Journal of Information Display, vol. 5, no. 3, 2004

Ultra Thin Film Encapsulation of OLED on Plastic Substrate

Sang-Hee Ko Park*, Jiyoung Oh, Chi-Sun Hwang*, Yong Suk Yang, Jeong-Ik Lee, and Hye Yong Chu

Abstract

Fabrications of barrier layer on a polyethersulfon (PES) film and OLED based on a plastic substrate by atomic layer deposition (ALD) have been carried out. Simultaneous deposition of 30 nm of AlO_x film on both sides of PES film gave film MOCON value of 0.0615 g/m^2/day (@38°C, 100 % R.H.). Moreover, the double layer of 200 nm SiN_x film deposited by PECVD and 20 nm of AlO_x film by ALD resulted in the MOCON value lower than the detection limit of MOCON. The OLED encapsulation performance of the double layer have been investigated using the OLED structure of ITO/MTDATA(20 nm)/NPD(40 nm)/AlQ(60 nm)/LiF(1 nm)/Al(75 nm) based on the plastic substrate. Preliminary life time to 91 % of initial luminance (1300 cd/m^2) was 260 hours for the OLED encapsulated with 100 nm of PECVD deposited SiN_x/30 nm of ALD deposited AlO_x.

Keywords : atomic layer deposition, encapsulation, thin film, OLED, plastic

1. Introduction

The flexible displays, which could be embedded in our clothes or show information in our environment on demand and be disappeared when we're done with it, have attracted much attention as the next-generation display. The microparticle-based display [1], Liquid Crystal Display (LCD) [2], and Organic Light Emitting Diodes (OLEDs) [3] have intrigued researchers as a candidate of flexible display.

Among them, OLEDs that could be thin, robust, lightweight, and have high information contents have been at the focus of much research activities [4, 5]. It has been recognized that the development of thin film barrier (encapsulation) layers for OLED and flexible substrates are the key technology for the realization of flexible OLEDs [6]. Although the requirement for barrier layer of OLED display is not elucidated clearly, it has been understood that long-lived flexible OLEDs need a moisture barrier layer which transmits less than 10^{-6} g/m^2/day of water and 10^{-5} cc/m^2/day of oxygen [7].

It has been reported that multi layer combinations of polymer and inorganic dielectric layer can be more than three orders of magnitude less permeable to water and oxygen than an inorganic single layer [7,8]. Several groups have reported multi-layered barriers consisting of inorganic thin films fabricated by plasma enhanced chemical vapour deposition (PECVD) or sputtering [9,10]. To be a good barrier layer, it should not only be free of pinholes also have good step coverage. Especially, it is also very important for the barrier deposition process to be compatible with the organic emitting layer beneath [11].

Our group reported a new approach to the barrier layers of flexible OLED substrates using travelling wave reactor typed atomic layer deposition (ALD) [12] and plasma enhanced ALD (PEALD) [13]. ALD is a technique using a binary reaction which is split into two self-limiting chemical reactions in a repeated alternate deposition sequence. Although ALD based on the surface chemical reactions can minimize the structural imperfection in films and substrate damage, the low growth rate is the main drawback for the application of barrier layer. In addition, use of water vapour or ozone in travelling type ALD method and oxygen plasma in PEALD method as the precursor of oxygen, made us hesitate to apply ALD-deposited AlO_x films as the barrier layer for OLED device

Manuscript received June 20, 2004; accepted for publication September 7, 2004. The Korea ministry of Information and Communications financially supported the accomplishment of present work.
* Members, KIDS.
Corresponding Author : Sang-Hee Ko Park
Basic Research Laboratory, Electronics and Telecommunications Research Institute, 161 Gajeong-Dong, Yuseong-Gu, Daejeon, 305-350, Korea.
E-mail : shkp@etri.re.kr **Tel** : +42 860-6276 **Fax** : +42 860-5202

directly.

However, we have observed that just nanometer order of AlO_x single film deposited by ALD technology has excellent barrier properties and is suitable even for the device encapsulation layer. In addition, we have optimized the structure of encapsulation layer for OLED on a plastic substrate, consisting of parylene, PECVD deposited SiN_x, and ALD deposited AlO_x.

