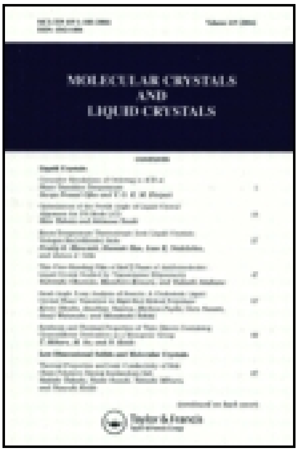

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Investigation of Barrier Layer Deposited on Flexible Polymers Substrate by Facing Target Sputtering System

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Investigation of Barrier Layer Deposited on Flexible Polymers Substrate by Facing Target Sputtering System

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A multilayer structure consisting of an indium tin oxide conducting layer, and a multi component barrier film (SiO_2 and ITO) on polyethylenenapthalate is proposed as a transparent flexible substrate for flat panel displays and photovoltaic. The ITO and barrier films were deposited by DC magnetron sputtering and a facing target sputtering (FTS) system, respectively. To improve the multilayer film properties, a 30, 50, 80 nm-thick barrier layer was sputtered by FTS method prior to the ITO film sputtering process. The ITO films on the barrier film showed good properties due to the decreased lattice mismatch between the ITO and barrier film.

Keywords Barrier film; facing target sputter; sputtering system; WVTR; ITO; flexible substrate

Introduction

Transparent and conducting indium tin oxide (ITO) films are used extensively as transparent electrodes for flat panel displays (FPDs), touch panels and photovoltaic [1,2]. Most properties of ITO films depend on the deposition techniques used and the composition of the ITO film [3,4]. In the case of flexible substrates, this study is based on the critical point for FPDs and photovoltaic applications [5,6]. In addition, flexible substrates have the advantages of mechanical flexibility, optical transparency, low weight and cost effectiveness [7].

Inorganic transparent oxide films (e.g. silicon and aluminum oxide) on polymer films have been used widely as gas barrier materials for display and photovoltaic applications [8]. A number of inorganic functional coatings on polymers have critical properties, such as water vapor impermeability and low resistivity. For example, water vapor impermeability is achieved by coating polymers with inorganic transparent oxide films, and a lower resistivity is achieved using a transparent conducting oxide (TCO) films, often coated on the barrier [6].

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Therefore, it is important to have a clear understanding of the mechanisms in multilayer, thin film barrier coatings on polymeric substrates. Recently, the behavior of thin films coated on flexible substrates was reported for an individual water vapor permeation oxide barrier [8,9] and TCOs [10,11]. However, the behavior of the complex multilayer is not well understood. For example, optimal thickness control, and lattice mismatch between the barrier and TCO films has not been considered. Therefore, to decrease the lattice mismatch between the barrier and TCO film, and improve the conductivity TCO films, this study developed barrier films with a new composition, such as multi component SiO_2 and ITO film, using facing target sputtering (FTS) system with two cathodes without substrate heating.

The electrical, surface roughness, optical and water vapor transmission rate (WVTR) properties of ITO/barrier film (multi component SiO_2 and ITO) on the polyethylenenaptha-late (PEN) were investigated, and the influence of the barrier film thickness is discussed. These results show that the barrier film has a significant effect on the properties of the ITO film.

Experimental

Multi components of SiO_2 and ITO (SiO_2:ITO) films were grown on unheated PEN substrates using a FTS system with two cathodes. Two sintered ceramic targets (SiO_2 and ITO), 3 inch in diameter, were set facing approximately 70 mm apart. A barrier film was deposited to a thickness of 30, 50 and 80 nm by radio frequency (13.56 MHz) magnetron sputtering. The total working pressure, Ar flow rate and RF power were 3 mTorr, 20 sccm and 150 W, respectively. After deposition of barrier film, the ITO films were deposited on the substrates using an ITO target (In_2O_3:SnO_2 = 90:10 wt%) using a conventional DC magnetron sputtering process. The ITO films were deposited at an Ar flow rate, working pressure and DC power of 20 sccm, 3 mTorr and 150 W, respectively.

The permeability of the films was measured using a Mocon permeability tester (AQUA-TRAN model 1, Mocon). The permeability measurements were carried out at a relative humidity (RH) of 37.8°C@100%. The test cells were divided into two chambers separated by the sample. The inner and outer chamber was filled with nitrogen and water vapor, respectively. The permeation rate through the film is referred to as the transmission rate (TR).

