INFLUENCE OF CRYSTALLINE SURFACES ON THE TLBR RADIATION DETECTOR PERFORMANCE

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Abstract - **Thallium bromide (TlBr) is an important material for room temperature detectors. Due to its high photoelectric absorption efficiency and large bandgap, thallium bromide is a good candidate for X- and γ-ray spectrometry. In this study, TlBr detectors were fabricated from the crystals purified by the multipass zone refining and grown by the Bridgman method. Detectors were prepared using TlBr wafers of about 0.3mm thick, with surface submitted at different mechanical and chemical treatments. The results of surface quality of TlBr wafers, evaluated by scanning electron microscopy, are presented. Spectrometric performance of the TlBr detector was assessed by excitation with ^{241}Am gamma-ray source at room temperature. The dependence of the radiation on the detector was affected by the condition of the crystalline surface. This study has important implications for adequate processing of TlBr surfaces for radiation detector applications.**

I. INTRODUCTION

Thallium Bromide crystals are semiconductors composed of high atomic number elements (Z_{Tl}=81 and Z_{Br}=35), with high resistivity (>10^{10}Ωcm) and density (7,5g/cm^3). It has been the subject of many investigations due to specific technological features. Its good response to X and γ-rays at room temperature makes it suitable for a number of applications such as radiation detector, photodetector and small dimension devices with high radiation efficiency like intra-operable surgical probes [1].

The performance of radiation detectors is controlled by both intrinsic and extrinsic factors. Carrier lifetime, mobility and the atomic number of the material used for radiation detectors represent intrinsic parameters, while factors such as crystallographic perfection and impurity levels can also play a major role in the ultimate performance of radiation detectors [2]. TlBr has a wide band gap and a high density. Neverthless, the crystals have low electron and hole mobility and significant hole trapping [3]. Because of these crystalline characteristics, efficient detectors can only be obtained with thin samples of less than 1mm thick. Although it is harder than other semiconductors (PbI_2 and HgI_2), TlBr is not sufficiently hard (Knoop hardness of 12) to prevent damage during its fabrication process [4]. TlBr is also highly toxic and must be handled with care [5].

The crystal cutting, surface polishing and subsequent etching are important processes during the manufacturing of radiation detectors, such as CZT and TlBr. The routine procedure to fabricate TlBr detetor involves mechanical polishing of the crystal followed by chemical etching with a bromine methanol solution. In some circumstances the material exhibits a negligible polarity effect. However, polarity effects can be introduced by surface processing and effectively removed by appropriate polishing and chemical etching. Careful study of this polarity effect is necessary to optimize the surface treatment process, and it is also helpful to improve the detector design and performance [6].

In this work, the surface quality of the TlBr wafers prepared at different mechanical and chemical were evaluated. The influence of the quality surface in the detector performance was also evaluated. Spectrometric response of the TlBr detectors was assessed by excitation with ^{241}Am gamma-ray source at room temperature.

II. EXPERIMENTAL

To produce pure TlBr crystals, commercial TlBr material (99,0%) was purified by zone refining process. 20 zone refining passes were carried out in a furnace at the speed of 2 cm/hr. A small section of TlBr purified material was used for Bridgman crystal growth. Details of the purification and crystal growth were described in our previous paper [7]. TlBr crystal with a 1cm diameter and 3cm long was obtained for detector characterization. The crystal purity was evaluated by ICP-MS technique. The impurities concentrations found were <10ppm.

The crystal was cut using a diamond saw in lower speed, in order to have less damage and smaller depths in the resulting layer. Usually, the crystal cutting process causes a large number of defects distributed along the resulting layer depths [4]. The slices with dimensions 1 x 1 x 0.03 cm^3 were polished using abrasives of 12 to 3μm to thin down and smooth them. In order to remove damage from the crystal cutting, wafers surfaces were then etched using a 10% bromine in methanol solution and rinsed with a methanol solution. So, the crystalline slices are polished to delete the defects on the surface arising at the cutting and the etching solution is used to remove the damages caused after the mechanical procedure. This etching provides a suitable crystal surface for electric contacts deposition [7]. Thermal

Manuscript received October 18, 2004. This work was supported in part by the Fundação de Amparo à Pesquisa do Estado de São Paulo – FAPESP under Grants 01/09049-5 and 02/10195-9.

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annealing was also performed to improve the crystallinity of some slices, at 100ºC.

