

# DC Sputtered Indium-Tin Oxide Transparent Cathode for Organic Light-Emitting Diode

Haiying Chen, Chengfeng Qiu, Man Wong, and Hoi Sing Kwok

**Abstract**—The performance of top-emitting organic light-emitting diodes depends not just on the choice of the transparent cathodes but also on their techniques of formation. Compared to the damage induced by radio frequency sputtering of indium-tin oxide cathode, that induced by dc sputtering was verified to be less severe and relatively independent of the sputtering power. Consequently, a high dc sputtering power of 120 W could be employed to achieve a high deposition rate of 0.1 nm/s. Adequate emission efficiency was maintained, even with a relatively thin 7-nm copper (II) phthalocyanine buffer layer.

**Index Terms**—Indium-tin oxide, organic light-emitting diodes, sputtering, top-emission, transparent cathode.

## I. INTRODUCTION

ORGANIC light-emitting diodes (OLED) as pixels for flat-panel displays are being actively pursued because of their relatively simple structure, high brightness, and self-emitting nature [1]–[3]. A top “through-the-cathode” emitting diode is of considerable interest, particularly for displays fabricated on opaque substrates [4] and high-resolution displays made of vertically stacked full-color pixels [5].

Indium-tin oxide (ITO) is a natural candidate for realizing a transparent electrode. It has been pointed out that a composite layer consisting of sputtered ITO and “damaged” copper (II) phthalocyanine (CuPc) is an acceptable electron-injecting electrode [6]. For a given CuPc thickness, the sputtering power must be high enough to produce a sufficient population of defect states in CuPc to allow easy electron injection from ITO, yet low enough to minimize any damage to the underlying emission layer. For a given sputtering power, the CuPc must be thin enough to minimize any additional electrical resistance or absorption of the emission, yet thick enough to absorb all sputtering-induced damage. Conventionally, RF sputtering at power levels of no more than 5 W [2]–[4] has been used to form ITO cathodes on CuPc-buffered OLEDs based on tris-8-hydroxyquinoline aluminum ( $\text{Alq}_3$ ). Though low-power RF sputtering has been used to reduce damage to the underlying organic layers, the resulting deposition rate is exceedingly low. Values of 0.03 nm/s [5] and 0.06 nm/s at 50 W [7] have been reported. At 5 W, the required thickness of CuPc buffer layer is 6 nm [6] but the deposition rate is a mere 0.005 nm/s [4].

Manuscript received January 6, 2003; revised January 30, 2003. The work was supported by the Research Grants Council, Hong Kong Special Administrative Region. The review of this letter was arranged by Editor T.-J. King.

The authors are with the Centre for Display Research, Department of Electrical and Electronic Engineering, The Hong Kong University of Science and Technology, Kowloon, Hong Kong.

Digital Object Identifier 10.1109/LED.2003.812550

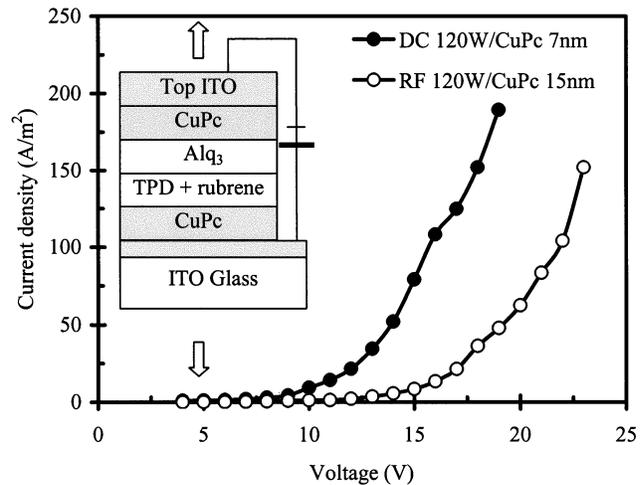


Fig. 1. Current–voltage characteristics of optimized top-emitting OLEDs with dc and RF sputtered top ITO. Shown in the inset is the diode configuration.

