Properties of IZTO Thin Films Deposited on PEN Substrates with Different Working Pressures

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ABSTRACT

In this work, the properties of Indium-Zinc-Tin-Oxide (IZTO) thin films, deposited on polyethylene naphthalate (PEN) with a SiO₂ buffer layer, were analyzed with different working pressures. After depositing the SiO₂ buffer layer on PEN substrates by plasma-enhanced chemical vapor deposition (PECVD), the IZTO thin films were deposited by RF magnetron sputtering with 1 to 7-mTorr working pressure. All the IZTO thin films show an amorphous structure, regardless of the working pressure. The best morphological, electrical, and optical properties are obtained at 3-mTorr working pressure, with a surface roughness of 2.112-nm, a sheet resistance of 8.87-Ω/sq, and a transmittance at 550-nm of 88.44%. These results indicate that IZTO thin films deposited on PEN have outstanding electrical and optical properties, and the PEN plastic substrate is a suitable material for display devices.

Key words : IZTO, Transparent conductive oxide, SiO₂ buffer layer, Thin film

1. Introduction

Recently, as wearable display technology receives greater attention, requisite flexible display technology is also being more intensively studied. When thin film glass or polymer plastics such as polyethylene terephthalate (PET), polyimide (PI), polyether sulfone (PES), polycarbonate (PC), polyethylene naphthalate (PEN), etc. are used as a substrate, devices with flexible properties can be produced.¹⁾ The above plastic substrates have limitations, however, in use for processes requiring heat treatment, since they are easily deformed by heat owing to their somewhat inferior physical characteristics such as elongation and tensile strength as well as their poor heat resistance compared with glass substrates currently in wide use. However, since plastics have not only flexible properties but also are reasonably affordable and light in comparison with glass, thereby allowing mass production, they are nonetheless receiving attention as substrates for display devices.²⁾ In addition, plastics have problems of degraded thin film properties by infiltration of moisture and gases absorbed. The problem of these impurities has been somewhat improved by applying a buffer layer.²⁾

Although among plastics PET has a low glass transition temperature (T_g), which causes its properties to be deteriorated by crystallization and presents processing limitations, many studies on its application as a new substrate for flexi-

ble display devices have been conducted due to its strength of having a very low coefficient of thermal expansion (CTE) in comparison with other plastic substrates. This is considered an even more important property than the glass transition temperature.^{1,3)} PEN meanwhile has been improved through recently developed polymer synthesis techniques to overcome its weakness of a low glass transition temperature, while having a low coefficient of thermal expansion as the strength of PET.⁴⁾ Despite this strength, however, it is very difficult to find an example of the studies where PEN has been applied as a substrate for flexible display devices.

Among display devices such as organic light-emitting diodes (OLEDs), liquid crystal displays (LCDs), etc., indium-tin-oxide (ITO) is being used as a transparent conductive oxide due to its high transmittance and electrical conductivity. However, it is difficult to deposit ITO on plastic substrates at a low temperature, as high electrical and optical properties are exhibited when crystallized by deposition at an elevated temperature.³⁾ To solve this problem, ITO can be replaced by indium-zinc-tin-oxide (IZTO). By doping with zinc, crystallization is hindered due to the immiscibility of zinc and tin within the In₂O₃ structure, thereby producing a stable amorphous structure. Since IZTO with this amorphous structure allows easy deposition even at a low temperature while having a low surface roughness and high electrical and optical properties, it is suitable for use as a transparent conductive oxide of next-generation flexible display devices.²⁾

In this paper, before depositing IZTO thin film, SiO₂ was formed as a buffer layer on PEN plastic substrates having a high glass transition temperature together with a low coefficient of thermal expansion, followed by an analysis of the

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structural, morphological, electrical, and optical properties of IZTO thin films deposited under various working pressures. Also, changes in the properties of IZTO thin films as a function of working pressure have been studied as a figure of merit calculated from measured results.