The crystal was cut in 8 slices and each slice suffered different treatments before the detector preparation. The following treatment procedures were carried out:

1st slice: no polishing (Po) and no chemical etching (CE), named M20-1;

2nd: with Po and no CE, (M20-2);

3rd: no Po and with CE for 10 sec., (M20-3);

4th: with Po and with CE for 10 sec., (M20-4);

5th: with Po and with CE for 30 sec., (M20-5);

6th: with Po and with CE for 10 sec., (M20-6). The Br-methanol solution was prepared 5 days before;

7th: with Po, with CE for 10 sec and thermal annealed for 5 days at 100ºC prior to gold deposition, (M20-7); and

8th: no Po, with CE for 10 sec and thermal annealed for 5 days at 100ºC prior to gold deposition, (M20-8).

Then, the simplified typical steps of the detectors fabrication consist of the following technological operations: (a) the cutting of crystals; (b) the input inspection for defects and material parameters; (c) the polishing of the selected slices; (d) chemical etching of the wafers; (e) the deposition of contacts; (f) the assembly of the detectors and (g) the measurement of final detectors performance [4]. Each stage can affect the crystal quality and introduces its own defects and imperfections [8].

The crystal surfaces after mechanical (cutting and polishing) and chemical treatments were examined by scanning electron microscopy (SEM). Radiation detectors were fabricated using gold vacuum deposition on both sides of the wafers. The TlBr detector was characterized by measuring its spectral responses using ^{241}Am gamma radiation source.

III. RESULTS AND DISCUSSIONS

Figs. 1 to 4 show the micrographs of the TlBr surface prepared with different treatments by SEM. Each slice was examined in wholly and its uniformity was verified.

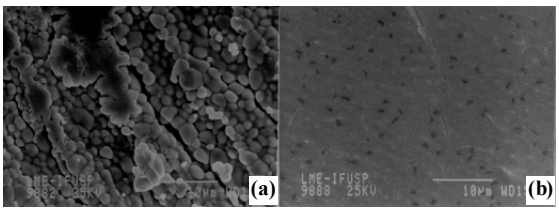

Fig. 1. SEM micrographs of TlBr crystal (X3500) (a) no Po and no CE: M20-1 and (b) with Po and no CE: M20-4.

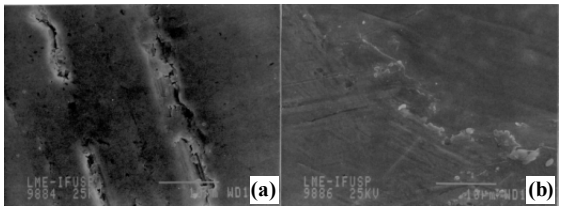

Fig. 2. SEM micrographs of TlBr crystal (X3500) (a) Po and no CE: M20-2 and (b) no Po and CE for 10 sec: M20-3.

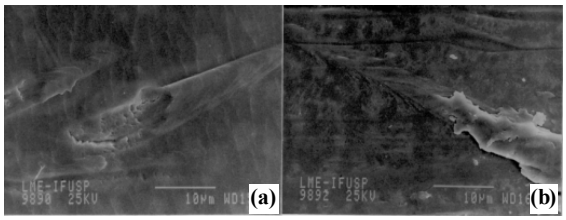

Fig. 3. SEM micrographs of TlBr crystal (X3500) (a) Po and CE for 30 sec: M20-5 and (b) Po and CE for 10 sec after 5 days of preparing of the Br-methanol solution: M20-6.

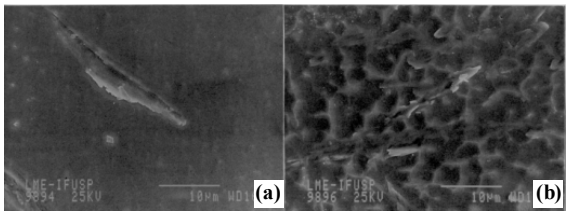

Fig. 4. SEM micrographs of TlBr crystal (X3500) (a) Po and CE for 10 sec and annealed by 5 days: M20-7 and (b) no Po and with CE for 10 sec and annealed for 5 days: M20-8.

The purpose of evaluating the surfaces etched with a Br-methanol solution prepared five days before was to verify the possible degradation of this solution. As the preparation of this solution is a hard and dangerous task, due to the Br toxicity, depending on the results of the detector performance, it would be no necessary to prepare the solution at each chemical treatment. Thus, many detectors could be prepared in different days, using the same solution for chemical etching.

