Optical and electrical properties of Ti-doped In₂O₃ film deposited on flexible transparent substrates by ion beam assisted deposition under different oxygen flow rate

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Optical and electrical properties of Ti-doped In₂O₃ film deposited on flexible transparent substrates by ion beam assisted deposition under different oxygen flow rate

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Abstract

A flexible transparent is an important technology to improve flexible electronic and flexible display devices. Actually, the deposition process of films coated on a flexible substrate will no more than 200 °C or room temperature. In order to achieve high transmission and lower resistivity, this paper reported the thin films of ITiO deposited by ion beam-assisted electron beam under the condition of different oxygen flow rates at room temperature. The electrical, optical, and morphological properties of ITiO films were investigated under the condition of different oxygen flow rates. At the 30 sccm oxygen flow rate, the surface roughness was 5.4 nm, high transmittance and optical bandgap of ITiO films were 89.2% at 470 nm wavelengths and 3.28 eV, respectively. The lowest resistivity of film was $2.1 \times 10^{-4} \Omega\text{-cm}$ at 30 sccm oxygen flow rate. Furthermore, the carrier concentration and hall mobility were experimentally investigated, which presented by $42.8 \text{ cm}^2/\text{Vsec}$ and $9.23 \times 10^{20} \text{ cm}^{-3}$. The use ITiO film coated on a flexible substrate with ion beam-assisted electron beam evaporation at room temperature can improve the

lower resistivity and high transmission of the flexible device. It was also demonstrated that ITiO film with ion beam-assisted deposition technique is suitable for flexible electronic and flexible display devices.

Keyword : titanium doped indium oxide, oxygen flow rate, ion beam-assisted deposition, flexible transparent substrates

1. Introduction

The most common flexible display and flexible electronics devices are basically developed by transparent conducting oxide (TCO). The oxide materials are useful in allowing the device to coat on polyethylene terephthalate (PET) substrate, which can be bent into various shapes such as indium tin oxide (ITO), indium zinc oxide (IZO), and aluminum indium zinc oxide (AIZO)¹⁻³. These materials oxides are semiconductors, which has a wide band gap, there are also other advantages including good insulators, transparent electronics, and photonics. These oxide materials have been widely used in many flexible devices, for instance, flexible solar cells, electroluminescence (EL), thin film transistors (TFT) and others⁴⁻⁶.

Indium oxide or Indium (III) oxide (In_2O_3) is the one of the oxide materials that is used as a precursor to doping agents due to its several advantages like does not oxidation with water, it is heat resistant when mixed with non-metals and metals, good electrical conductivity, low resistivity, high visible light transmission, UV absorption reflects more infrared reflections, wear resistant, and chemical corrosion^{7,8}. However, there are some disadvantages such as high-cost, poor electrical conductivity and also can create problem like toxic. To address these problems, it can be improved by adding titanium dioxide (TiO_2) doped indium oxide (ITiO). Because, it offers more effective like low resistance sheet properties. In addition, the transmission of ITiO film is near-infrared. The properties of the ITiO layer show that the wavelength is 1500 nm when compared to 1000 nm for a resistance sheet of 8 ohm/sq⁹⁻¹².

According to previous studies, the direct current (DC) and radio frequency (RF) magnetron sputtering were particularly used to be the coating process of ITO. In the coating process, oxygen must be controlled because ineffective control has directly affected to the coating layer, may not adhere well. Especially, the controlled pressure is poorly controlled,

which has a negative effect to the adhesion-related on surface. For solving the problems, the ion beam-assisted process is use, which can be controlled the oxygen flow rate resulting in hardness and the coefficient of friction accumulates is decreased. Consequently, it has enhanced adhesion when using ion beam-assisted techniques^{13,14}. For this reason, we are focusing on the ion beam-assisted technique for ITiO film-coated PET substrates, which could be enhanced properties of transmittance and lower resistivity.

This paper proposes a titanium doped indium oxide film coated on the surface of PET by ion beam-assisted techniques at room temperature. The structural, electrical, and optical properties of deposited ITiO films on PET substrate by ion beam-assisted deposition at room temperature on different oxygen flow rate conditions were investigated. The conditions of ITiO films by ion beam-assisted process are significant to performance flexible transparent substrates.

