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Transparent barrier coatings for flexible organic light-emitting diode applications

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1. Introduction

Transparent barrier coatings on polymers are receiving much attention in industries [1,2]. A major application area, namely, polymer-based organic light-emitting diode displays require perfect encapsulation against inward permeation of water and oxygen [3]. In this study, a multiple stack of SiN_x, SiO_x, and parylene layers was deposited by plasma-enhanced chemical vapor deposition and a parylene reactor. Comparing the results of film stress, transmittance, surface roughness and permeability, we determined a multilayer composed of parylene/SiO_x/SiN_x…parylene/SiO_x /SiN_x (PON…PON) on the polycarbonate substrates and the thickness of SiN_x, SiO_x and parylene. It is believed that the barrier of multilayer structure can achieve the flexible organic light-emitting diode requirements.

2. Experiment

The SiN_x and SiO_x films were deposited by Plasma-enhanced CVD and the parylene coatings were deposited by the Parylene reactor. The optimum deposition parameters of SiN_x films were SiH_4/NH_3 flow rate 130/20 sccm, rf power 180 W, pressure 800 mTorr, and temperature 80°C; SiO_x films were SiH_4/N_2O flow rate 150/100 sccm, rf power 80 W, pressure 800 mTorr, and temperature 80°C to prevent any deformation of the parylene coatings and the PC substrates. Internal stress in the SiN_x and SiO_x films on 4" Si wafers were determined by a Tencor-KLA (FLX-2320) laser profilometer. Surface morphologies of SiN_x, SiO_x and parylene were analyzed by atomic force microscopy (PSI Auto Probe). Wyko NT1100 (Veeco) optical profiler was used to obtain the large fields of view over SiO_x surface. Measurements of OTR and WVTR permeation were carried out on a 10-cm^2 active sample area, at 40°C, using MOCON “Ox-tran 2/61” and “Permeation W3/61” instruments, respectively; for OTR measurements, we used 0% relative humidity. Finally, we capped OLEDs with single and double PON layers on glass substrate for comparison.

3. Results and discussion

In Fig. 1, SiN_x and SiO_x films show a critical coating thickness and further increasing the coating thickness did not reduce the permeation significantly. Besides, SiN_x films have better impermeability than SiO_x films. It is known that moisture reacts with Si-O bonds and affects the barrier performance. However, transmittance in visible light region is one of the requirements for display to use as barrier films. To solve this problem, we combined SiN_x and SiO_x layers to enhance the transmittance and the impermeability as well. For the barrier films to have good mechanical properties, internal stress is a major concern. Higher compressive internal stress in the coatings is beneficial to the film density, barrier performance and adhesion [4]. Fig. 2 shows the internal stress versus the film thickness of SiN_x at different RF power conditions. Thicker coatings are associated with lower compressive stress because the stress relaxation process occurs as the coating growths. By adjusting the RF power, the WVTR and OTR value of 50 nm SiN_x film can be reduced from 0.18 g/m^2/day to 0.035 g/m^2/day and 5.5 cc/m^2/day to 0.95 cc/m^2/day with the internal stress varying from -38.8 MPa to -102 MPa. To better understanding the moisture-resistant behavior of the barrier films, the bending results of SiN_x were illustrated in Fig. 3. The WVTR values of thicker coatings tend to increase while the values of thinner coatings do not change significantly. Consequently, we chose the SiN_x and SiO_x films with lower thickness but higher compressive internal stress. In Fig. 4, the SiO_x/SiN_x on PC demonstrated higher value of transmittance than SiN_x/SiO_x on PC. In Table 1, both SiN_x/SiO_x and SiO_x/SiN_x on PC substrate reduce the WVTR values to 0.01 g/m^2/day. Nevertheless, only SiO_x/SiN_x reduces the OTR values significantly. The surface roughness of SiN_x was about 1.6 nm. But the high surface roughness and thick sharp spikes of SiO_x films show in Fig. 5 would cause more pinhole defects in the growing SiN_x films. Therefore, parylene layers were used as a smoothing and defect-decoupling organic medium. The parylene layer shows an undulated morphology with no sharp spikes that could cause a reduction in the barrier performance. By inserting a parylene layer into the SiN_x/SiO_x structure, the WVTR and OTR values were reduced to 0.01 g/m^2/day and 0.1cc/m^2/day. In Fig. 6, the SiO_x layer shows some scratches which might affect the barrier performance. Hence, it is necessary to provide a thick parylene layer as a protective layer on the top of the barrier films. In Fig. 7, the OLEDs capped with double PON layers show no dark spot and better emission than the single layer one. It proves that repeating the PON layers can improve the barrier performance.

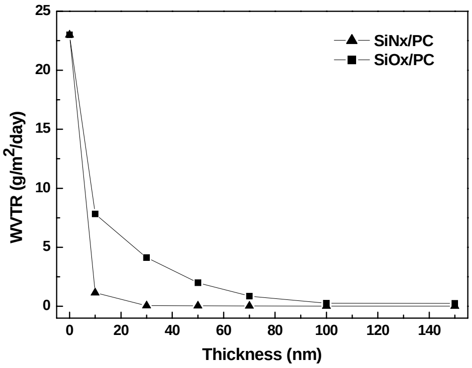

Fig.1. WVTR of PECVD SiN_x and SiO_x films on PC substrates as a function of film thickness.

