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Preparation of ZnO Based Thin Films for OLED Anode by Facing Targets Sputtering System

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In this study, we prepared ZnO-based thin films-AZO, GZO and GAZO for OLED anode by facing targets sputtering (FTS) system. The reason of the used ZnO-based thin films is that it can be prepared to obtain high transparency in the visible range, low resistivity, stability of chemical and stability in hydrogen plasma by added materials such as Al, Ga. The electrical and optical properties of the as-doped AZO, GZO and GAZO thin films were evaluated with UV/VIS spectrometer, 4-point probe, XRD, AFM, FESEM.

And the performance of the OLED device such as the operating voltage and its efficiency were evaluated by J-V-L (current density-voltage-luminance) measurements

Keywords AZO; GAZO; GZO; OLED

Introduction

Recently, Organic light Emitting device (OLED) is receiving attention as a next-generation display because it has emissive bright colors, wide viewing angle and fast response time. While existing display is required a complex process and circuit, OLED has a simple process and structure so which can be applied various field. For these reason, to improve the efficiency of OLED, various studies are being done [1,2]. Such as efficient injection of electrons, optimized surface property and electrode is an example. Among the various studies, transparent electrode is a fundamental part of the drive device, and study of materials and structures have been reported. Now, among the TCO materials, Indium Tin Oxide (ITO) have reputation for high transmittance and low sheet resistance in OLED [3]. On the other hand ITO has weakness as like pinhole and dark spot caused by polycrystalline structure and rough surface. Also, raw material price is highly and unstable in H₂ plasma and toxic. Therefore, it is required electrode materials that can be replaced with ITO [4]. ZnO-based thin film is stable in H₂ plasma atmosphere, and has a high transmittance

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and it can be processed in low temperature so it is receiving attention as a material which can be applied to flexible display. However, electrical conductivity of ZnO thin film is lower than ITO thin films so, it is unsuitable for used electrode. Therefore, to improve the property of ZnO thin film, study of preparation by adding dopant (Al, Ga) is being [5]. In this study, we prepared AZO, GZO, GAZO thin film on glass substrate and analyzed the property of as-deposited AZO, GZO and GAZO thin films. To confirm the suitability of the AZO, GZO and GAZO thin film for an OLED anode, we investigated an OLED with an anode of AZO, GZO and GAZO thin film deposited by the FTS system.

Experimental

Before the deposition, the substrate was ultrasonically cleaned for remove the impurities on the glass substrate. The substrate cleaned with acetone, de-ionized water (D. I. water) and alcohol, isopropyl alcohol (IPA) for 10minutes and then dried with N₂ gas. The targets used for deposition are AZO (Al₂O₃ 2 wt.%, ZnO 98 wt.%) and GZO (Ga₂O₃ 3 wt.%, ZnO 97 wt.%). Thickness of thin films fixed at 150 nm and it prepared at room temperature. More detail experimental condition showed in Table 1.

In this study, we prepared AZO, GZO and GAZO thin films by facing targets sputtering system on the glass substrate. The FTS system designed to array two sheets of targets facing each other and the substrate is located in a plasma-free area apart from the center of plasma. As a result, sputtering atmosphere is stable and the FTS system can suppress high particle bombardment to the substrate [6–8]. So, it though that the life time of OLED which used thin film prepared by FTS system is more longer than the life time of OLED which used thin film prepared by conventional sputtering system. Also, the FTS system has another advantage. It can prepare new material thin film in stable atmosphere by installing a different target. And component ratio of thin film can change by adding the power supply. For this reason, the FTS system is though that it is suitable equipment for preparing of various thin films. The electrical, structural and optical properties of the as-deposited GAZO

Table 1. Sputtering condition

Deposition Parameter	Sputtering condition		
	AZO	GZO	GAZO
Target	AZO 4 inch	GZO 4 inch Zn 4 inch	GZO 4 inch AZO 4 inch
Gas flow	Ar = 10 sccm	Ar = 10 sccm O ₂ = 0.2 sccm	Ar = 10 sccm
Input Current		0.15A	
Substrate		Glass	
Base pressure		4.1 × 10 ⁻⁶ Torr	
Working Pressure		1 m Torr	
Thickness		150 nm	
Temperature		Room temperature	

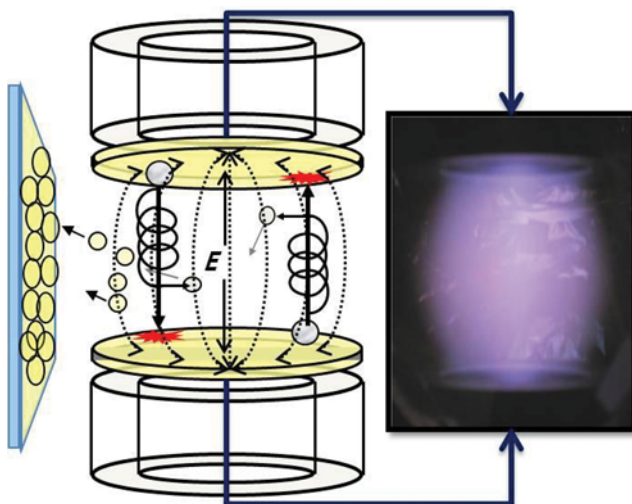


Figure 1. Schematic of facing targets sputtering (FTS) system.