The TlBr morphology and defects caused by diamond saw on the surface of the crystal cut, without polishing, can be observed in Fig. 1a. The method and the quality of the cutting have great impact on the depth of these layers and defects concentration in them [4]. Knoop hardness of TlBr is only 12 units, so during the cutting and polishing of the small thickness crystalline wafers, the plastic deformation can take place. Gostillo et al. [4] suggest that to avoid this deformation TlBr crystal should be glued to the special hard substrate. The manual polishing was slower mechanically allowing to reduce the risks of a deep scratch or chips, as well as other major defects of a mechanical processing. The chemical etching have been used to remove strains and defects.

As it can be observed in Figs. 1 to 4, there are significant differences in the resulting surfaces under varying conditions. Some samples were fractured using N_2(l), after detector performance measurements, to verify if the polishing and etching treatments caused damages in the material morphology. This task was also difficult due to plasticity this material. Using SEM to study the inner section of the slices, we confirmed that there was no alteration in the internal morphology, after the procedures.

However, both mechanical polishing and chemical etching can affect the surface leakage current. The centers resulting from mechanical polishing may both enhance the carrier recombination on the surface by increasing surface trapping sites and affect the surface leakage current by

providing more conductive pathways and changing the electric-field distributions [6].

The typical γ-ray spectrums of ^{241}Am obtained from the TlBr detectors are shown in Figs. 5 to 8. A bias voltage of 100V and a shaping time of 3μs were applied. The FWHM of the 59keV peaks for TlBr detectors were about 28.3keV (47.9%) to M20-1; 38.9keV (65.9%) to M20-2; 31.8keV (53.9%) to M20-3; 19.4keV (32.8%) to M20-4; 21.8keV (36.9) to M20-5; 31.8keV (53.9%) to M20-6; 13.5keV (22.8%) to M20-7 and 36.6keV (62.0%) to M20-8. The width of the peaks may be due to incomplete charge carrier collection and polarization effects.

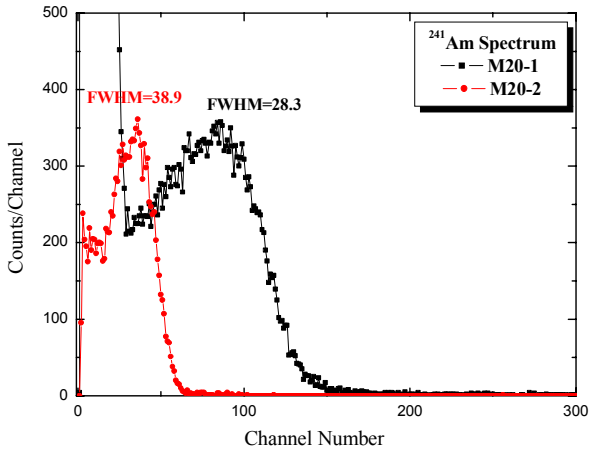

Fig. 5. ^{241}Am spectrum obtained from M20-1 and M20-2 TlBr detectors.

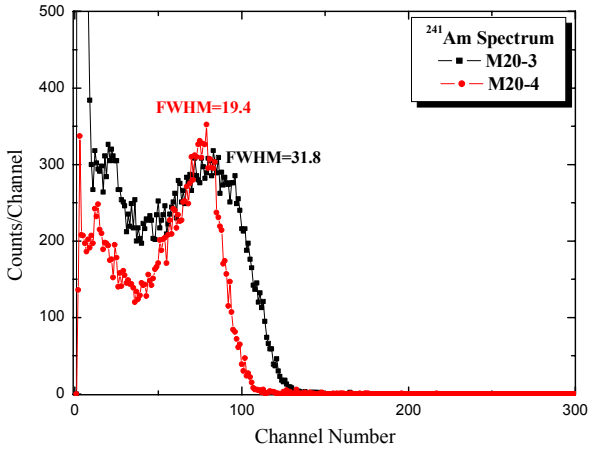

Fig. 6. ^{241}Am spectrum obtained from M20-3 and M20-4 TlBr detectors.

The best results obtained were with the detectors produced from wafers with polishing and etching for 10 seconds, and also with thermal annealing (M20-4: Fig. 6 and M20-7: Fig. 8, respectively).