The electrical properties, resistivity (ρ), hall mobility (μ) and free carrier density (n) of the ITO films were measured using Hall-effect measurements (HMS-3000, ECOPIA) in a van der Pauw geometry. The surface morphology of the films was analyzed by atomic force microscopy (AFM, NanoScope 3, Digital instruments) and the transmittance of the films were measured from 200 to 800 nm using a UV-Vis spectrophotometer [Lambda 950, Perkin Elmer].

The mechanical properties of ITO film were estimated by the changes in resistance during the cyclic bending test, which were monitored by a computer system with an digital multimeter [Agilent 34401A]. The samples used in the cyclic test were rectangular in shape, 20 mm × 50 mm. The bending tests were carried out at a frequency of 0.08 Hz and constant linear vertical movement with a 20 mm stroke. The cyclic bending stress was applied dynamically on the films using a moving jig.

Results and Discussion

Figure 1 shows the effect of the barrier film thickness on the WVTR of the (a) barrier (SiO_2:ITO)/PEN and (b) ITO (150 nm)/barrier (SiO_2:ITO)/PEN films with FTS system and

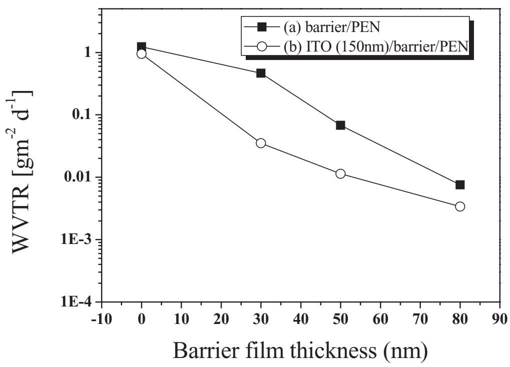


Figure 1. Change in the WVTR for (a) barrier/PEN and (b) ITO(150 nm)/barrier/PEN for each barrier film with various thicknesses.

conventional DC magnetron sputtering method. In the case of (a) barrier (SiO_2:ITO)/PEN, a faster decrease in the WVTR value with barrier film up to 80 nm was associated with the decrease in high energetic particles though the FTS system [12,13] Generally, the permeation increases because a thin layer is limited by transport through coating defects, such as pinholes, grain boundaries, micro cracks, intrinsic stress [4,14]. The coating defects in the conventional DC magnetron sputtering process deposited thin film result from film damage during growth of a thin film caused by the bombardment of high energetic particle s, such as negative oxygen ions (O^-) or reflected argon atoms (Ar^o) [12]. Therefore, the loss of high energetic particles during the growth of a barrier film was controlled by the FTS system, as suggested in recent studies. In the case of (b) ITO (150 nm)/barrier (SiO_2:ITO)/PEN films, the WVTR values showed a similar tendency with the barrier film thicknesses. Therefore, the ITO films are strongly affected by the permeability of the barrier film. Also, the WVTR values in Figs. 1(b) relatively decreases for all films, as compared to

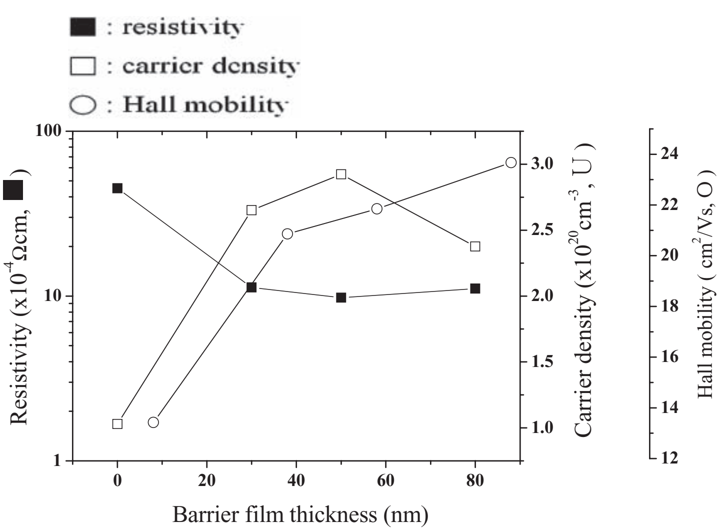
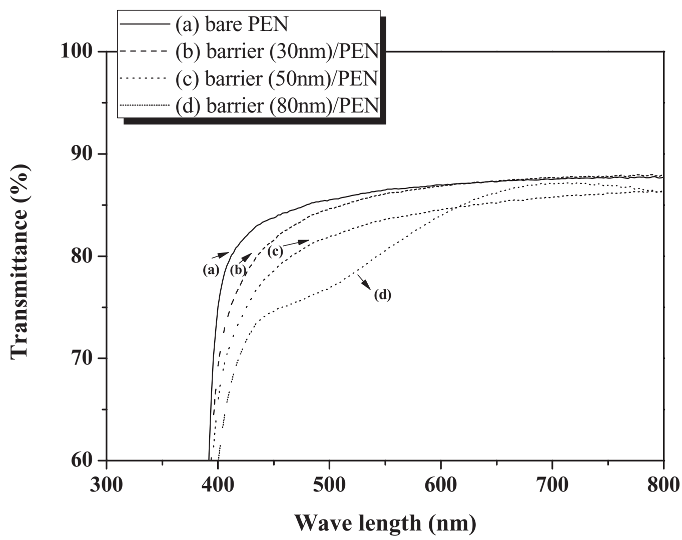