2. Experiments

Aluminum oxide thin films with thickness in the range of 10 nm~50 nm were grown on a PES substrate in a 12 × 16 inch large travelling wave ALD reactor with nitrogen as a carrier gas in the temperature range of 80~100°C. Trimethylaluminum (TMA) and H_2O were used as precursors of Al and O, respectively. The sequence of pulses for one cycle deposition of AlO_x is TMA (0.5sec.)/ N_2 (0.8 sec.)/ H_2O (0.5 sec.)/ N_2 (2.5 sec.). Water vapor transmission rates (WVTRs) were measured for both PES substrate and aluminum oxide coated samples on a 50 cm^2 active sample area at 38±2°C, 100 % R.H. using MOCON permatran-W1A for 72 hours. Morphological properties were examined by scanning electron microscope (SEM) and AFM. UV-vis spectra were taken using Hitachi U-3501 spectrophotometer. The device structure of OLEDs grown by vacuum thermal deposition was ITO/ 20 nm MTDATA/ 40 nm NPD/ 60 nm Alq/ 1 nm LiF/ 75 nm Al. Polyethyleneterephthalate (PET) film was used for the substrate of OLED device with emitting area of 2 × 2 mm^2.

3. Results and Discussion

First of all, the dependences of barrier properties of AlO_x films on the deposition temperature and thickness were investigated. With the considerations of low deposition rate of the ALD method and application of encapsulation layer for OLED, we have prepared ultra thin aluminum oxide films of 20~50 nm deposited at lower than 100°C. Table 1 shows barrier properties of uncoated PES film and AlO_x coated one. While most of inorganic single layers deposited by other chemical or physical methods showed proper barrier properties with the thickness higher than 100 nm, single AlO_x films deposited by ALD exhibited good barrier properties of the MOCON values lower than 1.3 g/m^2/day. The barrier property has increased with increasing thickness and elevating deposition temperature and the lowest MOCON value of 0.102 g/m^2/day was obtained from the 50 nm of film deposited at 80°C in our experiment conditions.

One of the main advantages of travelling wave typed ALD is the ability of film coating on both sides of substrate or device simultaneously. The PES film coated both sides

Table 1. The barrier properties of PES substrates coated with AlO_x, SiN_x, parylene or double layer

Deposition Temperature(°C)		Thickness (nm)	WVTR (g/m^2/day @ 38°C)	[a]WVTR (g/m^2/day @ 38°C)
uncoated PES			[b]92.8	
AlO_x	80	20	1.25	
		30	0.409	0.0615
		40	0.141	
		50	0.102	
	90	30	0.303	
	100	30	0.214	0.0276
SiN_x	R.T.	200	highest limit	
SiN_x/AlO_x	R.T./80	200/20	0.058	
parylene	R.T.	3μm	[b]24	
parylene/AlO_x	R.T./80	3μm /20	0.199	

[a] WVTR of PES coated both sides with AlO_x
[b] WVTR for 5cm^2 active area

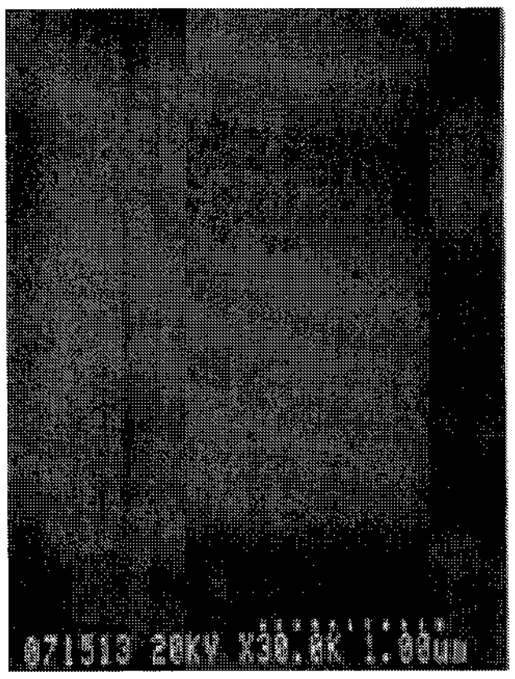


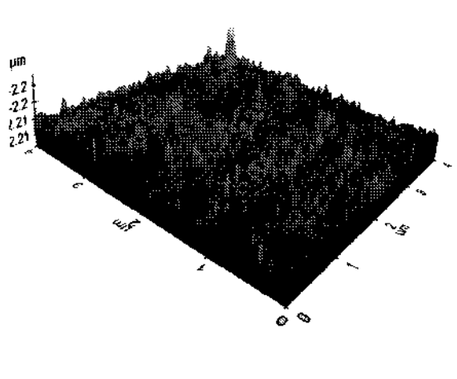
Fig. 1. SEM image of AlO_x thin film deposited @80°C by 500 cycles.

with 30 nm of AlO_x showed 0.0615 g/m^2/day of MOCON value that is, to our knowledge, the lowest value for single inorganic layer with the thickness less than 100 nm. We believe that both sides coating can also reduce the film stress caused by the difference of coefficients of thermal expansions between the plastic substrate and the dielectric film.

