Research Article

High Performance of IZO Coated on PET Substrate for Electroluminescence Device Using Oxygen Plasma Treatment

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Received 27 August 2020; Revised 6 March 2021; Accepted 31 May 2021; Published 12 June 2021

Academic Editor: Simona Binetti

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Thin films of indium zinc oxide (IZO) were deposited on polyethylene terephthalate (PET) substrate with varying plasma power (from 100 W to 300 W) using the radio-frequency (RF) magnetron sputtering technique and electroluminescence (EL) devices. The IZO films that were obtained from this process were treated with oxygen plasma powers using the plasma-enhanced chemical vapor deposition (PECVD) system. After this treatment, the microstructural, electrical, and optical properties of IZO films were observed and reported. The result showed that the IZO/PET films was fabricated at the lowest resistivity ($2.83 \times 10^{-3} \Omega \cdot \text{cm}$), while the optical characterization displayed the maximum transmittance of 95% in the visible region with a smooth morphology and good crystalline structured, affected by the 300 W of plasma power with the optimum carrier concentration ($4.93 \times 10^{21} \text{ cm}^{-3}$) and hall mobility ($42.12 \text{ cm}^2/\text{V} \cdot \text{sec}$), respectively. The luminance properties and the EL efficiency were also investigated and shown a 300 W highest point of plasma power with 84 cd/m^2 and 0.924 lm/W . The film properties were found responsible for producing and improving the performance of IZO/PET substrate, suitable for displaying the devices.

1. Introduction

Flexible display devices such as organic light-emitting electrochemical cell (OLEDs) and light-emitting diodes (LEDs) have become interesting to study because of their displayed characteristics toward lightness, thinness, and wide viewing angles [1, 2]. However, these light-emitting devices are quite difficult to manufacture in some manufacturing processes, for example, vacuum evaporation processes or metal organic chemical vapor deposition (MOCVD). Consequently, new light-emitting materials like electroluminescence (EL) panel have been considered as the best choice method for photoelectronic devices. This is because the light-emitting materials have unique advantages such as less limitation and simple device structure of electrode materials, as well as its ability to fabricate large-area devices [3, 4]. In addition

to the EL properties, the transparent conductive oxide (TCO) materials are widely used as conductive oxide (such as optoelectronic devices, flat panel displays, and solar cell). Most studies on the TCO is concentrated on indium-tin oxide (ITO) [5] which is commonly used for the EL devices because of their good electrical properties, for instance, high electrical conductivity, optical transparency, and thermal stability of 500°C [6–8]. Therefore, the new candidate is necessary for ITO replacement due to the scarcity of in-reserves [9]. The transparent conducting zinc oxide (ZnO) films are commonly used as potential substitutes for ITO. The conductive ZnO has become increasingly interesting because they have a wide band gap of 3.37 eV with a large exciton binding energy of 60 meV semiconductors in the visible wavelength range which can enable the efficient excitonic emission at room temperature [10, 11]. Outside of these

