Preparation and the study optical and electrical properties of thin films for optoelectronic applications

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Abstract
Conductive transparent In$_2$O$_3$ thin films with (222) preferred orientation were prepared by thermal oxidation (TO) in static air of indium thin films at condition \((250^\circ\text{C}/25 \text{ min})\). Detailed structural, electrical, and optical characteristics of the film are presented. The data are interpreted to give a direct band gap of \((3.6) \text{ eV}\) and indirect band gap of \((2.5) \text{ eV}\). The In$_2$O$_3$ film has sheet resistance as low as \((20) \Omega/\square\) in absence of any post-deposition annealing conditions. The mobility of these films was estimated to be \((31) \text{ cm}^2. \text{V}^{-1}. \text{s}^{-1}\).

Keywords: thermal oxidation, In$_2$O$_3$ thin film, figure of merit.

Introduction
Indium oxide In$_2$O$_3$ film is a very important transparent conducting oxide (TCO) material used for optoelectronic applications such as photovoltaic, flat panel display, and heat reflective coating [13]. It exhibits high transparency (> 70%) in the visible region. Several deposition techniques have been employed for deposition of polycrystalline indium oxide films, such as d.c magnetron sputtering [4], reactive ion plating [5], sol-gel technique [6], reactive evaporation [7], metalorganic CVD [8], ultrasonic spray CVD [9], chemical spray pyrolysis [10], and laser ablation [11]. In the majority of the published studies, In$_2$O$_3$ films have been prepared by reactive sputtering and evaporation, since these two techniques gave the best results. Thermal oxidation has been recently used for preparation some semiconducting oxide materials. Cu$_2$O and Ag$_2$O films for ultra large-scale integration applications have been prepared by TO technique using oven at low temperatures [12,13]. Here we report the use of thermal oxidation technique to grow highly oriented In$_2$O$_3$ film and the structural, the optical, and the electrical properties was investigated in present work.

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2. Experiment
2.1 Film Preparation

High purity (99.999%) indium powder was deposited on an optically flat well-cleaned corning glass substrate at room temperature by using thermal resistive technique under vacuum pressure down \((10^{-6})\)torr. After film deposition, the substrate was loaded into TO system. Schematic diagram of the TO system is presented as Fig. (1) [14]. The oxidation process was occurred using tube oven under static air at condition \((250^\circ\text{C}/25\text{min})\). Fig. (2) Display the temperature-time curve for TO process. It is clear that this curve contains three distinct region heating, oxidation cycle and cooling on the other hand, cooling rate of \(25^\circ\text{C}/\text{min}\) calculated from coolie's cycle after oxidation process. The \(\text{In}_2\text{O}_3\) film thickness was measured and found to be 100 nm.

2.2 Film Characteristics

The crystal Structure of \(\text{In}_2\text{O}_3\) film was investigated using XRD analysis \((\text{Cu } K_\alpha \text{ source; } \lambda = 1.54506 \text{ Å})\) with scan speed of 3°/min with 20 mode. The transmittance measurement was performed using spectrophotometer (Shimadzu type) in the spectral range 300-900nm. To measure the electrical properties, ohmic contacts have been made on \(\text{In}_2\text{O}_3\)film by depositing of high purity Al film of 500 nm thick through special thin metal sheet mask. Seebeck coefficient measurement was done by measuring of thermovoltage developed under a temperature difference in the interval \((5)\) K. The dark electrical resistivity was measured at room temperature.

2. Results and Discussion

Fig. (3) Show a photograph \(\text{In}_2\text{O}_3\)film deposited on glass substrate, it is uniform, transparent (around 80% in the visible and NIR regions) and smooth film. The film has good adhesive properties.

The XRD spectrum of \(\text{In}_2\text{O}_3\)film is presented in Fig. (4). There is strong and sharp single diffraction peak at \(20 = (30.6)\). It can be indexed to the (222) crystal plane of bcc structure with \(a = (10.104)\) Å, which is very close to the ASTM Powder Diffraction Data Card No. ASTM-6-0416. Moreover, no metallic indium phases are present in the spectrum. The highly oriented film is so important in order to reduce the effect of film texturing on transport properties. The crystal quality of \(\text{In}_2\text{O}_3\)film prepared by this method is much better than that film prepared by spray pyrolysis, laser ablation, and DC magnetron sputtering techniques, no highly-oriented plane was a direct and indirect optical energy gaps in the allowed transition were obtained from optical absorption measurements as shown in Fig. (5) and they found to be \((3.62)\) eV and \((2.68)\) eV consequently. These results in good agreement with reported results [1].

The room temperature electrical resistivity of the \(\text{In}_2\text{O}_3\)film was found to be \((2\times10^4)\Omega\cdot\text{cm}\). The relationship between electrical resistivity and optical transmittance \((T)\) was correlated and derived by Haacke [15]. This relationship defined by Figure of Merit \((\Phi_{Tc})\) which can be given by:
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\[ \Phi_T = \frac{T^{10}}{R_s} \quad (1) \]

Where \( R_s \) is the electrical sheet resistance measured in thin film.

This parameter could be used efficiently to describe TCO materials. It is obvious from the table that our results are encouraged and competitive to the results obtained by Adurodija et al. [16].

Thermoelectric measurement shows that the In\(_2\)O\(_3\) film has n-type conductivity. Fig. (6) shows the thermovoltage of In\(_2\)O\(_3\) film as a function of the temperature. Seebeck coefficient \( S \) can be given by equation [19]:

\[ \frac{1}{s} = -13.62E_{fo}/A(\gamma - 1/T) \quad (2) \]

Where \( E_{fo} \) is fermi energy. \( A \) is a parameter related to the carrier energy. and \( \gamma \) is constant. We plotting \( 1/s \) versus \( 1/T \) as shown in Fig. (7). From slope and intercept of this figure, we can obtain the values for the parameters \( E_{fo}/A \) and \( \gamma \). The experimental result, show that \( E_{fo}/A \) of In\(_2\)O\(_3\) was around \((0.11)\)eV and \( t = (3.3)K^{-1} \). These values give an indication that the In\(_2\)O\(_3\) film is degenerate semiconductor.

4. Conclusions

We have prepared and characterized of highly oriented In\(_2\)O\(_3\) thin films with aid of thermal oxidation. Figure of merit of these films after comparing with those prepared by other methods suggests the feasibility of producing good quality In\(_2\)O\(_3\) window layer for solar cells and transparent electrodes by simple, reliable and inexpensive TO technique.
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Fig. (1): Furnace

Fig. (2): Temperature time curve for TO process

Fig. (3): A photograph of an In$_2$O$_3$ film prepared by thermal oxidation

Fig. (4): XRD pattern of In$_2$O$_3$ thin film
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Fig. (5): Optical bandgap of In$_2$O$_3$ thin film (a) direct gap (b) indirect gap

Fig. (6): Thermovoltage as a function of temperature.

Fig. (7): Reciprocal of seebeck coefficient vs 1/T.