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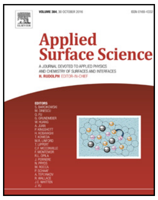

Full Length Article

Transparent thin films of indium tin oxide: Morphology–optical investigations, inter dependence analyzes

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ABSTRACT

Using a fast and eco-friendly deposition method, ITO thin films with different thicknesses (0.5 μm–0.7 μm) were deposited on glass substrates by radio frequency magnetron sputtering technique. A comparative analysis of these oxide films was then carried out. AFM investigations showed that the deposited films were smooth, uniform and having a surface roughness smaller than 10 nm. X-ray diffraction investigations showed that all samples were polycrystalline and the grain sizes of the films, corresponding to (222) cubic reflection, were found to increase with the increasing film thickness. The optical properties, evaluated by UV-VIS-NIR (190–3000 nm) spectrophotometer, evidenced that the obtained thin films were highly transparent, with a transmission coefficient between 90 and 96%, depending on the film thickness.

Various methods (Swanepoel and Drude) were employed to appreciate the optimal behaviour of transparent oxide films, in determining the dielectric optical parameters and refractive index dispersion for ITO films exhibiting interference patterns in the optical transmission spectra. The electrical conductivity also increased as the film thickness increased.

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1. Introduction

Indium tin oxide (ITO) have been widely studied in the last decades by many prestigious research groups, mainly because of the broad spectrum of applicability of these thin films. However, the optical and electrical properties of these thin films depend heavily on the influence of oxygen pressure correlated with thickness of the films, the preparation method and the growth parameters (like deposition rate or temperature).

The choosing a suitable transparent conductive oxide (TCO) may contribute to the advancement of research in photovoltaics [1,2] and optoelectronic devices applications such as light emitting diodes, transparent coatings for solar energy, photo transistors, lasers or flat-panel displays [3–5]. ITO is one of the most appropriate TCO candidate for such applications [6,7] being the most used TCO material due to its excellent structural, electrical and optical properties (low resistivity and high transmissivity) [3,8,9]. In addition, it may be obtained on large surfaces. These are the main reasons, it has been used as thin layer in different devices such as transparent conductors [10] or solar cells [11], where the performance is always influenced by the size and morphology.

As part of optoelectronic devices, it is involved in the control of transmission [8] and electrical conduction [8,9]. The quality of the oxide thin films is highly influenced by the deposition technique [3,7,12,13,14]. Radio frequency magnetron sputtering (rfMS) [15,16], diode (DC) magnetron sputtering [17] reactive ion beam sputtering [18], spray pyrolysis method [19], sol gel process [20] and pyrosol method [9] are among the methods used for deposition of ITO thin films. The rfMS deposition method [12,15,16] used to obtain ITO thin films with very good qualities, has several advantages as compared to other techniques [13,14,16,17]. For instance, it allows the deposition of uniform films on large areas with a good adherence to the substrate and low residual tensions. Variations of deposition parameters lead to structural changes [3] and development of physical processes in these oxide films. ITO is an ideal material as a contact electrode in $CuInGaSe_2$ (CIGS) solar cells [11].

The topic of this work is interesting as a contribution to the development of new optoelectronic devices. Changing the optical and electrical properties by tuning the coating process is an important field of investigation.

In this paper, we report a comparative analysis of ITO thin films obtained by rfMS with different thickness. To track changes in the performance of structural and optical properties of oxide

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thin films, we varied the thickness of the layers. In this study, different approaches were used to measure the optical parameters of transparent oxide films with different thicknesses. Estimating the thickness and transmission of ITO thin films from Swanepoel [21] and Drude methods [22,23], implied determinations of several parameters and investigation of the optical properties. Refractive index dispersion, absorption spectrum, the real and the imaginary parts of dielectric constants, optical conductivity and mobility are some of the important characteristics that could be determined by using these valuable methods. Calculation of plasma frequency is also possible using the Drude method [22–24].

In this paper, we have shown that quality of the obtained transparent oxide thin films, improves with increasing thickness of the deposited layer. This way, the increase in thickness determined an increase of the electrical resistivity at room temperature, for the ITO films (i.e. 7.1 × 10 − 4 Ωcm for ITO 05, 5.5 × 10 − 4 Ωcm for ITO 06 and 4.6 × 10 − 4 Ωcm for ITO 07).