properties, ZnO is a promising material for a variety of electronic devices such as thin films optoelectronic, transparent electronic, sensing, and photovoltaic [12, 13]. However, in comparison with most TCO electrodes, the electrical conductivity is still lower. In order to increase ZnO conductivity, some elements need to be incorporated into ZnO, such as aluminum (Al), gallium (Ga), and indium (In), that have been used experimentally as substitutional dopants for ZnO which can effectively change the electrical and optical properties as well as geometry (size and morphology) of the films. Therefore, gallium-doped zinc oxide (GZO) and aluminum-doped zinc oxide (AZO) are experimentally found to be the interesting option. However, there have been extensively studied of those dopants, which have been completely done by using glass substrates [14–17]. Shi et al. [18] fabricated the conductive AZO thin films of $7.56 \times 10^{-4} \Omega \cdot \text{cm}$ resistivity with the thickness of 380 nm. Kim et al. [19] fabricated AZO thin films with the lowest resistivity of $7.8 \times 10^{-4} \Omega \cdot \text{cm}$ and thickness of 200 nm. Nevertheless, these dopant types can create some problems, such as heavy brittle and unfold able. Among the various element dopants, indium is a unique and interesting material due to its qualities, energy band gap (3.37 eV), high optical transmission, high chemical stability, and high electrical conductivity which has been widely used in many microelectronics (such as solar cells, flat panel displays materials, and sensors) [20–22]. For these reasons, indium-doped zinc oxide (IZO) films seem to be a good material for ITO films [23]. The deposition of IZO films on flexible substrates can be reliable with energy conversion technology like plastic which is flexible, lightweight, and cheaper than glass substrates [24, 25]. Among various plastic materials, polyethylene terephthalate (PET) is a promising alternative for flexible substrates due to its good thermal stability and spin ability, as well as surface inertness which performs the optical transmission of over 85% in the visible range [26, 27]. In order to prepare IZO films, various deposition techniques are considered such as sol-gel processing, ion beam sputtering, magnetron sputtering (MS), and radio frequency (RF) magnetron sputtering. Among these methods, RF magnetron sputtering is an excellent method due to its ability to coat heat-sensitive substrates, high-purity films, dense pore-free coating, and good adhesion (30 MPa) [28, 29]. However, the IZO film seems to be problematic since it is based only on RF magnetron sputtering with surface roughness, easy cracks, and uneven surface or edge [30]. To address these problems, an oxygen plasma treatment process is used in order to fabricate the IZO film surface and make it smoother and without cracks [31]. Alternatively, a collision with cosmic rays at atmospheric pressure is the most adopted technique of plasma production due to the fact that it produces multiple electrons in gas molecules and charges particles through a pulling electron density process. The particle-causing electrons are then positively charged (ionization) [32, 33], showing the gas collision mechanism as follows (Equation (1)):

In addition, oxygen is considered important because this part shows the oxygen mechanism as the ozone precursor by reaction [34] as follows (Equation (2)):

There are many surface treatment methods, laser treatment [35], coupling agent coating [36], and oxygen plasma treatment [37]. Particularly, oxygen plasma treatment appears to be the best choice. Due to the advantages of this method, it has a high efficiency, no damage to substrate, and environmental friendliness [38]. To applicate, there are many researches that have studied the application of IZO films, for example, Ullah Sana et al. developed the properties IZO films by boosting highly transparent and conductive [39], and Goncalves Goncalo et al. studied the improving transparent of IZO films of orange, green, and blue organic light emitting [40, 41]. Consequently, in order to achieve multiple properties of high-quality crystallinity and enhanced electrical and optical properties, the IZO films are considered significant. In this study, we fabricated indium-doped zinc oxide (IZO) films and coated on PET substrate by radio-frequency (RF) magnetron sputtering technique which was later treated with oxygen plasma. We investigated the electrical and optical properties of conductive IZO thin films under different conditions of plasma power from 100 W to 300 W. This process influenced the structural characterization of the films growth.

2. Materials and Methods

2.1. Preparation of IZO Film Deposited on PET by Oxygen Plasma Treatment. With regard to the IZO films deposited on PET production, the IZO films were deposited on PET substrate (MELINEX® 453) by the RF magnetron sputtering technique with $4 \times 4 \text{ cm}^2$. To prepare the ZnO-doped In_2O_3 , the researchers used IZO alloy target (AEM company) that has a purity and density of 99.99% consisted of ZnO (10 wt.%) and In_2O_3 (90 wt.%). The first step in the thin film production process was to clean the substrate ultrasonically (UC-2000) in order to remove contamination before the deposition process. In addition, the distance between the target and the substrate was held at 7 cm, and the vacuum chamber (SUS304) was pumped to a base pressure of 2×10^{-6} Torr. The working pressure was set at 4 mTorr and controlled during the deposition process. The film thickness of 500 nm was grown on the PET substrate at room temperature by changing the RF power deposition.