The formation of conformal film by ALD resulted in coverage of pin-holes in inorganic or organic layer beneath. Deposition of 30 nm of AlO_x on 200 nm of SiN_x thin film, having MOCON value as high as upper limitation with 50 cm^2 active area, gave the MOCON value lower than the detection limit of MOCON measurement. This indicates the defects decoupling effect of ALD deposited AlO_x film. Superior step coverage property of ALD deposited AlO_x film can even be applied to the coverage of negative angle sloped separator used in the passive matrix OLED panel.

The surface SEM image of AlO_x film on a PES film is shown in Fig. 1. The amorphous aluminum oxide thin films showed flat morphology and also good adhesion to the plastic substrate. When thin films are used for the device packaging, it would be very important for films to have good adhesion to different materials such as metal, organic materials, and substrate. Fig. 2 showed atomic force microscopy (AFM) of AlO_x coated PES substrate. The RMS surface roughness of the 50 nm thick AlO_x film was 3.38 Å while that of uncoated PES film was 4.06 Å. The transmittance of the AlO_x coated film for the whole visible light range is more than 85 % with the air reference and higher value than that of bare PES film with the reference of PES film as shown in Fig. 3. The high transparency of barrier film is suitable for the top emitting OLED encapsulation.

For the application of device encapsulation, we tested ALD deposited AlO_x film as an encapsulation layer



(a) (b)

Fig. 2. AFM images of (a) bare PES and (b) 50 nm AlO_x coated PES.

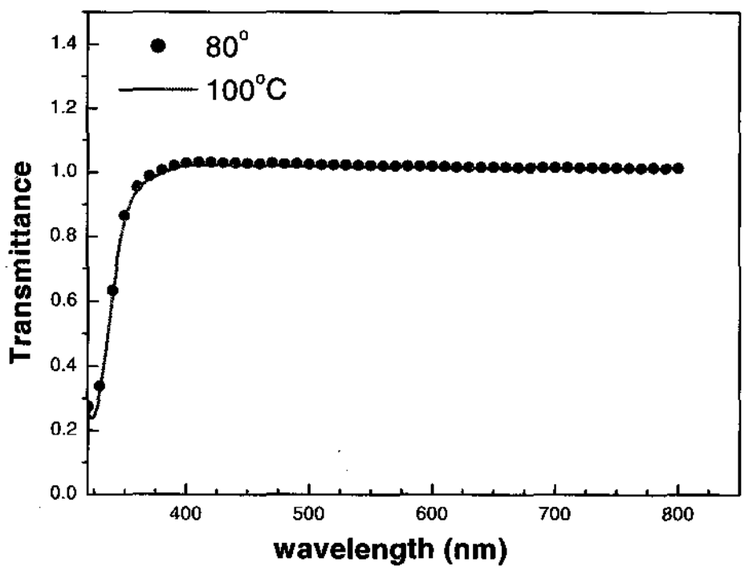


Fig. 3. Transmittance of 50 nm AlO_x coated PES film for the visible light range with the PES substrate reference.