In the present work, the performance of top-emitting OLEDs with dc sputtered transparent ITO cathodes is studied. A deposition rate as high as 0.1 nm/s was obtained using 120-W sputtering power. Compared to RF sputtering, the damage induced by dc sputtering was verified to be less severe and relatively independent of the sputtering power. Consequently, it was possible to use a thinner CuPc buffer layer in an OLED with a high-power dc sputtered ITO cathode.

## II. SAMPLE PREPARATION

Commercial ITO-coated glass, with a sheet resistance of 20 ~ 30  $\Omega$ , was used as the starting substrate. After glass precleaning [8], the constituent organic layers of the OLEDs were deposited by thermal vacuum evaporation of commercial grade CuPc  $\text{Alq}_3$  and N,N'-diphenyl-N,N'-bis(3-methylphenyl-1,1'-biphenyl-4,4'-diamine (TPD) mixed with 5 vol% 5,6,11,12-tetraphenylnaphthacene (rubrene). The deposition rates of the organic thin films were 0.2–0.4 nm/s at  $\sim 1$   $\mu$ torr base pressure. The film thickness and the deposition rates were determined *in situ* using a quartz crystal monitor.

A four-layer organic stack consisting of CuPc(20 nm)/TPD:rubrene(40 nm)/ $\text{Alq}_3$ (50 nm)/CuPc was formed, as shown in the Inset of Fig. 1. The TPD:rubrene layer was the hole-transport and emission layer. No emission from the electron-transporting  $\text{Alq}_3$  layer was observed. The effects of varying the thickness of the final electron-injecting CuPc buffer layer were studied.

After the formation of the organic layers, the samples were transferred in air to an AJA International Model ATC1200H

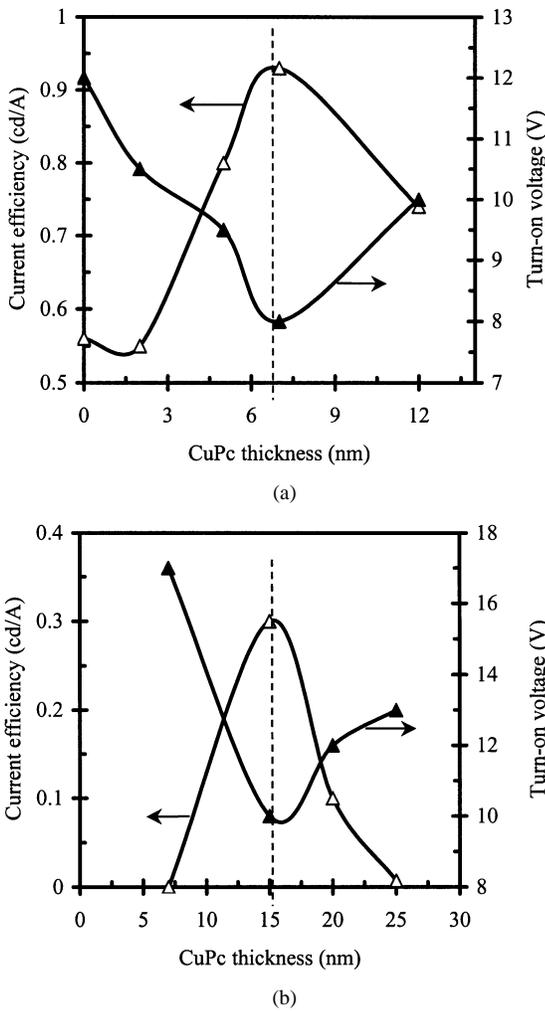


Fig. 2. Dependence of the current efficiency and turn-on voltage on CuPc thickness of OLEDs with 120 W (a) dc and (b) RF sputtered top ITO. Corresponding optimal thickness of CuPc is indicated by a dashed line.

sputtering chamber. A 99.99% bulk density ITO target, with an indium:tin ratio of 9:1 was used. Sputtering was carried out at a pressure of 10 mtorr in argon mixed with 1% oxygen. A cathode area of 12 mm<sup>2</sup> was defined through a shadow mask. The ITO thickness, ~ 75 nm, was determined *ex situ* using a surface profilometer. The deposition rates were ~ 0.01 and ~ 0.02 nm/s for dc power levels of 5 and 20 W, respectively. At 120 W, a high rate of ~ 0.1 nm/s was achieved.