Figure 2. Resistivity, carrier density and Hall mobility of ITO films deposited on the barrier film with various thicknesses.

that in Fig. 1(a). This behavior can be explained by an increase the permeability path with ITO film deposition on the barrier film [6].

Figure 2 shows the resistivity, carrier density and Hall mobility of the multilayer deposited with ITO films on a barrier film with various thicknesses. In the case of the ITO (150 nm)/PEN film, relatively high resistivity (4.508×10^{-3} Ωcm) was obtained without the barrier film. However, the resistivity was significantly lower with increased the barrier film. On the other hand, no large change in the resistivity of the ITO films was observed when the barrier film thickness was increased from 30, 50, 80 nm. In the case of ITO films on a barrier film, the lattice mismatch between ITO and the barrier film was decreased by the additional ITO composition in the barrier film. The decreased mismatch between ITO and the barrier film probably induced an increase in carrier density, i.e., the resistivity



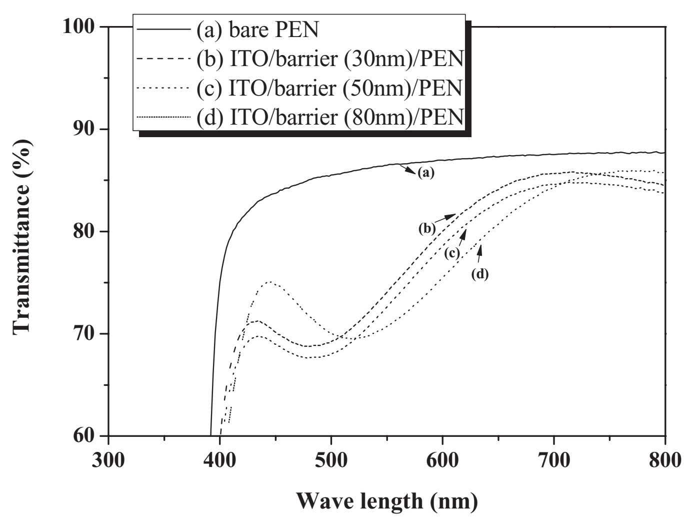


Figure 3. Transmittance of the barrier/PEN and ITO(150 nm)/barrier/PEN deposited on barrier films with various thicknesses. (a) PEN, (b,c) barrier/PEN and (d) ITO(150 nm)/barrier/PEN.

decreased and the Hall mobility increased with a constant ITO film thickness. On the other hand, the carrier density of the ITO(150 nm)/barrier layer (80 nm)/PEN film decreased with increasing Hall mobility. Generally, the scattering center of the carrier that dominates the mobility in degenerated TCO films is believed to be ionization impurity scattering and neutral impurity scattering [15]. The Hall mobility increased due to a decrease in ionization impurity scattering caused by the decrease in carrier density [15]. Therefore, the resistivity of the ITO film increased slightly with a barrier film thickness of 80 nm, which could be related to ionization impurity scattering.

Figure 3 shows the optical transmittance over the wavelength range, 380∼780 nm, of the barrier/PEN and ITO/barrier/PEN films under different barrier film thicknesses: 30 nm, 50 nm and 80 nm. The transmittance of the barrier/PEN film was about 80% at the visible light region for all films. The transmittance of films were decreased by increases in the barrier layer thickness. This can be explained by the similar refractive index (RI, denoted by n) between ITO and barrier film. Thus, the transmittance decreased at visible light region due to the increase in reflectance with the increase of barrier film.