2. Experimental

A solid strategy (regarding experimental, theoretical and applied research) to obtain efficient transparent conductive films using the rfMS deposition technique, is developed in this paper. In this respect, the ITO thin films were deposited by rfMS method [15,25,26], at room temperature onto MENZEL-GLASER optical glass substrates with dimensions of 2 cm × 2 cm × 0.6 cm. The substrates were cleaned ultrasonically in a chromic acid and distilled water bath. Deposition was performed using a VARIAN ER 3119 equipment, with the following main components: two magnetrons, a variomatch, a HV-8Cryo pump and a TM-300 thickness monitor with 6 MHz gold crystal (ScoTech). Deposition rate and thickness of thin films were measured during deposition, after introducing the information on density and acoustic impedance of the material to be deposited, as well as the correction factor related to the target-substrate distance. Commercial individual sputtering targets (from MaTeck) were employed with 99.99% purity (In_2O_3/SnO_2, in a 90/10 wt% ratio), 100 mm in diameter and located at about 90 mm below the substrate holder. The deposition chamber has a volume of 0.2 m^3 and the power of the magnetron is 1 kW. In our experiments, a mixture of high purity (99.995%) gases (Ar flow rate of 30 sccm and O_2 flow rate of 1.5 sccm), was used. The deposition chamber was evacuated at 7.5×10^{-4} Pa before deposition whereas the pressure during deposition was maintained around 4.5×10^{-2} Pa.

The following deposition parameters were used in preparing the ITO samples on substrates at room temperature: rf power applied to magnetron (90 W), electrical current applied to the target (0.2 A), preset thickness of the layer to be deposited (0.5 μm (ITO 05), 0.6 μm (ITO 06) and 0.7 μm (ITO 07), respectively). In the conditions mentioned above, and a deposition rate of about 2.5 Å/s, ITO thin films were obtained. Deposition rate and thickness of thin films were measured during deposition, after introducing the information on density and acoustic impedance of the material to be deposited, as well as the tooling factor related to the target-substrate distance. These deposition parameters, ensured the achievement of uniform ITO thin films of different thickness as a function of deposition time.

Following, white light interferometry principles were employed, by using an Ambios interferential microscope, to measure the thickness of the obtained samples. The investigations on morphological properties of ITO samples were realized by atomic force microscopy (AFM) using an XE-100Park Systems microscope. The AFM images were acquired in standard tight loop, contact mode (height mode) and with a scan speed was 2 μm/s.

The crystalline structure of the ITO films was investigated by X-ray diffraction (XRD) standard technique, using $Cu_{K\alpha}$ radiation ($\lambda = 1.5418$ Å) in the range of $2\theta = 15$–80 degrees of arc. The XRD measurements were performed using a Brucker D8 Advance diffractometer.

The transmission spectra of the obtained oxide thin films in UV-VIS-NIR range was obtained using a double beam Perkin-Elmer Lambda 950 spectrophotometer. The spectra were recorded at room temperature in the 190 nm–3000 nm wavelengths range. The electrical properties were determined by using the four points methods through the medium of a set-up containing a Keithley 6517A electrometer and a RLC bridge. An electrical analysis of the conduction mechanisms specific for different voltage ranges was also performed.

3. Results and discussion

AFM analysis was used to investigate the microstructure and morphology of the oxide thin films. Fig. 1(a)–(c) shows two-dimensional (5 μm × 5 μm) AFM images of the oxide thin films. The roughness (root mean square) is estimated from the topographic images between 2.9 nm and 9.5 nm. Following these scans, the presence of nanoparticles (Table 1) was noticed. It was also found that nanoparticle size increases as the thin film thickness increases, from about 138 nm (for ITO 05 sample), 154 nm (for ITO 06 sample), to about 168 nm (for ITO 07 sample). The same behavior, was observed, with increasing of film thickness in the case of pores dimension between nanoparticles. The surface line profiles for the above-mentioned samples show the presence of gaps (pores) between nanoparticles, specific for the nanostructured morphology. The gaps have sizes of hundreds of nanometers and depths in range of tens of nanometers for thicknesses above 500 nm (Fig. 1(a) AFM line profile). The average sizes between nanoparticles of 132 nm for ITO 05, 156 nm for ITO 06 and 171 nm for ITO 07, were determined. This proves that the nanoparticles are dense and uniformly distributed at the film surface. No cracks or pores with depths close to the films thickness were observed. This is due to aggregation of grains followed by increasing in crystallinity.

The performed investigations show that deposition conditions have a major influence on the roughness, for a given thin film. In addition to the effect on the crystalline structure of thin films, the radio frequency process has an important role on nano-structuring the surface. Thus, a change in morphology was noticed with increasing of thickness.

For thicker layers, the sputtering time was extended, leading to an increase in roughness. This is the case for the thickest sample (0.7 μm), which has the higher roughness value ($R_{rms} = 9.5$ nm) of all samples. Generally, smooth and uniform thin films with roughness values less than 10 nm (on small areas) were obtained for all deposited samples. In these conditions, scattering on the surface was slightly improved, leading to a slight drop of optical transparency in the visible range.

The high intensity sharp peaks in X-ray diffraction (XRD) pattern (Fig. 2) indicate that all ITO thin films samples, with various thickness, have very good crystallization. Positions of all the reflection peaks can be indexed as the characteristic structure of cubic ITO phase (JCPDS File No. 6 416). Lattice constant for the cubic structure was calculated with the following equation [27,28]:

$$\frac{1}{d_{hkl}} = \frac{h^2 + k^2 + l^2}{a^2}, \tag{1}$$

where d_{hkl} is the interplanar spacing and h, k, l are the Miller indices.

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