### III. RESULTS AND DISCUSSION

For a 120-W dc sputtered ITO cathode, the dependence of the OLED performance on the thickness of CuPc is shown in Fig. 2(a). The “turn-on voltage” was defined to be the minimum voltage at which electroluminescence was visually detected. With increasing CuPc thickness between 2 and 7 nm, damage to the Alq<sub>3</sub> layer is progressively reduced and increasingly confined to within the CuPc layer. This results in a reduced turn-on voltage because of improved electron mobility in Alq<sub>3</sub>. A further increase of the CuPc layer beyond 7 nm leads to an apparent increase in the turn-on voltage, due to increased absorption of rubrene emission by CuPc [9]. The corresponding

TABLE I  
COMPARISON OF 120-W dc AND RF SPUTTERED ITO AND THE PERFORMANCE OF THE CORRESPONDING OLEDs WITH OPTIMIZED CuPc THICKNESS. TRANSMISSION WAS MEASURED AT THE PEAK RUBRENE EMISSION WAVELENGTH OF 565 NM

		DC Sputtered ITO	RF Sputtered ITO
ITO properties	Sheet resistance $\Omega/\square$	120	150
	Transmittance at 565nm %	85	85
	Deposition rate nm/s	0.1	0.06
OLED performance	Optimized CuPc thickness nm	7	15
	Turn-on voltage V	8	14
	Power efficiency lm/W	0.28	0.08
	Current efficiency cd/A	0.9	0.3

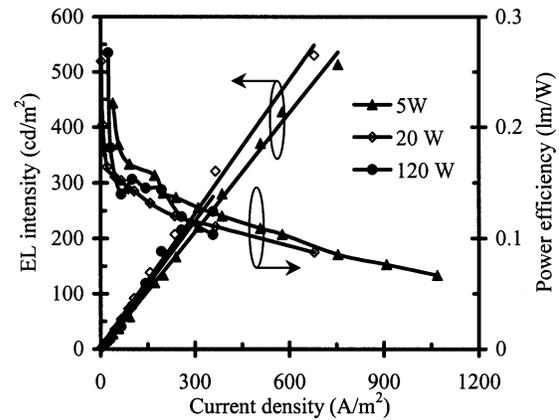


Fig. 3. Dependence of the EL intensity and power efficiency on the operating current density of OLEDs with dc sputtered top ITO. Note the relative independence on the sputtering power.

increase in the driving voltage for a given current level was determined to be relatively minor because of the relatively high electron mobility in CuPc [10]. Similar trends for the turn-on voltage and power efficiency were observed for OLEDs with 120-W RF sputtered ITO cathode [Fig. 2(b)]. The optimized CuPc thickness was 15 nm, more than double that for an OLED with dc sputtered ITO. The current–voltage characteristics of optimized OLEDs with 120-W dc and RF sputtered ITO are compared in Fig. 1 and Table I. The sheet resistance of the ITO was measured on films with equivalent thickness deposited on glass.

Shown in Fig. 3, the luminous current (0.9 cd/A) and luminance power (0.28 lm/W) efficiencies of OLEDs with dc sputtered ITO cathode, are relatively independent of the sputtering power. This is quite different from the RF case. With 6-nm thick CuPc buffer layer, the emission layer is protected only at low RF power of 5 W but severely damaged at a higher power of 50 W [4], [5].

The relative independence of device efficiencies on dc sputtering power may be traced to the physical process of dc sputtering. During sputtering, any sputtered species have to pass through the plasma before reaching the substrate. At a pressure of 10 mtorr, the mean-free path of ~ 1 cm, is significantly shorter than the 20-cm substrate-to-target distance in the ATC1200H system. Therefore, the kinetic energy of sputtered species upon leaving the target is mostly transferred to the plasma by collisions. It is the voltage drop across the anode

sheath that determines the final kinetic energy of and the degree of damage imparted on the organic layer by the ionized species extracted from the plasma. For a dc gas discharge, the anode sheath voltage is relatively insensitive to and increases only logarithmically with the dc sputtering power density [11]

$$V_p - V_{an} = a + b \ln W$$

where  $V_p$  and  $V_{an}$  are voltages of the plasma and anode, respectively.  $W$  is the power density,  $a$  and  $b$  are the parameters related to ion impingement rate and electron temperature, respectively.