After the IZO films deposition processes, the films were treated by plasma-enhanced chemical vapor deposition (PECVD) system [40] with a symmetrical position and size reactor in the two parallel plates. In the plasma treatment of IZO films, the PET substrate was placed on the grounded electrode cathode with the RF signal applied to the upper electrode at 13.56 MHz under various RF power conditions from 100 W to 300 W in 1 hour. Moreover, the oxygen gas was pumped to 50 sccm constant and 26.6 Pa of working pressure that was controlled with the substrate temperature on the surface (at about 50°C, as can be seen in Figure 1).

2.2. Preparation of EL Device with IZO Film Deposited on PET Substrate Electrode. The element of IZO PET/Phosphor/insulator (BaTiO_3)/Ag electrode were considered and used for EL device with $3 \times 3 \text{ cm}^2$ size. A thickness from $10 \mu\text{m}$ to $50 \mu\text{m}$ was prepared by phosphor power ($\text{ZnS}:\text{Cu}$, $20 \mu\text{m}$ - $32 \mu\text{m}$) that was purchased from Osram Sivanier Company. As a binder ink system, 1 gram of phosphor and 1.5 gram of binder were mixed by stirring for 10 min at 800 rpm, then dried treatment was applied at 130°C for 25 min. It should be noted that the phosphor was deposited by insulator for two times. Furthermore, the Ag electrode deposited on the insulator with a size of $2.5 \times 2.5 \text{ cm}^2$ was dried at 130°C for 25 min [39, 42, 43]. In this work, the morphology properties of IZO film deposited on PET substrate by oxygen plasma treatment for the EL device were studied by atomic force microscope (AFM: Prak, X-100). The microstructure of the films was analyzed by X-ray diffraction (XRD: JDX-3530) with Cu K- α ($\lambda = 1.5418 \text{ \AA}$) source and a scanning range 20° - 80° of 2θ . The optical properties were investigated using the UV-visible spectrophotometer (PerkinElmer: LAMBDA 950). The electrical properties such as resistivity, carrier concentration, and hall mobility were measured by hall coefficient using van der Pauw measurement [44]. Moreover, the luminance properties of the films were examined through spectral brightness analyzer (Konica Minota; CS-2000). A 5 V 60 Hz voltage was applied by digital function generator (Hantek; HDG2012B).

3. Results and Discussion

3.1. Structure Properties. The investigation of IZO films deposited on PET substrate was prepared by oxygen plasma treatment in this study. In order to make a systematic study, the plasma power from 100 W to 300 W were experimentally controlled and the working pressure as well as the distance between target and substrates were fixed. The surface morphology of the IZO films was studied by the AFM analysis as shown in Figures 2(a)–2(d) where various shapes of columnar microstructures are presented. As presented, the significant point has been found at RF power 300 W which shows a greater characterization of uniform grain distribution and has a diameter of 50 nm resulting in the smooth chemical particles on the surface, and this condition can remove the unrelated chemical particles from the IZO films [45]. Consequently, the results showed that the plasma power is a crucial factor in the film growth. It was observed that at lower plasma power, the IZO films with less density and bigger grain size were obtained, which presented low qualities for application in display technology. On the other hand, when the plasma power is increased, IZO films displayed more consistent surface characteristics in the formation of columnar microstructure showing IZO films surface formation of granular structures with smaller grains size [46–48]. This is because the preparation of IZO films at higher plasma power is an effect on the atoms on high deposition, IZO atoms increasing kinetic energy, and high diffusion coefficient. Thus, the electrons were easily moved through the free space causing high grain size and film density of the IZO films.

FIGURE 1: The preparation of IZO film by oxygen plasma treatment for EL devices.