2. Experimental Setup

As previous studied, the preparation of ion beam-assisted deposition process was reported^{15,16}. The resistive sources were conducted by turbo-pumped coating chamber at 6-pocket 270°. The electron beam gun was controlled under 14 kW with a quartz crystal rate/thickness. The ion source by Kaufman & Robinson, Inc. was placed in coating chamber, as seen in Fig. 1. The evaporation source material of ion beam-assisted deposition system was used 10 % titanium dioxide (TiO_2 ; Sigma-Aldrich) with 99.99% of purity and 90% indium oxide (In_2O_3 ; Sigma-Aldrich) with 99.99% of purity. The pressure of evaporation was $\sim 4 \times 10^{-3}$ mtorr at room temperature. The quartz crystal monitor was monitored for evaporation rate and thickness of

film. In during the deposition process, ion energy and ion current density was controlled at 160 eV and $100 \mu\text{A}/\text{cm}^2$, respectively.

The PET substrates were cleaned by ultrasonic washer with acetone at 45 min, alcohol at 45 min, the organic compounds was removed by de-ionized water, and after that was blow dried with nitrogen gas. The obtained PET substrates were coated with ITiO by ion beam-assisted on the condition of oxygen flow rate at 10, 20 and 30 sccm. The flow rate was determined by a mass flow controller (MKS). The electrical and optical properties of ITiO film were measured by a sheet resistance meter (Dasol Eng; FPP-HS8), hall-effect measurements (ECOPIA HMS-5000) and UV spectrophotometer (Hitachi; U-3900) the structure of film was investigated by atomic force microscopy (AFM) (WITec GmbH; alpha300 RA) and X-ray diffractometer (XRD) (Rigaku; D/max 2100H) under Cu Ka irradiation ($\lambda = 1.5418\text{\AA}$)

3. Result and Discussion

The structure, electrical and optical properties of ITiO films deposited on flexible (PET) substrate by ion beam-assisted deposition were controlled with an oxygen flow rate range of 10, 20, and 30 sccm. The deposition rate in all films was deposited at 200 nm of thickness, 0.5 nm/sec rate of deposition, 160 eV of ion energy and $100 \mu\text{A}/\text{cm}^2$ of ion current density.

Fig. 2 shows the X-ray diffraction (XRD) pattern of ITiO films deposited on flexible (PET) substrate by ion beam-assisted deposition. As can be seen in Fig. 2, the strong peak of all films has corresponded In_2O_3 (222) plane at 35.05° (2θ) and Ti (101) plane at 25.62° (2θ). All peaks of (400), (440), and (622) planes were found in agreement with In_2O_3 and TiO_2 planes were not found as extra peaks. However, TiO_2 doped indium oxide resulted in slow cumulative

growth of grain size in indium. From previous reports ¹⁷⁻²⁴, the amount of ion energy and ion current density was obtained from the positive ions colliding to target will increase with increasing oxygen flow rate. The energy of ions was dependent on the oxygen flow rate, electrons are released from the replacement of In_2O_3 atoms cause charged oxygen vacancies. Oxygen vacancies can increase the double charge of oxygen. The existence of TiO_2 will result in n doping of the lattice due to the doped materials that would be increased electrons to the conduction band ²⁵⁻²⁷.

In order to confirm the effect of oxygen flow rates on the structural properties of the ITiO films by ion beam-assisted deposition, the AFM of the ITiO films deposited at different oxygen flow rates at 10 to 30 sccm were investigated. Fig. 3 shows the AFM of the ITiO films deposited on flexible (PET) substrate by ion beam-assisted deposition. As the oxygen flow rates increase up to 30 sccm, the crystallite size of the ITiO films at 10 sccm of oxygen flow rate was less than that 30 sccm of oxygen flow rate. Increasing the oxygen flow rate excites crystal growth and induces film crystallinity improvement. Moreover, AFM of the films has measured the roughness of the films, it depended on the fabrication parameters of the films. Normally, they use root-mean-square (RMS) evaluate to the roughness of the films. The root means square (RMS) uses the same classifying surfaces, RMS can be defined as the standard deviation of the surface height ²⁸. The root mean square (RMS) surface roughness of the ITiO films in all oxygen flow rates conditions was lower to 5.4 nm of RMS surface roughness. Increasing oxygen flow rates result in the mobility of the atom decrease, thereby providing rough thin films.

