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Influence of multi-depositions on the final properties of thermally evaporated TlBr films

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ABSTRACT

Thallium bromide is a promising candidate material for photodetectors in medical imaging systems. This work investigates the structural, optical and electrical properties of thermally evaporated TlBr films. The main fabrication parameter is the number of depositions. The use of sequential runs is aimed to increase the thickness of the films, as necessary, for technological applications. We deposited films using one–four runs, that led to maximum thickness of about 50 μm. Crystallographic and morphological changes were observed with varying deposition runs. Nevertheless, the optical gap and electrical resistivity in the dark remained constant at about 2.85 eV and $10^9\,\Omega$ cm, respectively. Thicker samples have a larger ratio of photo-to-dark signal under medical X-ray exposure, with a larger linear region as a function of applied voltage. The results are discussed aiming at future technological applications in medical imaging.

1. Introduction

Thallium bromide (TlBr) is a semiconductor material with simple cubic structure, as CsCl, with lattice parameter $a=3.97$ Å [1]. It has also a large optical band gap of 2.68 eV [2] and a high stopping power for X-rays and γ-rays [3], mainly due to its high atomic number (Tl=81 and Br=35) and its mass density of 7.56 g/cm^3 [4]. Due to these properties, thallium bromide is a promising material to be used as a detector in medical imaging systems that use ionizing radiation [5–9].

For applications in medical imagers, large area detectors such as 40×40 cm^2 are needed [10]. For that aim, the film form is the most suitable. Nevertheless, most of the research up to date has only focused on the production of bulk millimeter-sized crystals [11,12]. In previous investigations, the alternative film deposition methods have been explored such as spray pyrolysis [13] and thermal evaporation [14]. They have even been compared with better results obtained from the last case [15]. In addition, for the thermal evaporation technique, an important deposition parameter such as the distance between evaporation-boat and substrate was already deeply investigated. The data have shown that larger separations lead to (i) smaller deposition rates; (ii) a final material with a better structure and morphology, and (iii) larger ratios of the photo-to-dark currents when the system is irradiated under X-ray mammographic energy range [14].

The aim of the present paper is to contribute for the continuing optimization of subject. We extend the investigation to another fabrication parameter using the thermal evaporation technique, i.e. the number of depositions. The aim is to investigate variations in the final properties of material as the optimized thickness for ionizing radiation detection is achieved. The samples are structurally and morphologically characterized and selected films are also investigated as photosensors using a mammographic system.

2. Experiment

Thermally evaporated TlBr films were produced on Corning glass 7059 and quartz substrates (1 cm^2). The source to substrate distance was kept at 3 cm and the boat current was about 58 A, that lead to boat temperatures below 400 °C, according to the previous calibration of the system. The chamber has a total volume of 4 L and the base pressure before deposition was about 10^{-5} mbar. We studied the influence of an important deposition parameter on the final properties of the samples, i.e. the number of depositions (n). The main focus was to obtain thick samples, aimed for the medical imager detectors. For that, 0.25 g of starting powder (Merck 99.9%) was placed on top of the tungsten crucible for each run. Films were fabricated using one–four deposition runs in order to increase the final thickness of the samples. The substrates were held at room temperature, which was not monitored during deposition. Samples were produced in triplicate

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and the corresponding deviations are taken into account in the error bars of the following figures.

The morphology of the samples was investigated using scanning electron microscope (Zeiss, EV050), with an accelerating voltage of 20 kV. Elemental microanalysis was based on energy dispersive spectroscopy (EDS). An IXRF accessory, 500 Digital Processing model was attached to the SEM system for that purpose.

The crystalline structure of the samples was studied by X-ray diffraction experiments, using the Cu anode K_α radiation of a SIEMENS D5005 equipment (wavelength of 1.5406 Å). The scanning was performed with steps of 0.02°C, using coupled θ–2θ configuration. The optical gap of the samples was inferred from experiments of optical transmittance as a function of wavelength (Beckman Coulter, DU 640 model UV–vis spectrophotometer).

Electrical resistivity in the dark, at room temperature, was estimated from current versus voltage experiments (Hewllet Packard 4140B). For electric transport measurements, carbon contacts (about 1 cm apart) were applied on the surface of the samples using the coplanar configuration. Selected samples were exposed to a medical diagnosis X-ray system (General Electric, Senographe 500T), in the mammography energy range. The X-ray tube accelerates electrons under 30 kVp towards a molybdenum anode (k_α and k_β of 17.5 and 19.6 eV, respectively). The total exposition time and dose were controlled using 50 mAs.

3. Results and discussion

Morphologies of the samples fabricated using $n=1$ and 4 are presented in Figs. 1(a) and (b), respectively. For all the samples, regardless of the number of depositions, a structure with grooves and organized grains is observed. These grooves become more pronounced with increase in deposition steps. Their extension and diameters seem to increase for large number of depositions. This might indicate that new deposition runs are not capable to properly cover the holes that originate these undesired grooves. The grains also increase in size with increase in deposition runs. This might indicate some degree of coalescence between preexisting grains of the previous depositions.