The thickness of the films was measured by using a surface profilometer (Dektak 150). There has been found that the highest thickness was presented by 650 nm, which is located at plasma power of 300 W. Note that the plasma power has an evident effect on film thickness. The film preparation at higher plasma power displays a thicker film than preparation at lower plasma power. For the reason, ions were stimulated to easily move along with high energy, due to the rising plasma power during growth. Hence, a momentum transfer with collisions of atoms occurred on the film surface, which led to obtained thicker films. Additionally, Figure 3 shows the XRD pattern of IZO films deposited on PET substrate by oxygen plasma treatment. The IZO films shows the formation of the polycrystalline structure which refers to the JCPDS 36-1451 standard. All samples displayed indium (101) diffraction peaks located at around 36.9° (2θ). Each conditions showed the peak at (002), (101), (102), and (004) at approximately 34.5° , 48.3° , and 68.9° , respectively, after further investigation. Figure 3 also showed that the intensity of the XRD peak steadily increases with the rise in the crystallinity of the IZO films on the plasma power. This corresponds to Figure 2 with the highest intensity plasma power at 300 W.

Moreover, the Bragg's equation [49] was used to calculate layer spacing distance d as can be seen in Table 1. The given Table 1 provides information about the grain size that was calculated by the Scherrer's formular [50] (Equation (3)).

$$D = \frac{0.9\lambda}{\beta \cos \theta}. \quad (3)$$

The grain size of the IZO films were a gradual decrease from 16.8 nm to 12.2 nm along with the increasing of plasma power. Note that the grain size seen from AFM is correlated from the grain size determined by XRD analysis.

3.2. Optical and Electrical Properties. Figure 4 illustrates the optical transmittance spectra of the IZO films deposited on PET substrate at different plasma power from 300 to 800 nm wavelength regime. The given line graph shows the average transmittance range at 450 nm to 750 nm. The wider transmittance spectrum for high substrate temperature samples implicated the more absorption efficiency for the thin films. As can be seen, the transmittance of the IZO film with 100 W was nearly 86% within the visible light region and at

FIGURE 2: AFM image for the morphologies of the IZO films deposited on PET substrate by oxygen plasma treatment with the different plasma power for EL device. (a) Untreated. (b) 100 W. (c) 200 W. (d) 300 W.

FIGURE 3: The XRD pattern of the IZO films deposited on PET substrate by oxygen plasma treatment at the plasma power of 300 W, 200 W, 100 W, and untreated.

200 W presented by around 90%. It was also found that the maximum transmittance of 95% was present at the IZO films of plasma power at 300 W. This is because of changing con-

TABLE 1: XRD data of the IZO obtained from study to those of the films deposited by plasma power of 300 W, 200 W, 100 W, and untreated.

Plasma power (W)	2θ (°)	FWHM (°)	d (Å)	Grain size (nm)
Untreated	33.13	0.412	2.4215	16.8
100	31.32	0.568	2.2398	14.9
200	30.65	0.653	2.1569	13.1
300	30.11	0.745	2.1086	12.2

ditions in rising plasma power. This is an essential factor for the plasma to have many argon ions causing the target atoms to lose their energy from the collision with argon ions. For these reasons, atomic energy was insufficiently deposited on the substrate, resulting to a decreased deposition rate. Therefore, the transmittance value was increased due to the reduced thickness of the films. Note that the optical transmittance of the films has little effect on plasma power which is the response of the complex refractive index of the material and the surface roughness [51, 52].

The calculation of the absorption coefficient (α) was used to calculate the optical band gap of IZO films (Equation (4)) [53].

FIGURE 4: The optical transmittance spectra of the IZO films deposited on PET substrate by oxygen plasma treatment at the plasma power of 100 W, 200 W, 300 W, and untreated.