The detailed investigation of the transmittance (%) of ITiO films deposited on flexible (PET) using ion beam assisted deposition was specially controlled with different oxygen flow rates, which was investigated by the UV-Visible spectrophotometer under the wavelength range

300 to 800 nm, as shown in Fig. 4. Obviously, the transmittance of the ITiO films in all oxygen flow rate conditions was shown by 84% over in the visible region. There have been found that the maximum transmittance value of ITiO films was presented by 89.2% under the oxygen flow rate of 30 seem with the wavelength of 470 nm. Therefore, it can be summarized that the wavelengths of 450 to 500 nm are suitable for applying with a solar cell, comparing in all oxygen rate conditions. Note that because solar energy has a high intensity in the absorbance wavelength 450-550 nm, which has a wavelength close to the film obtained ²⁹. In addition, when the oxygen flow rate is steadily increased, the absorption coefficient (α) of the film would be shifted onto the ultraviolet (UV) (<350 nm), which can be calculated by Lambert's formula ^{11,12,16,30}. In this point, it affects to the optical band gap values of the films.

$$\alpha = \frac{1}{d} \ln \left(\frac{1}{T} \right) \quad (1)$$

where T and d are transmittance and film thickness, respectively. In order to move to the conduction band, the energy level was defined by appropriate electron excitation and move to the conduction band. At low energy, the absorption can be grown with optical energy in the absorption edge of the semiconductors. The absorption coefficient is directly allowed to transit through a simple parabolic scheme form, which can be found a function of incident photon energy as follows:

$$\alpha h\nu = A(h\nu - E_g)^{1/2} \quad (2)$$

where $h\nu$ is photon energy and E_g is the optical band-gap.

The optical band-gap of ITiO films can be determined by means of extrapolation from $(\alpha h\nu)^2$ versus $h\nu$. As a continuously increased of oxygen flow rate, the optical energy band gap

of 3.23 eV to 3.28 eV were found, as seen Fig. 5. Thus, the increasing of oxygen flow rate results the significant resistivity reduction and the visible range of films is fairly good transparency. The energy bandgap of ITiO films gives more energy to make a low wavelength range of the incident light. Note that the light transmittance of ITiO film is flowed through to the PET substrate under the condition of different wavelengths, where the titanium dioxide has an energy level larger than 3.2 eV³¹⁻³².

The oxygen flow rates of ITiO films have an evident effect to the light transmittance range of films in the UV range. As can be seen in Fig. 6, the resistivity and sheet resistance of ITiO films deposited at 160 eV of ion density under different conditions of oxygen flow rates was presented. Looking in more detail, there have been found that the lowest resistivity of the ITiO films was illustrated under the condition 30 sccm of oxygen flow rate, which was presented by $2.1 \times 10^{-4} \Omega\text{-cm}$. In addition to this point, the resistivity at 10 sccm and 20 sccm of oxygen flow rates were nearly constant at $2.7 \times 10^{-4} \Omega\text{-cm}$ and $2.6 \times 10^{-4} \Omega\text{-cm}$, respectively.

It is important to note that the decreased of resistivity is the result of rising in the oxygen flow rates because the deposition process has a sufficient oxidation of ITiO films, which may cause in the oxygen absorption in the grain boundaries. The sheet resistances of the ITiO films density under different oxygen flow rates were found that 34.1 Ω/sq , 32.6 Ω/sq , and 28.6 Ω/sq , respectively. It demonstrates that a minimum at 30 sccm oxygen flow rate.