Fig. 2(a) presents the picture of a tilted SEM of a sample fabricated using $n=4$. Some features are observed at the top of the surface of the sample. These might be due to the evaporation of agglomerates of the original powder, as already discussed in Ref. [14]. The columnar-like growth can be better seen in cross-section SEM presented in Fig. 2(b). White arrows are presented in order to visualize the preferential growth orientation of each run. A maximum variation of up to 12° was observed between all the runs. This might be either due to the format of disposition of the original powder inside the evaporation-boat, or to a larger extent, to the influence of the grains and grooves of the surface of the samples in the previous runs. Fig. 2(c) shows that the final thicknesses of the samples scale linearly with the number of deposition, as expected. Considering that each deposition takes about 3 min, a final deposition rate of about 5 μm/min was achieved. Note that this is a reasonable value for industrial applications. The EDS analysis of the original powder revealed a Br:Tl peak ratio of about 0.74. For the case of the final films, this number is reduced by about 10%. This variation was already discussed in a previous contribution and it is most probably associated to the difference in vapor pressures of Br and Tl [14].

The XRD data for samples deposited using $n=1$–4 are presented in Fig. 3. A strong orientation is observed given the fact that only 100 and 200 diffraction peaks are observed. Other diffraction peaks were observed in Figs. 3(c) and (d), and they might be associated to the variations of growth direction discussed in the SEM of Fig. 2(b). Note that the most intense

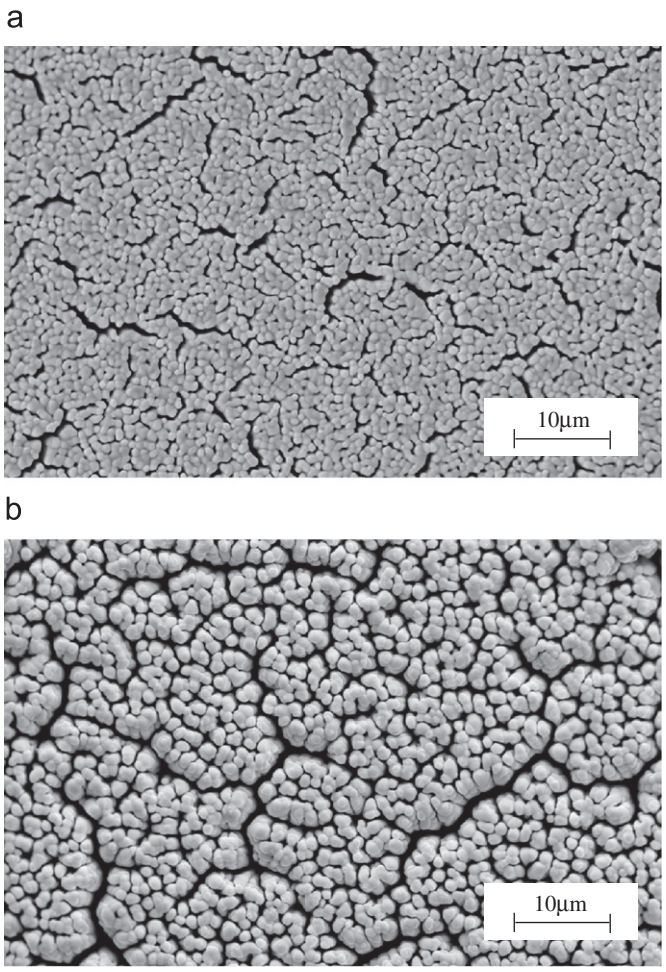

Fig. 1. Scanning electron microscopy images of surface of the samples for (a) one deposition ($n=1$) and (b) four sequential depositions ($n=4$). Each run was performed using an evaporation-boat to substrate distance of 3 cm, and 0.250 g of the starting thallium bromide powder.

peak is 100 for the first two depositions, while the most intense is 200 for the final runs. Besides, after $n=3$ less intense peaks are observed due to 110 and 211 planes. A reasonable limit for the intensity of the diffracted X-ray beam would be about 1% of incoming light. It can be considered that for the diffraction experiment the X-ray path is more than twice the maximum penetration thickness. According to the absorption coefficient of TlBr at this energy range [3], a maximum penetration would be close to 1 μm. Note from Fig. 2(c) that each deposition run results in a more than 10 μm thick layer. Thus, the XRD data discussed above correspond only to the topmost region of each sample, without any information from the underlying previous layers.

A deeper analysis of the XRD data can be performed by calculating the ratio of the integrated area of the 100 peak to the integrated area of the total spectrum, i.e. A_{100}/A_{Total}. This is presented in Fig. 4. It should be noted that for the first two runs no variation was observed and after the third run a decrease of about 45% was measured. This shows that the structure of the material is less crystalline with the contribution of grains oriented along other directions. The ratio of the integrated area of the 100 peak to the integrated area of the 200 peak, i.e. A_{100}/A_{200}, is also presented in Fig. 4. A similar behavior can be seen, which is probably related to the variation of grain orientation and possible internal stresses.

The optical gap as a function of the number of depositions is presented in Fig. 5. The values of the optical gap for the present samples are larger than previously reported in the literature of