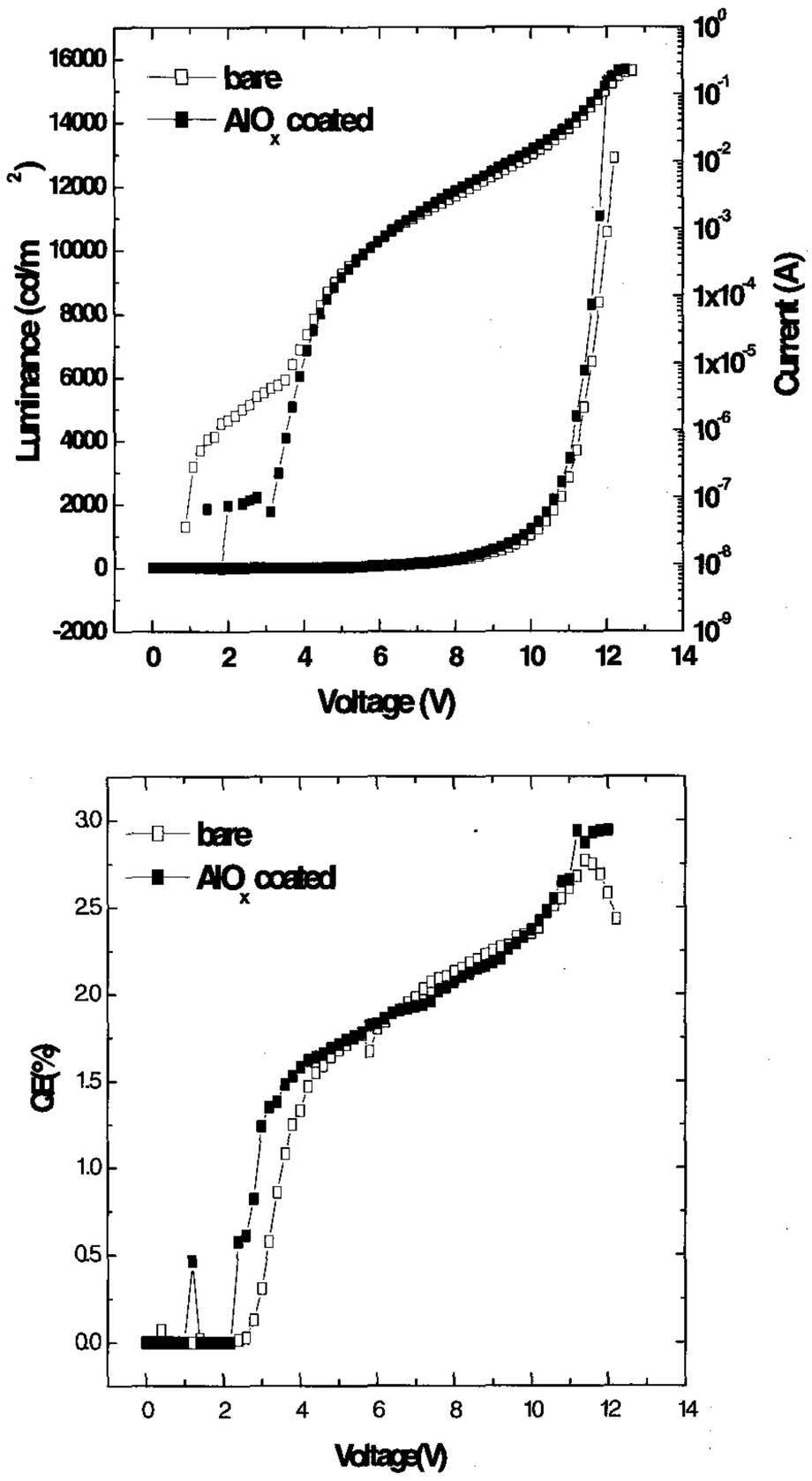


Fig. 4. I-V-L and efficiency characteristics of uncoated OLED and 1 mm parylene/30 nm AlO_x coated OLED on a plastic substrate.

of OLED on a plastic substrate of PET film, having good chemical stability. The OLED was prepared by the vacuum evaporation and 30 nm thick AlO_x were deposited continuously in a separated reactor at 80°C. Since the oxygen precursor, water, is the severe source of device damage, we have tried to use the minimum amount of water. The damage of OLED device caused by precursors could be minimized compared to other plasma enhanced deposition methods such as PECVD or sputtering because precursors including water vapour, oxygen plasma, or TMA are pulsed for just less than 0.5 second for each cycle even in a reactor size larger than 12".

Although we have tired to minimize the device damage during the deposition, the direct deposition of AlO_x film caused formation of several dark spots with the device fabricated on the plastic substrate. We tried to prevent device damage by deposition of organic polymeric buffer layer, parylene, between the device and the inorganic encapsulation layer. One micrometer thick parylene and 30 nm thick AlO_x were deposited continuously in a separated reactor at R.T. and 80°C, respectively and we could succeed in obtaining better OLED characteristics optically and electrically. Fig. 4 shows I-V-L curves and efficiency of OLED on PET substrate encapsulated with parylene and AlO_x films. The parylene/AlO_x coated device showed no big changes in I-V-L and efficiency compared to those of the uncoated device.