In the M20-3 and M20-4 spectra can be observed a ^{241}Am peak with energy of about 27keV. The others spectra were of different profiles – no marked contrast was observed between the shape of the 59.5 keV and that of minor energy peak.

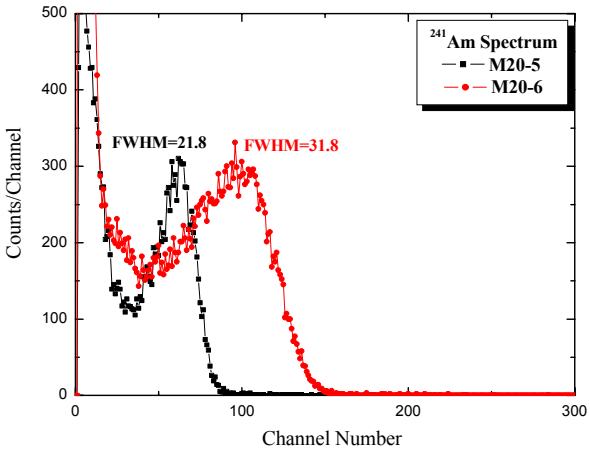

Fig. 7. ^{241}Am spectrum obtained from M20-5 and M20-6 TlBr detectors.

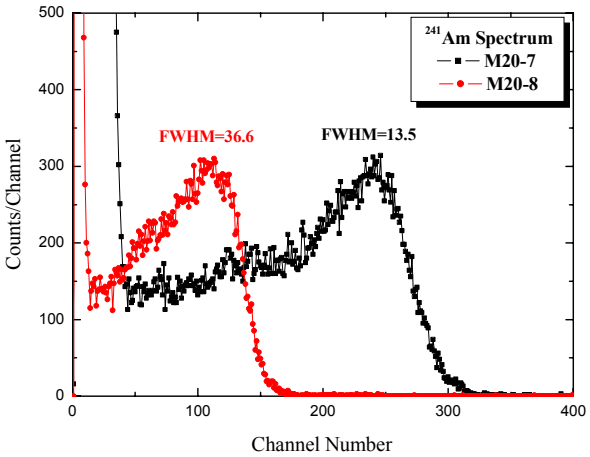

Fig. 8. ^{241}Am spectrum obtained from M20-7 and M20-8 TlBr detectors.

However, more studies should be carried out to characterize several variables that can affect the surface uniformity. Experiments have been performed to verify the influence of the surface quality in the detector response, as well as if the structural defects limit the performance of TlBr detectors, such as chemical impurities. The influence of chemical impurities on TlBr detector performance was shown in our previous work [7].

Nevertheless, the M20-4 and M20-7 detectors have shown good performance as nuclear radiation detectors at room temperature. However, to obtain a better resolution is necessary a complete charge collection [9], which was not completely observed in our measurements. The origin of the charge trapping is believed to be a direct consequence of the relative softness of TlBr. According to Owens et al. [10,11] any mechanical treatment, as cutting and polishing, generates a high concentration of intrinsic structural defects by local deformation limiting the performance of a TlBr detector. Therefore, it is necessary careful handling and suitable surface treatment procedures for detectors production. Further improvement of the detector performance can be achieved optimizing the mechanical treatment, investigating the structural and surface properties more deeply and reducing electronic noise in the detection system. Because of the TlBr soft hardness, somewhat plastic nature, we are

improving our fabrication procedures in the hope of preventing fabrication stress-induced crystal defects.

IV. CONCLUSION

Preliminary results show that the best detector responses are obtained from the detector prepared with the slices polished and chemically etched for 10sec. The best overall energy resolution and sensitivity to gamma-rays was achieved by the detector annealed after the polishing and chemical etching. A significant improvement in the detector characteristics was obtained by annealing. It was found that the slices without polishing and with etching by 10 sec had similar results to those polished and etched with a solution prepared 5 days before the treatment. The slices without polishing and etching resulted in the decrease of the charge collection efficiency.

Thus, the importance of polishing and etching in the TlBr crystalline surfaces, for application as a radiation detector, was demonstrated by the spectrometric results found.

Despite not having a good resolution, for applications where a radioisotope with well-known energy is used, like in surgical probes for nuclear medicine, this TlBr detector shows to be suitable.

Further works are needed to establish the optimal surface processing conditions. The results of such investigations would improve the development of TlBr crystal growth technology for obtaining detectors with stable spectroscopic performance.

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