In the case of RF sputtering, power affects in a complex manner both the current impinging on the substrate and the ion energy. In 120-W RF plasma, the voltage drop across the anode sheath can be as high as 150 V [12], while in a 120-W dc plasma, it is only in the range of 10 V [11]. The higher sheath voltage leads to correspondingly higher ion energies in an RF plasma [13], hence induces more severe damage. Consequently, a thicker CuPc is required to protect the Alq<sub>3</sub> from RF than dc sputtering damage, at equivalent power settings.

#### IV. CONCLUSION

DC sputtering has been employed to deposit transparent indium-tin oxide cathodes for top-emitting organic light-emitting diodes. It is shown that the emission efficiencies are relatively independent of the sputtering power. This allows 120-W high-power sputtering, leading to a relatively high deposition rate of 0.1 nm/s. The optimized thickness of the necessary copper (II) phthalocyanine buffer layer was determined to be 7 nm. By comparison, it is 15 nm for 120-W RF sputtered indium-tin oxide.

#### REFERENCES

- [1] C. W. Tang and S. A. VanSlyke, "Organic electroluminescent diodes," *Appl. Phys. Lett.*, vol. 51, no. 12, pp. 913–915, 1987.
- [2] Y. Kijima, N. Asai, N. Kishii, and S. Tamura, "RGB luminescence from passive-matrix organic LED'S," *IEEE Trans. Electron Devices*, vol. 44, pp. 1222–1228, Aug. 1997.
- [3] G. Gu, V. Bulovic, P. E. Burrows, and S. R. Forrest, "Transparent organic light emitting devices," *Appl. Phys. Lett.*, vol. 68, no. 19, pp. 2606–2608, 1996.
- [4] S. R. Forrest, P. E. Burrows, Z. Shen, G. Gu, V. Bulovic, and M. E. Thompson, "The stacked OLED (SOLED): A new type of organic device for achieving high-resolution full-color displays," *Synth. Metals*, vol. 91, pp. 9–13, 1997.
- [5] G. Parthasarathy, C. Adachi, P. E. Burrows, and S. R. Forrest, "High-efficiency transparent organic light-emitting devices," *Appl. Phys. Lett.*, vol. 76, no. 15, pp. 2128–2130, 2000.
- [6] G. Parthasarathy, P. E. Burrows, V. Khalfin, V. G. Kozlov, and S. R. Forrest, "A metal-free cathode for organic semiconductor devices," *Appl. Phys. Lett.*, vol. 72, no. 17, pp. 2138–2140, 1998.
- [7] L. S. Hung and C. W. Tang, "Interface engineering in preparation of organic surface-emitting diodes," *Appl. Phys. Lett.*, vol. 74, no. 21, pp. 3209–3211, 1999.
- [8] C. Qiu, H. Chen, M. Wong, and H. S. Kwok, "Dependence of the current and power efficiencies of organic light-emitting diode on the thickness of the constituent organic layers," *IEEE Trans. Electron Devices*, vol. 48, pp. 2131–2137, Sept. 2001.
- [9] A. B. Djurišić, C. Y. Kwong, T. W. Lau, W. L. Guo, E. H. Li, Z. T. Liu, H. S. Kwok, L. S. M. Lam, and W. K. Chan, "Optical properties of copper phthalocyanine," *Opt. Commun.*, vol. 205, pp. 155–162, 2002.
- [10] L. S. Hung and M. G. Mason, "Voltage reduction in organic light emitting diodes," *Appl. Phys. Lett.*, vol. 78, pp. 3732–3734, 2002.
- [11] B. Chapman, *Glow Discharge Processes*. New York: Wiley-Interscience, 1980.
- [12] U. Kortshagen and L. D. Tsendin, *Electron Kinetics and Applications of Glow Discharges*. New York: Plenum, 1998.
- [13] M. Bender, J. Trube, and J. Stollenwerk, "Characterization of a RF/DC-magnetron discharge for the sputtering deposition of transparent and highly conductive ITO films," *Appl. Phys. A: Mat. Sci. Process.*, vol. 69, pp. 397–401, 1999.