The increasing oxygen flow rate causes oxygen-deficient ITiO films. The increasing oxygen incorporation causes the oxygen-deficient of the film thus sheet resistance of ITiO film depends on oxygen vacancy into evaporated films¹⁶. At 30 sccm oxygen flow rate, the resistivity and sheet resistance of films presented the lowest value because the oxygen was etched on

surface area, which cause surface modification by ion beam-assisted process, as can be seen in Fig. 3. Moreover, it shows a jagged hole and a groove on the surface is decrease. The lower oxygen flow rates have importantly affect to the ITiO films, which cause to the surface have a free space and electron concentration will be increased^{33,34}.

The hall mobility and carrier concentration and of ITiO thin films by deposited on flexible substrate by ion beam-assisted deposition under different oxygen flow rates, as seen in Fig. 7. The hall mobility was slightly enhanced 36.2 - 42.8 cm²/Vsec with increasing oxygen flow rates from 10 to 30 sccm. As can be seen, the highest mobility was located by 30 sccm of oxygen flow rate about 42.8 cm²/Vsec. At low resistivity and sheet resistance of films result a high hall mobility because it attributed to a formation of In₂O₃ in the films. Thus, the electrons or ions are released from the replacing to indium (In) atom in the sublattice, and from oxygen vacancies with double charge^{26,35}. The carrier concentration of ITiO film under different oxygen flow rate has reached the peak of 1.12×10^{21} cm⁻³ under oxygen flow rate 10 sccm and reduced to 9.23×10^{20} cm⁻³ under oxygen flow rate 30 sccm. When, the rising oxygen flow rate result increase carrier concentration. The on energy and ion density will cause kinetic energy move to the condition band^{36,37}. Moreover, the relationship between electrical resistivity and carrier concentration could be explained by the morphological properties of the film with AFM morphology in Fig. 3. It was confirmed that the rising oxygen flow rate could not grow crystalline grains that was observed from the porosity on the surface. The increasing oxygen flow rate results in a decrease grain size, thereby reducing resistivity, increased mobility, and increased concentration.

4. Conclusions

The effect of ITiO films coated on the surface of PET by ion beam-assisted techniques at room temperature under different oxygen flow rates. During the deposition process, 160 eV of ion energy, 100 $\mu\text{A}/\text{cm}^2$ of ion density, 100 nm of thickness, and 0.5 nm/sec of deposition rates were experimentally used. In an experiment, the lowest resistivity of ITiO film was $2.1 \times 10^{-4} \Omega\text{-cm}$ at 30 sccm of oxygen flow rate. The optical transmittance and optical band-gap of ITiO film were 89.2% (470 nm) and 3.28 eV under 30 sccm of oxygen flow rate. The hall mobility and carrier concentration of ITiO film were $42.8 \text{ cm}^2/\text{Vsec}$ and $9.23 \times 10^{20} \text{ cm}^{-3}$ at 30 sccm of oxygen flow rate, respectively. According to these results, under oxygen flow rates at ion energy and ion density constant are attractive methods to produce a high-quality ITiO film which is a suitable 30 sccm oxygen flow rate. The use ion beam-assisted techniques under different oxygen flow rates can improve the crystalline and electrical properties of ITiO film on PET substrate.

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Figure and Table

Fig. 1 Schematic diagram of ion beam-assisted deposition system.

Fig. 2 X-ray diffraction (XRD) pattern of ITiO films deposited on flexible (PET) substrate by ion beam-assisted deposition at 10, 20 and 30 sccm oxygen flow rates.

Fig. 3 AFM of ITiO films deposited on flexible (PET) substrate by ion beam-assisted deposition at 10, 20 and 30 sccm oxygen flow rates.

Fig. 4 Transmittance spectra of ITiO films by deposited on flexible (PET) substrate by ion beam-assisted deposition under different oxygen flow rates at 10, 20 and 30 sccm.

Fig. 5 Energy bandgap of ITiO thin films by deposited on flexible (PET) substrate by ion beam-assisted deposition under different oxygen flow rates at 10, 20 and 30 sccm.

Fig. 6 Resistivity and sheet resistance of ITiO thin films by deposited on flexible (PET) substrate by ion beam-assisted deposition under different oxygen flow rates at 10, 20 and 30 sccm.

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