2. Experimental Procedure

2.1 Cleaning of substrate

PEN substrates of 1 inch × 1 inch size (125-um, Teijin-Dupont, TEONEX Q65HA) were ultrasonically cleaned for 3 minutes in alcohol and deionized water since deformation may be caused by acetone, and subsequently dried using nitrogen gas.

2.2 Deposition of SiO₂ buffer layer

SiO₂ as a buffer layer was deposited to a thickness of 30-nm by using plasma enhanced chemical vapor deposition (PECVD), as shown in Table 1.

2.3 Deposition of IZTO thin film

IZTO thin films were deposited by RF magnetron sputtering with working pressure changed as shown in Table 2, from an IZTO (In₂O₃ 90 wt.%; ZnO 5 wt.%; SnO₂ 5 wt.%) ceramic target of 2 inch diameter manufactured by sintering.

2.4 Measurement method

Structural properties of the deposited IZTO thin films were analyzed by the X-ray diffraction method (HR-XRD, Xpert-Pro, MRD), while the morphological properties were analyzed using an atomic force microscope (AFM, SII Nano Technology, SPA400). Electrical and optical properties were investigated by using Hall-effect measurement (Accent, HL5500PC) and a UV-Vis spectrometer (Varian, Cary-500), respectively. By using the sheet resistance and transmittance measured by the above methods, a Figure of Merit designed by Haacke was calculated to study the suitability

as a transparent conductive oxide under varied working pressures.

3. Results and Discussion

Figure 1 shows the X-ray diffraction pattern of an IZTO thin film deposited on a PEN substrate with the formation of a 30-nm thick SiO₂ buffer layer. The intensive high peaks of the PEN substrate corresponding to 2θ = 23°, 27° were confirmed to have been reduced by the deposition of SiO₂, and IZTO was confirmed to have an amorphous structure under all working pressures since no other peaks were detected.

Fig. 1. X-ray diffraction patterns of IZTO thin film with various working pressures.

Fig. 2. AFM image and surface roughness of IZTO thin films with various working pressures.

Table 1. Process Conditions of the PECVD

Working Pressure	800 mTorr
Working Temperature	Room Temperature
RF power	40 W
N density	20 sccm
N ₂ O density	200 sccm
SiH ₄ density	600 sccm

Table 2. Process Conditions of RF Magnetron Sputtering

Base Pressure	4.5 × 10 ⁻⁶ Pa
Active gas (Ar) density	20 sccm
Working pressure	1, 3, 5, 7 mTorr
Working temperature	Room temperature
RF power	50 W
Deposition time	15 min

Figure 2 shows a morphological image and surface roughness of the cleaned PEN substrate and the IZTO thin film deposited thereon. The surface roughness is given by the root mean square (RMS) value. Bare PEN substrates show a very low surface roughness of 0.412-nm, and there are no pinholes or cracks in any of the IZTO thin films, as the SiO₂ deposited earlier properly performed the role of a buffer. The surface roughness of the IZTO thin films thus had relatively low values of 2.112 ~ 3.576-nm according to the working pressure, even though the IZTO thin films were deposited to considerable thickness, as shown in Table 3 below. The lowest surface roughness of 2.112-nm was exhibited when the working pressure was 3-mTorr. Since carriers are generated by substituting zinc and tin in the IZTO thin film, it has been reported that the surface morphologies can affect the electrical properties and better properties tend to be exhibited with lower surface roughness.^{2,3)}