thin films were then examined by hall-effect measurement, and by using an atomic force microscope (AFM), an X-ray diffractometer (XRD), and UV-VIS spectrometer. And optimized as-deposited thin film applied to OLED anode.

Figure 2 showed a cross-sectional view of OLED structure. The device structure has the configuration of ZnO-based thin films (1500 Å)/TPD (400 Å)/Alq₃ (700 Å)/LiF (5 Å)/Al (700 Å). The ZnO-based thin films were treated by plasma for 1 min before used as OLED anode. The performance of the OLED device such as the

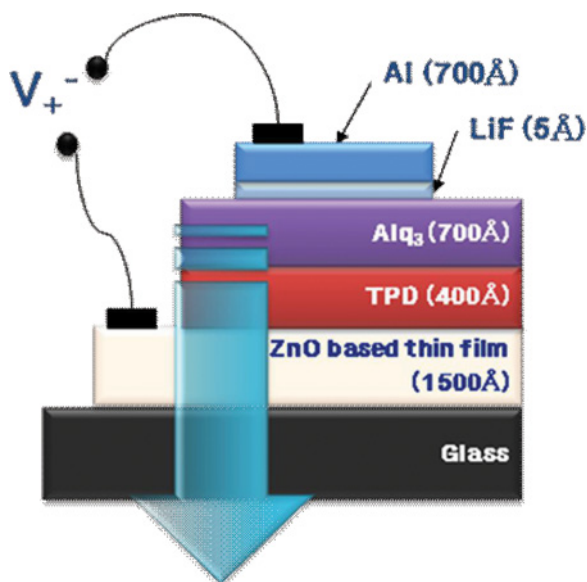


Figure 2. Structure of organic light emitting device (OLED).

operating voltage and its efficiency were evaluated by J-V-L (current density-voltage-luminance) measurements.

Results and Discussion

Figure 3 shows the optical properties of ZnO-based thin film with the optimum conditions. As deposited all the ZnO-based thin films showed approximately 85% transmittance (Wavelength = 550 nm). But transmittance of GZO thin film showed lower than AZO thin film and GAZO thin film because of using a metal target. We could observe that transmittance of as-deposited thin films has improved by adding the dopant (Al and Ga) at the same time. These result, caused by the photon scattering and ionized impurity scattering decreased due to the increasing of crystallinity due to the Al and Ga [9–10].

The relation between the absorption coefficient (α) and the photon energy ($h\nu$) can be express as [8]

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

α = Absorption coefficient, t = Thickness of thin film and T = Transmittance.

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

$h\nu$ = Photon Energy and A = constant.

As a result, ZnO based thin film showed 3.45~3.71 eV of optical band gap energy. Generally, optical band gap energy will increase because of Ga and Al is

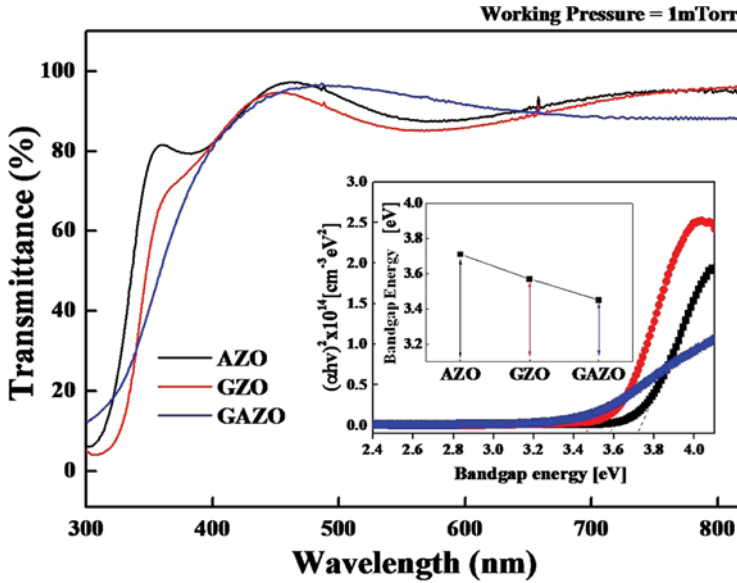


Figure 3. Optical properties of ZnO based thin films prepared on glass substrate.

substituted and crystallinity is increased. But in this study, optical band gap of GAZO thin film showed the lowest value (3.45 eV). This result though that, Ga and Al formed metallic oxide by reacts with surplus oxygen and increasing the amount of metals in thin film [11]. Although, GAZO thin film showed the lowest optical band gap energy, but it considered that GAZO thin film has high transmittance and low sheet resistance so, as deposited GAZO thin film is suitable of the OLED anode application. This property of sheet resistance can be assured with Figure 4.