FIGURE 5: The spectrum phosphor of the IZO films deposited on PET substrate by oxygen plasma treatment at the plasma power of 100 W, 200 W, 300 W, and untreated.

$$\alpha = \frac{1}{d} \ln \left(\frac{1}{T} \right), \quad (4)$$

where T is the transmittance and d is the layer thickness. In this point, the optical band gap (E_g) is related to the absorption coefficient (α) via the Tauc relation (Equation (5)) [54].

$$(\alpha h\nu)^2 = A(h\nu - E_g), \quad (5)$$

where A is a constant of the semiconductor and $h\nu$ is the photo energy. Therefore, the calculated values of the optical band gap of IZO films are summarized. As a result, there have been found that the values of E_g was shown in the range between 3.7 eV and 3.95 eV, which is the

FIGURE 6: Electrical resistivity (ρ) of the IZO films deposited on PET substrate by oxygen plasma treatment at the plasma power of 100 W, 200 W, 300 W, and untreated.

FIGURE 7: Carrier concentration (N) and hall mobility (μ) of the IZO films deposited on PET substrate by oxygen plasma treatment at the plasma power of 100 W, 200 W, 300 W, and untreated.

determinant of the position of emission energy in photon energy form. Note that ZnO films typically have an average energy band gap of about 3.37 eV. To compare with IZO films, we found that there was a gradual shift up in energy band gap which led to the increasing of light transmittance range of IZO films in the UV range [55]. Moreover, we found that the plasma power (100 W, 200 W, and 300 W) displayed a blue shift. As seen in Figure 5, the maximum value was shown by the condition of 300 W plasma with an average wavelength of 523 nm, which shows the EL device in the visible range.

Moreover, the electrical properties of the IZO films deposited on PET substrate were investigated by hall coefficient using van der Pauw measurement. A magnetic field was set constant at 0.5 T. Figure 6 presents information on electrical resistivity with the variation of plasma power. With increasing plasma power from 100 W to 300 W, the result shows a dramatic decline from $8.92 \times 10^{-3} \Omega \cdot \text{cm}$ to $4.85 \times 10^{-3} \Omega \cdot \text{cm}$. We found that the lowest point was shown by

TABLE 2: Comparison the electrical properties of doping of ZnO with high-performance ZnO/Ag/ZnO.

Deposition parameters	Substrate	Resistivity	Transmittance (%)	References
IZO	PET	$2.83 \times 10^{-3} \Omega \cdot \text{cm}$	95	This study
ZnO/Ag/ZnO	PET	$8.11 \Omega \cdot \text{sq}^{-1}$	94.8	[57]

approximately $2.83 \times 10^{-3} \Omega \cdot \text{cm}$ at plasma power of 300 W. Furthermore, the dependence carrier concentration and mobility with different plasma power condition is shown in Figure 7. Looking at Figure 7, there has been a steady increase trend in the value of carrier concentration (N) which is shown the highest value by $4.93 \times 10^{21} \text{ cm}^{-3}$ at 300 W. As the same time, the mobility (μ) trend has been steadily raised to $42.12 \text{ cm}^2/\text{V}\cdot\text{sec}$ at 300 W. According to these results, it can be assumed that the hall mobility and the carrier concentration are gone up as the plasma power is increased. The reason for this result is due to the surface behavior modification and oxygen vacancies of plasma power that were etched on the IZO film [49, 51, 52]. Therefore, the obtained IZO film by oxygen plasma treatment at high hall mobility and carrier concentration may be suitable for photochemical and electrochemical applications. An improved IZO films with high carrier concentration and mobility can simultaneously be realized in this research, which is obvious from Figures 6 and 7. As a result, it is obvious that we provided IZO films of thickness 650 nm. According to the above mentioned, we would expect high thin film properties by increasing electrical and optical effective in the films of higher plasma power since these are prepared in a different condition of the plasma power. When plasma power is increasing, the mobility trend is in a steady rise and the carrier concentration have a similar trend. The mobility has a direct effective to the electrical properties. By increasing the plasma power, ions are accelerated by increasing power to move with high energy. As a result, atoms on the surface of films will have a high motion energy, which lead to the electron can easily to move from valence band to conduction band well. For this case, the mobility is increased and also carrier concentration; thus, it is a positive effect to the electrical properties [56].