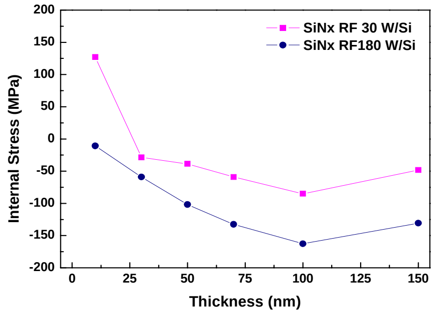

Fig. 2. Internal stress of SiN_x deposited on Si wafer as a function of film thickness.

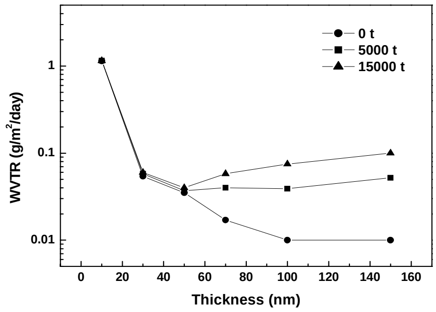

Fig. 3. WVTR of SiN_x film on PC substrates bended for 0, 5000 and 15000 times as a function of film thickness.

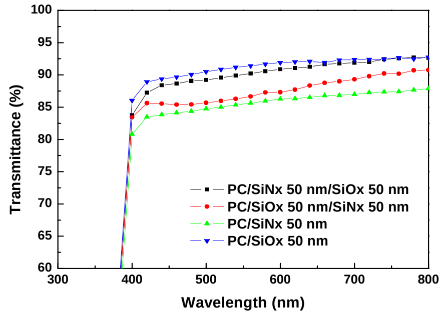

Fig. 4. Transmittance of SiN_x, SiO_x, SiN_x/SiO_x and SiO_x/SiN_x on PC substrates as a function of wavelength.

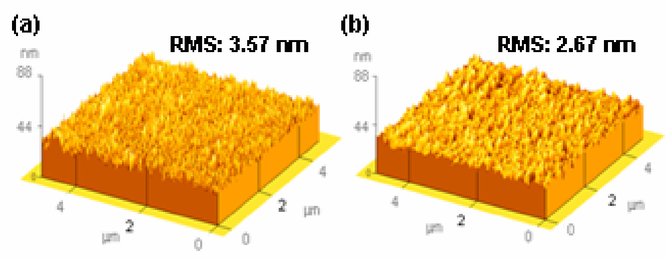

Fig. 5. Three-dimensional AFM surface morphologies (a) 50 nm-SiO_x (b) 150 nm-parylene/50 nm-SiO_x on PC substrate.

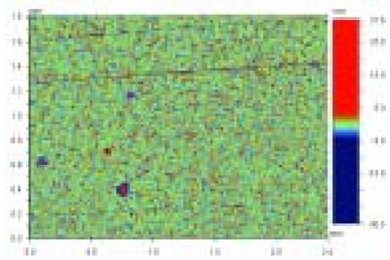

Fig. 6. Two-dimensional surface morphologies of 50 nm-SiO_x film on PC substrates.

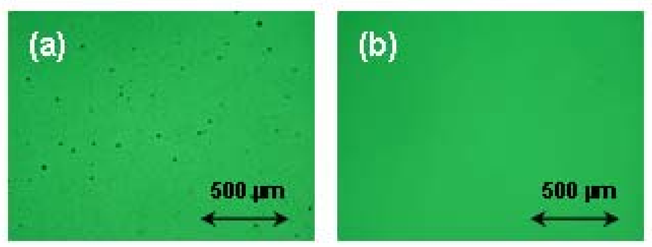

Fig. 7. Comparison of emission characteristics of (a) single PON and (b) double PON layers capped OLEDs after 100 h under 25°C, 40%RH.

Table I WVTR and OTR for various coatings

Barrier system	WVTR ($g/m^2/day$)	OTR ($cc/m^2/day$)
Polycarbonate (178 µm)	~20	~700
Parylene (150 nm)/PC	~20	~700
SiOx (50 nm)/PC	~1.15	~0.75
SiNx (50 nm)/PC	~0.035	~0.95
SiNx/SiOx/PC	~0.01	~0.7
SiOx /SiNx/PC	~0.01	~0.1
SiNx/Parylene/SiOx/PC	~0.01	~0.1

4. Conclusions

In this study, we have described the promising of using parylene(450 nm)/SiO_x(50 nm)/SiN_x(50 nm)…parylene(150 nm)/SiO_x(50 nm)/SiN_x(50 nm) on polycarbonate substrate as moisture and oxygen permeation barriers. Combining the SiO_x film with SiN_x, we prevent the aging effects of SiO_x films and enhance the transparency in visible light region as well. The SiN_x and SiO_x films with lower thickness but higher compressive internal stress were chosen to have both impermeability and good mechanical properties. Under optimum condition, the WVTR and OTR of SiO_x/SiN_x barrier coatings on PC at 80°C decrease to a value of near 0.01 g/m^2/day and 0.1 cc/m^2/day, respectively. By inserting an organic medium, parylene, the WVTR and OTR of SiN_x/parylene/SiO_x on PC at 80°C also decreased because the improved SiN_x film properties deposited on parylene layer. Finally, we capped the barrier films with parylene layer to avoid the inorganic layers from being scratched.

Acknowledgements

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