Figure 4 shows the sheet resistance and figure of merits of ZnO-based thin film with the optimum conditions. The optimum condition of thin films was setting by pre-test in laboratory. The sheet resistance of as-deposited ZnO-based thin film showed 42.8 ohm/sq at GAZO thin film. This sheet resistance is improved AZO (13%) thin film and GZO (25%) thin film. These results can be explained by relationship between Al^{3+} and Ga^{3+} . The electrical property of ZnO thin film can be improved by adding dopant (Al, Ga). The reason is that Al^{3+} is effectively substituted donor for Zn^{2+} [12]. Also it is though that electrical property was improves by increasing of Zn^{2+} substitution rate and occurrence of donor due to injection of Ga at the same time. But the sheet resistance of as-deposited ZnO-based thin films showed little higher than conventional ITO thin film. However, it is considered that the electrical property can improve by post-production process like as annealing and plasma treatment [13,14]. Based on electrical and optical properties, to compare about suitability as a transparent electrode, the figure of merit value of the GAZO thin film was calculated by following formula [15].

$$\phi_{TC} = \frac{T^{10}}{R_{sh}}$$

T = Transmittance and R_{sh} = Sheet resistivity.

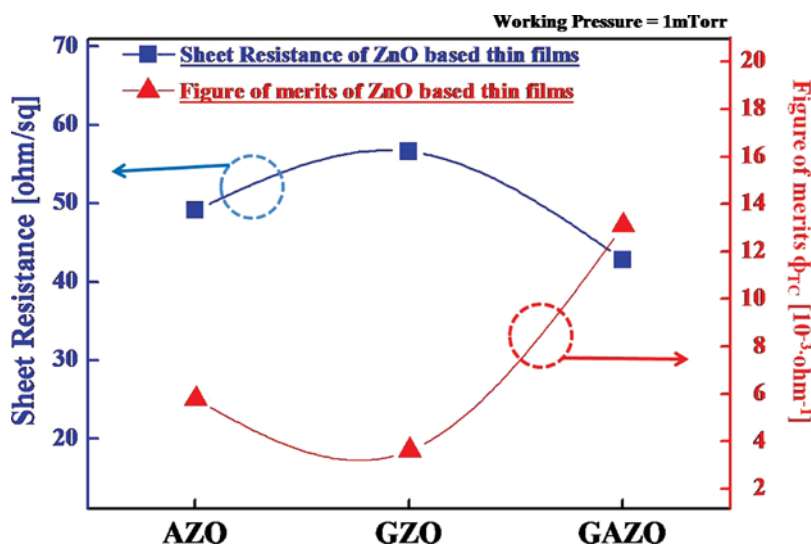


Figure 4. Sheet resistance and figure of merits of ZnO based thin films prepared on glass substrate.

As a result, we could observe that the GAZO thin films which showed high transmittance (94% at 550 nm) and lowest sheet resistance has the maximum figure of merits value.

Figure 5 shows the structural properties of as-deposited the ZnO-based thin films. As shown in X-ray diffractometer (XRD) pattern of as-deposited the ZnO-based thin films, we could observe the most improved crystalline in GAZO thin film. This result is though that Improvement of crystallinity is caused by injection of Al and Ga at the same time. By substitution of Al and Ga, crystallinity is increased and this result is effect to transmittance and electrical property [16].

In the atomic force microscope (AFM) image of as-deposited the ZnO-based thin films, the RMS (root-mean-square) value which expresses the surface roughness is important factor in the performance of OLED. Smooth surface of electrode increase the life time of OLED. On average, as-deposited ZnO-based thin films showed low RMS value, this value is lower than conventional ITO thin films (2 nm) and it showed very smooth [17]. Chief of all, RMS value of GAZO thin film showed the lowest surface roughness value (1.017 nm). As mentioned earlier, it is thought that the life time of the OLED fabricated on ZnO-based anode increase more than the life time of the OLED fabricated on ITO anode due to the relationship between the surface of electrode and life time of the OLED.

Figure 5 shows the field emission scanning electron microscope (FESEM) image of as-deposited the ZnO-based thin films. Crack or peak on the thin film surface make concentrated current, so it reduce the life time of device and efficiency of OLED [18]. However, as shown in the field emission scanning electron microscope (FESEM) image, as deposited ZnO-based thin films showed smooth surface without the defect in FESEM images. It is considered that ZnO-based thin film is suitable for application of OLED because of the ZnO thin films is composed of uniform particles on the surface.