Table 2 illustrates a comparison result of the films between IZO and ZnO/Ag/ZnO. As can be seen, this study performs an advantage potential, which provides the lowest resistivity than that of the film prepared by others.

3.3. Luminance Properties. The luminance properties of IZO films deposited on PET substrate by oxygen plasma treatment for EL devices were investigated by spectral brightness analyzer with a voltage of 5 V and 60 Hz (as shown in Figure 8). According to Figure 8, there has been a steady increased trend in the luminance values of IZO films. The significant point was shown at 300 W plasma power point which indicates the highest luminance of 84 cd/m^2 , 200 W, 100 W, and untreated plasma power values at 76 cd/m^2 , 64 cd/m^2 , and 52 cd/m^2 , respectively. Note that the maximum luminance was obtained from the EL devices with IZO films coated on PET substrate by oxygen plasma treatment because of the behavior of the voltage waveform and faradaic current. This caused the current in the phosphor between the IZO

FIGURE 8: Luminance and Eff_{el} of the IZO films deposited on PET substrate by oxygen plasma treatment at the plasma power of 100 W, 200 W, 300 W, and untreated.

films surface and treated plasma to charge, an effect of fast electron transfer. Looking in more detail, for the increased plasma, power was a direct effect to the improved luminance because the IZO electrode has many electrons and the film surface area that could have led to a faster transfer of electron, which resulted in electron collision. In this case, it is a major factor in the behavior of faradaic current and voltage waveform which has a positive effect on the IZO film surface [58, 59]. Furthermore, the EL efficiency (Eff_{el}) or called power efficiency was calculated by the luminance of EL devices which was multiplied by the area light emission per output power. The Eff_{el} can be showed in Equation (6) [35, 38].

$$\text{Eff}_{\text{el}} = \frac{I_v \cdot A}{V \cdot I_{\text{out}}}, \quad (6)$$

where I_v is a luminance (cd/m^2 or $\text{lm/m}^2/\text{sr}$). A_{surf} is light emitting surface. It was found that the highest Eff_{el} of IZO films coated on PET substrate was at 0.924 lm/W , 200 W, and 100 W of plasma power 300 W, and untreated at 0.845 lm/W , 0.712 lm/W , and 0.437 lm/W , respectively. At the best, the Eff_{el} of the EL devices with IZO film and oxygen plasma treatment was at 300 W plasma power. This can be explained by the increasing number of electrons into the phosphor and increasing phosphor Zn and Cu electrons' collision between ZnS:Cu phosphor.

4. Conclusions

In this work, we have studied the thin film properties of the IZO films deposited on PET substrate by RF magnetron sputtering method and oxygen plasma treatment which both had a great influence on the structural, electrical and optical properties of films. The difference of plasma power was

significantly used to be the condition in this study, which was experimented under various conditions by plasma power 300 W, 200 W, 100 W, and untreated. The IZO films were treated at O₂ plasma treatment power using the PECVD system. As a result, the IZO films coated on PET substrate of plasma power 300 W were seen to have lower resistivity than other conditions with the lowest point of $2.83 \times 10^{-3} \Omega \cdot \text{cm}$ and the highest luminescence of 84 cd/m². The use oxygen plasma treatment on IZO films for EL devices could have possibly improved the efficiency of the light intensity. In addition, these reasons can also demonstrate that IZO thin films by oxygen plasma treatment samples are feasible in the fabrication of the IZO thin films that are suitable for TCOs application and electronic display devices.

Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

Conflicts of Interest

The authors declare that there is no conflict of interest regarding the publication of this paper.

Acknowledgments

This work was supported by the Petchra Pra Jom Klao Master's Degree Scholarship from King Mongkut's University of Technology Thonburi (KMUTT), Thailand (grant number 11, 2019); under the project of the Research, Innovation, and Partnerships Office (RIPO); and National Research University Project of Thailand's Office of the Higher Education.

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