As shown in Fig. 3, the specific resistance and the sheet resistance of the IZTO thin films showed a declining trend from $3.90 \times 10^{-4} \Omega\text{-cm}$ to $2.13 \times 10^{-4} \Omega\text{-cm}$ and from 17.7- Ω/sq to 8.87- Ω/sq , respectively, as the working pressure was decreased from 7-mTorr to 3-mTorr, and increased again to $4.14 \times 10^{-4} \Omega\text{-cm}$, 21.8- Ω/sq , respectively, at 1-mTorr. It was reported that thin films having higher mobility are formed when the working pressure is lower and that thin films have a lower resistivity.⁵⁾ However, if the working pressures are lowered excessively, the sputtered particles' mean free paths are extended, and these particles get much higher energy, potentially resulting in defects in the thin films.⁵⁾ The high surface roughness when the working pressure is 1-mTorr observed in the AFM image of Fig. 2 can also be attributed to an increase in the mean free path, as described above. Because of this phenomenon, electrical properties have been confirmed to be superior at $2.13 \times 10^{-4} \Omega\text{-cm}$ for

Table 3. Thickness of IZTO Deposited by RF Magnetron Sputtering

Working Pressure (mTorr)	1	3	5	7
IZTO Thickness (nm)	190	240	220	220

Fig. 3. Resistivity and sheet resistance of IZTO thin film as a function of working pressure.

specific resistance and 8.87- Ω/sq for sheet resistance when the working pressure is 3 mTorr.

Figure 4 shows the transmittance of IZTO/SiO₂/PEN films as a function of working pressure and of a cleaned PEN substrate. As shown in this figure, the characteristics of PEN where light is absorbed in the ultraviolet domain while transmitted from 380-nm as a wavelength band for violet can be seen to be exhibited as they stand. While the average transmittance is low compared with PET and glass substrates, transmittance at 550-nm where human vision is the most sensitive is even higher when PEN was used.

In Fig. 5, the figure of merit (Φ_{TC}) for a transparent conductive oxide designed by Haacke has been calculated through equation (1), given below, by using transmittance of 550-nm, T_{550} , and sheet resistance, R_{sheet} , as determined from Fig. 4 and Fig. 3.⁶⁾

$$\Phi_{TC} = (T_{550})^{10} / R_{\text{sheet}} \quad (1)$$

As shown in Fig. 5, the calculated figure of merits show the same tendency as that for surface roughness and the

Fig. 4. Transmittance of PEN/SiO₂/IZTO films with various working pressures.

Fig. 5. Transmittance at 550-nm & Figure of merits of IZTO/SiO₂/PEN films with various working pressures.

electrical properties analyzed earlier. When the working pressure is 3-mTorr, not only the sheet resistance value is lowest but also the transmittance at 550-nm is highest, so the highest figure of merits, $3.30 \times 10^{-2} \cdot \Omega^{-1}$, has been calculated.

4. Conclusions

In this paper, optimum process conditions have been identified using a PEN substrate to overcome the low glass transition temperature impeding application of PET plastic. In addition, a 30-nm thick SiO_2 buffer layer is deposited to mitigate the drawbacks of plastic substrates, followed by an analysis of IZTO thin films deposited by RF magnetron sputtering with working pressure varied between 1 ~ 7-mTorr.

As a result, all IZTO thin films were confirmed to be amorphous through an X-ray diffraction analysis, while all IZTO thin films were also confirmed to be free of pinholes or cracks, regardless of the working pressure, through AFM measurement. In particular, when the working pressure was 3-mTorr, the lowest surface roughness of 2.112-nm was obtained. In addition, the highest electrical properties were observed through Hall effect measurement at a resistivity of $2.13 \times 10^{-4} \Omega\text{-cm}$, and a sheet resistance of 8.87- Ω/sq , while the transmittance at 550-nm, where human vision is the most sensitive, showed the highest value of 88.44%, when the working pressure was 3-mTorr. A figure of merit calculated using the sheet resistance and transmittance also showed the best value of $3.30 \times 10^{-2} \cdot \Omega^{-1}$ when the working pressure was 3-mTorr.

The results presented in this paper confirm that a PEN substrate with a high glass transition temperature accom-

panied by excellent heat resistance with an IZTO thin film deposited on the PEN to provide excellent electrical and optical properties is a promising material that can be applied to next-generation flexible display devices.

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