We have also put PECVD deposited SiN_x film between the parylene and the AlO_x film to minimize water vapour permeation during the AlO_x deposition. Furthermore, we expected adapting inorganic bi-layer system for the barrier of OLED on PET substrate would result in better encapsulation performance, because ultra thin ALD deposited film showed pin-hole decoupling effect as the micro-order thick organic film does. The SiN_x film was deposited on parylene coated OLED on a PET at room temperature under the plasma power of 25W with the gas ratio of N_2(400 sccm)/SiH_4(4 sccm) /NH_3(26 sccm) /H_2(100 sccm)/He(300 sccm). Both sides aluminum oxide coating was followed by 300 cycles at 80°C. When both sides of the device are coated simultaneously, permeation of water vapour and oxygen from the back side of the plastic substrate can be prevented effectively. Even though the SiN_x film deposited at room temperature showed poor barrier property due to the pin-holes, additional deposition of AlO_x by ALD method enhanced the film quality greatly.

For the comparison of life time, 30 nm thick AlO_x film was simultaneously deposited at front and back sides of the 1μm parylene coated device. Fig. 5 shows luminance decay curves of OLED devices packaged by parylene/SiN_x/AlO_x, parylene/AlO_x, and without encapsulation at constant driving current of 1mA and initial luminance of 1300 cd/m^2 in air at R.T. While the life time of uncoated device is less than 30 hours, 1 μm-parylene/30 nm-AlO_x coated OLED device maintains 80 % of initial luminance for 115 hours, and parylene/SiN_x (100 nm)/AlO_x (30 nm) coated OLED to 90 % of initial luminance (1300 cd/m^2) was 260 hours. Degradation of OLED was mostly due to the formation of new dark spots and growth of the existed dark spot as shown in Fig. 6.

The use of ALD deposited AlO_x thin film for the barrier layer can not only reduce the total thickness of barrier layer also increase the life time of plastic OLED

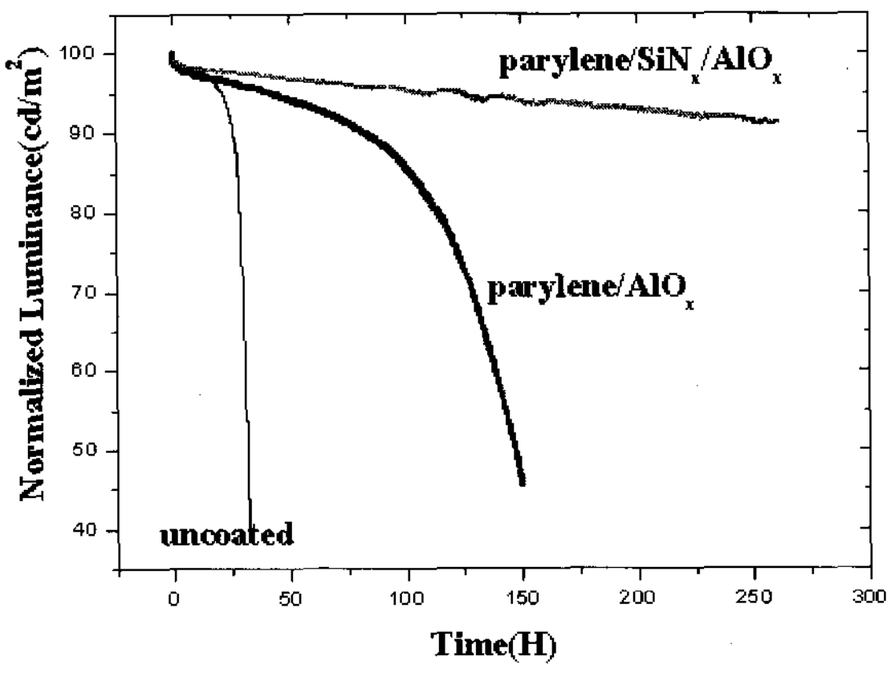


Fig. 5. Luminance decay curves of OLED devices on a PET packaged by parylene/SiN_x/AlO_x, parylene/AlO_x, and without encapsulation at constant driving current of 1mA and initial luminance of 1300 cd/m^2.

