requiring a new annealing treatment. The same behavior has been also observed by several authors [1, 8, 12]. The polarization effects were not observed in the detector operated until 3 hours at room temperature, and until 9 hours at -20°C. After these times, the polarization effects caused the degradation of the detector resolution due to the random amplitude and time transients generation. Additional studies have been carried out in order to understand the current reduction in function of the time. According to Ponpon et al. [13] the resulting time dependence of the current is directly related to the accumulation and release of charges on one (or more) trap level (s). This phenomenon, observed in wide bandgap semiconductor used for spectroscopy, has considerable consequences on devices properties, especially for the nuclear radiation detectors.

4. CONCLUSIONS

Thallium bromide radiation detectors have been fabricated from the grown crystals and spectroscopic performances have been evaluated from the γ- ray detectors at room and -20°C temperatures. The reduction of the leakage current was obtained for the TlBr detector cooled with a small Peltier cooler. Similar resolution results have been obtained for both operation temperatures, although the stability was slightly improved at lower temperatures. Nevertheless, the TlBr detector stability is still a problem and therefore the investigation on the degradation phenomena and the property improvement are important parameters to be studied.

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