Figure 4 (a), (b), (c), (d) shows the change in resistance ($\Delta R/R_0$) of the ITO films under dynamic stress mode, where R_0 and ΔR represent the initial resistance and difference between the initial and final resistance, respectively. A comparison of Fig. 4(a),(b),(c) and (d) showed that the resistance of the ITO films decreased remarkably with barrier layer thickness of 30 nm. The growth of the ITO films on the SiO_2:ITO (30 nm)/PEN film decreased its resistance significantly compared to that of the ITO films directly grown on PEN. This can be explained by structural differences in the films grown using FTS and

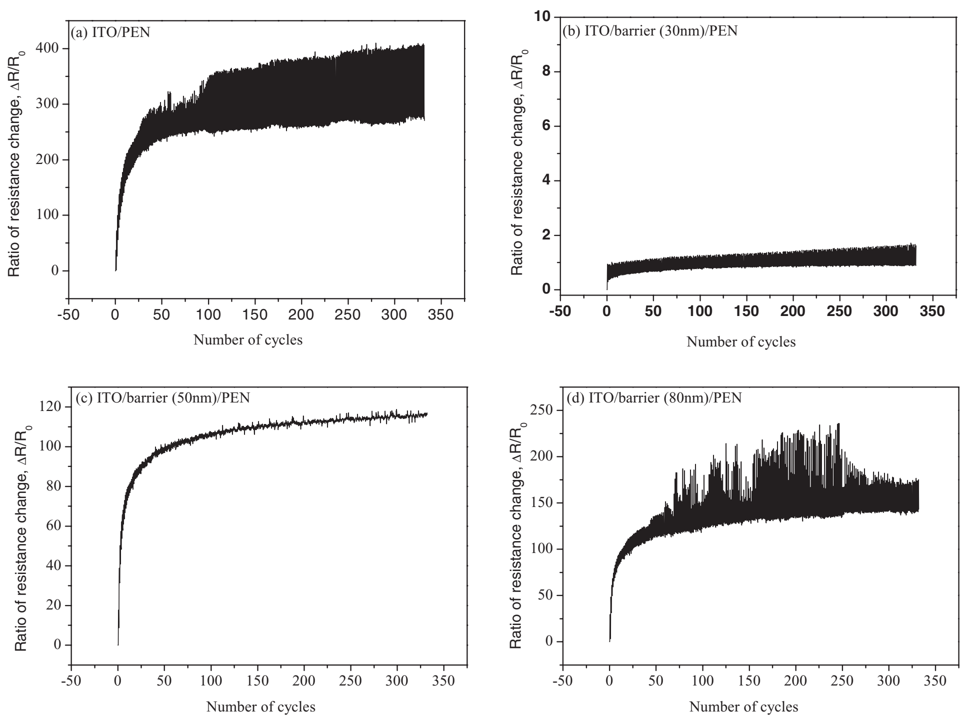


Figure 4. Change in the resistance for the ITO films on the barrier films with various thicknesses under cyclic mode.

conventional sputtering processes. A dense barrier layer with a small number of defects was grown by the FTS process due to a decrease in the bombardment of high energetic particles [12]. In contrast, the ITO films grown by the conventional sputtering process were exposed to the bombardment of high energetic particles. This led to the poor bending performance of the sputtered ITO films because of the formation of a large number of cracks even under small induced stress. However, an increase in the barrier film thickness in the ITO/barrier/PET films resulted in an increase in the resistance of the ITO films. This can be explained by difference of the internal stress between the ITO and barrier film. Very large internal stresses are induced at the interface between the ITO and the barrier film because of the large difference in the thermal expansion and elastic properties of ITO and barrier film [6]. The continuous stress caused by the increase in the barrier thickness leads to adhesive failure in the barrier films on the PEN substrate. As a result, this led to poor bending performance for the sputtered ITO films because of the formation of a large number of cracks even under a small induced stress.

Conclusions

The ITO/SiO_2:ITO/PEN films were investigated by carrying out the two-step process such as FTS and conventional sputtering processes without unheated substrate. Relatively good film properties (mechanical, electrical, and permeability) were obtained for the ITO films deposited with barrier film compare to ITO/PEN film. Also, it is therefore confirmed that ITO film properties were strongly affected by the barrier film. In particular, electrical property of ITO film on the barrier film was decreased with barrier layer due to decreased lattice mismatch between ITO and barrier film. The minimum resistivity of the 9.7×10^{-4} Ωcm was obtained at barrier film thickness 50 nm owing to the increase in the carrier concentration and Hall mobility. Also, the ITO film on the barrier film have good effect on the mechanical property and permeability as well as the electrical property.

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