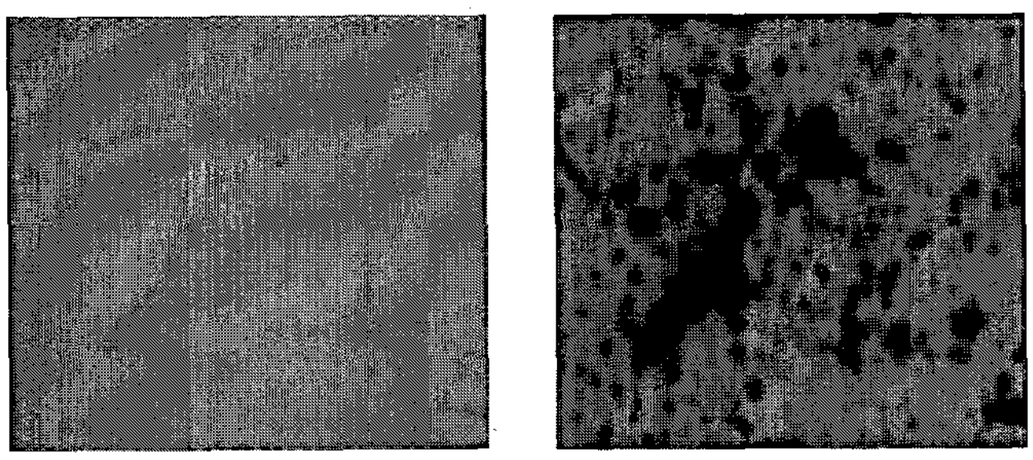


Fig. 6. CCD images of a light emitting area of the parylene/AlO_x coated OLED on a PET before and after the driving at 1mA to give 50 % reduction of first luminance under 24 °C, 45 % R.H.

device significantly.

4. Conclusions

The encapsulation of OLED device on a plastic substrate with the films deposited by the ALD method has been carried out for the first time. Since AlO_x films with even 30 nm-thickness shows excellent barrier properties, it is considered that ALD technology is suitable for the barrier layer fabrication. Due to the effect for decoupling of defect, introduction of ALD deposited AlO_x film can significantly enhance barrier properties for both a substrate and an OLED device on a plastic with ultra thin thickness.

References

[1] B. Comiskey, J. D. Albert, H. Yoshizawa, and J. Jacobson, *Nature*, 394, 253 (1998).

[2] E. Lueder, *SPIE*, 3297, 64 (1998).

[3] K. R. Sarma, C. Chanley, S. Dodd, J. Roush, J. Schmidt, G. Srdanov, M. Stevenson, R. Wessel, J. Innocenzo, G. Yu, M, O'Regan, W. A. MacDonald, R. Eveson, K. Long, H. Gleskova, S. Wagner, and J. C. Sturm, in *Proc. of SPIE* (2003), p. 180.

[4] R. H. Friend, R. W. Gymer, A. B. Holmes, J. H. Burroughes, R. N. Marks, C. Taliani, D. D. C. Bradley, D. A. Dos Santos, J. L. Bredas, M. Logdlund, and W. R. Salaneck, *Nature*, 397, 121 (1999).

[5] M. Gross, D. C. Muller, H.-G. Nothofer, U. Scherf, D. Neher, C. Brauchle, and K. Merholz, *Nature*, 405, 661 (2000).

[6] K. Yamashita, T. Mori, and T. Mizutani, J. Phys. D: Appl. Phys., 34, 740 (2001).

[7] P. E. Burrows, G. L. Graff, M. E. Gross, P. M. Martin, M. K. Shi, M. Hall, E. Mast, C. Bonham, W. Bennet, and M. B. Sullivan, *Displays*, 22, 65 (2001).

[8] M. S. Weaver, L. A. Michaiski, K. Rajan, M. A. Rothman, J. A. Silvernail, J. J. Brown, P. E. Burrows, G. L. Graff, M. E. Gross, P. M. Martin, M. Hall, E. Mast, C. Bonham, W. Bennett, and M. Zumhoff, *Appl. Phys. Lett.*, 81, 2929 (2002).

[9] A. Yoshida, S. Fujimura, T. Miyake, T. Yoshizawa, H. Ochi, A. Sugimoto, H. Kubota, T. Miyadera, S. Ishizuka, M. Tsuchida, and H. Nakada, in *Proc. of the Society for Information Display* (2003), p. 856.

[10] A. B. Chwang, M. A. Rothman, S. Y. Mao, R. H. Hewitt, M. S. Weaver, J. A. Silvernail, K. Rajan, M. Hack, J. J. Brown, X. Chu, L. Moro, T. Krajewski, and N. Rutherford, *Appl. Phys. Lett.*, 83, 413 (2003).

[11] H. Lifka, H. A. van Esch, and J. J. W. M. Rosink, in *Proc. Society for Information Display* (2004), p. 1384.

[12] S.-H. K. Park, J.-I. Lee, Y. S. Yang, and S. J. Yun, in *Proc. 2nd International Meetings on Information Display* (2002), p. 746.

[13] S.-H. K. Park, G. H. Kim, and H. Y. Chu, in *Proc. 4th International Conference on Electroluminescence of Molecular Materials and Related Phenomena* (2003), p. 109.