After evaluation of as-deposited thin film properties, we expected that the GAZO thin film is the most suitable for OLED anode. So, to confirm these results,

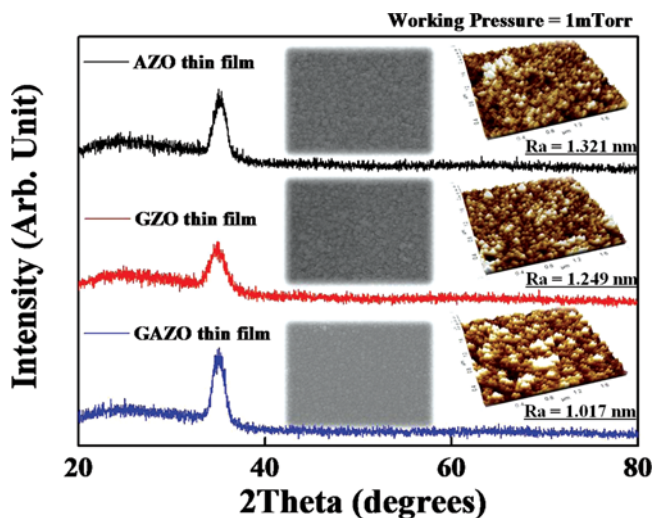
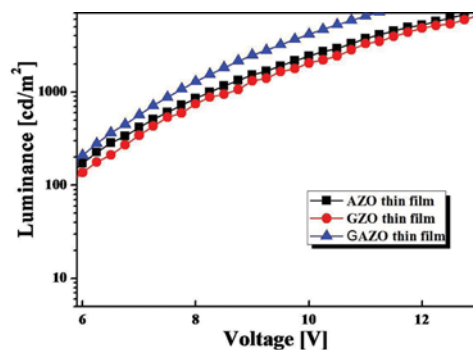


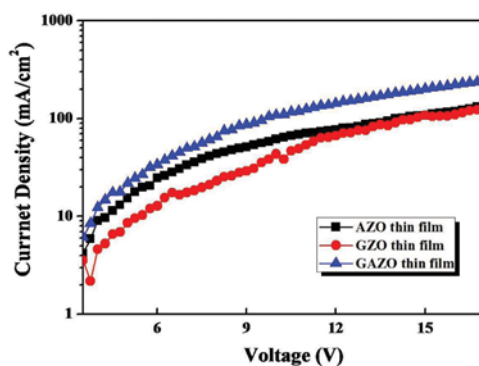
Figure 5. Structural properties of ZnO based thin films prepared on glass substrate.

we fabricated the OLED on AZO, GZO and GAZO anode after that compare the performance of OLED, respectively.

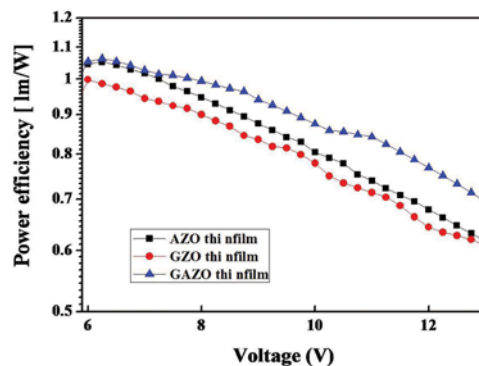
Figure 6 shows the performance of OLED fabricated on AZO, GZO and GAZO anode. We can observe that the OLED prepared on GAZO anode showed most excellent performance in luminance-voltage property. This result



(a) Luminance versus Voltage



(b) Current density versus Voltage



(c) Power efficiency versus Voltage

Figure 6. Performance of the OLED fabricated on ZnO based thin film.

of experiment can explain by the relation between the luminance of OLED and the sheet resistance of GAZO anode. Low sheet resistance of OLED anode increases the luminescence uniformity of OLED [19]. Therefore, as shown in Figure 6, the OLED fabricated on GAZO anode showed high luminance due to the low resistivity of GAZO anode.

Conclusions

In this study, to replace the ITO thin films for OLED anode, we prepared ZnO-based thin films by using a dopant (Al and Ga) and the properties of ZnO based-thin films were examined by various methods. As a result, ZnO-based thin films showed high transmittance (85%) and low sheet resistance. Chief of all, GAZO thin film showed the lowest sheet resistance (42.8 ohm/sq), high transmittance and smooth surface (RMS = 1.017 nm), so we can confirmed that GAZO thin film is most suitable to use OLED anode. And the result of investigation about the performance of OLED fabricated on ZnO-based thin film is that the performance of OLED fabricated on GAZO anode is most excellent.

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