Effect of thickness of a CuI hole injection layer on the properties of organic light emitting diodes

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Abstract—We report the influence of the thickness of a copper iodide (CuI) hole injection layer (HIL) on the performance of organic light-emitting diodes (OLEDs) with tris-8-hydroxyquinoline aluminum (Alq_3) active layer and aluminum cathode layer. The investigation of structural and morphological properties of thermally evaporated CuI thin films indicates that they are amorphous. OLEDs with the ITO/CuI/Alq_3/poly(ethylene glycol) dimethyl ether/Al structures were fabricated and current density-voltage, luminance-voltage and current efficiency-current density were analyzed. The maximal luminescence current efficiency is obtained for the devices with a CuI thickness of 12nm.

Display technologies that utilize organic light-emitting diodes (OLEDs) were commercialized at the end of the last century and have been continually developed until the present time [1-3]. One active direction of current research is the synthesis and characterization of new hole-injection and electron-injection materials. Hole injection layers (HIL) and electron injection layers (EIL) enhance the luminescence current efficiency of OLEDs [4-6]. For example, polyaniline [7], poly(3,4-ethylene-dioxythiophene) (PEDOT) [8-9], metal phthalocyanines [10] and metals with high work functions [11-12] are successfully used as HILs. These materials decrease the hole injection barrier between the anode and the organic active layer.

Recently we have reported that CuI thin films can be used as effective HILs in the pentacene-based photovoltaic structures [13], as an efficient injection layer of holes from the ITO anode in a light-emitting diode structure based on Alq_3 [14], and solid-state dye-sensitized solar cells [15].

CuI thin films deposited by thermal evaporation are on average 80% transparent over the 400–800nm wavelength range [13]. CuI material has low cost and therefore is promising for practical applications in organic optoelectronic devices.

The aim of this work is to characterize the structural and morphological properties of CuI thin films and to investigate the influence of the thickness of the CuI hole

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injection layer on the performance of organic light-emitting devices.

CuI, Alq_3, poly(ethylene glycol) dimethyl ether (PEGDE) and Al layers for OLEDs were deposited using thermal evaporation of the corresponding powders from molybdenum boats in a vacuum chamber with the base pressure lower than 10^{-3}Pa [14]. The layers were grown on ITO-coated glass substrates (Sigma Aldrich, 100Ω/sq) ultrasonically pre-cleaned using acetone and water. The substrates were held at room temperature during the deposition. The deposition rates of Alq_3, PEGDE [16] and Al were 0.3nm/s, 0.4nm/s and 6nm/s, and the resulting thicknesses were 40nm, 3nm and 150nm, respectively. CuI powders were heated to 300°C which resulted in a deposition rate of 0.3nm/s. The thickness of CuI layers in five reported OLEDs was 0nm, 5nm, 8nm, 12nm, 18nm, and 30nm, as determined from the ellipsometry measurements. The active area of the ITO/CuI/Alq_3/PEGDE/Al devices was 3.3mm[2].

The current density-voltage (J-V) curves of the OLEDs were recorded using an 'L2 – 56' curve tracer [14]. The luminance of the devices was measured with a calibrated Si photodiode. CuI surface morphology was studied in a contact mode using 'Solver 47P – Pro' atomic force microscope (AFM) over the 2.5μm^2 area. X-ray diffraction (XRD) patterns of CuI Kα radiation (λ=0.154nm) were collected using a 'Rigaku Rapid' diffractometer in a grazing incidence configuration with 3° angle.

XRD and AFM studies lead to a conclusion that deposited CuI thin films have an amorphous structure. As shown in Fig. 1, the only diffraction peaks present in the XRD patterns can be indexed to ITO structure [17-18]. Figure 2 shows the surface morphology of a CuI thin film measured using AFM. The CuI surface is smooth, which is consistent with the presence of an amorphous phase. The amorphous structure of a CuI hole injection layer is favorable for OLED applications, because it may enhance the device stability under continuous operation [19-20] and does not cause possible crystallization of the active layer.
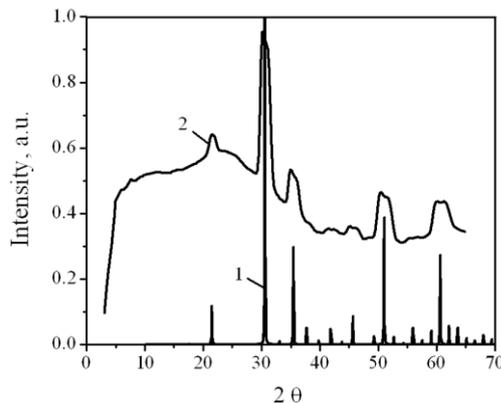
Fig. 1. X-ray diffraction patterns of the ITO/CuI thin film stacks on glass substrates.
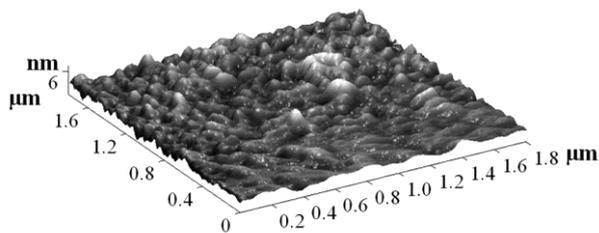
Fig. 2. Atomic force microscopy images of the CuI surface.

The results of the current density-voltage study are shown in Fig. 3. J-V curves of the ITO/CuI/Alq$_3$/PEGDE/Al devices with a different CuI thicknesses were fit using a trap-charge limited current (TCLC) model [1]:

$$J_{TCLC} = N_c \mu q \left(\frac{\varepsilon \varepsilon_0 l}{N_t q(l+1)} \right)^l \left(\frac{2l+1}{l+1} \right)^{l+1} \frac{V^{l+1}}{d^{2l+1}} \qquad (1)$$

where μ is the charge carrier mobility, q is the charge, d is the device thickness, ε is the relative dielectric constant, ε_0 is the vacuum permittivity, N_c is the density of states at the conduction band minimum, N_t is the trap density. In Eq. (1) $l = E_t/k_b T$, where E_t is the characteristic trap energy, T is the operation temperature, and k_b is the Boltzmann's constant.

The results of the TCLC model analysis lead to the conclusion that the CuI layer decreases the number of charge traps and enhances the hole injection properties of OLEDs. The insertion of the CuI layer into the structure leads to a decrease of l from 5.7 to 5.2. We attribute this change to the elimination of interface charge traps at the interface with Alq$_3$ [1]. The CuI layer also improves hole injection properties of the devices. As shown in Fig. 4, this improvement is associated with the decrease of a potential barrier for holes at the Alq$_3$ interface [1].
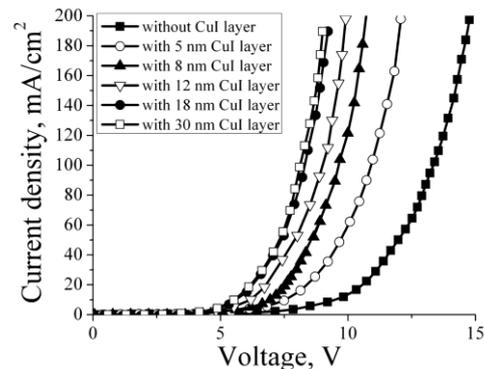
Fig. 3. Current density-voltage curves of the ITO/CuI/Alq$_3$/PEGDE/Al device.
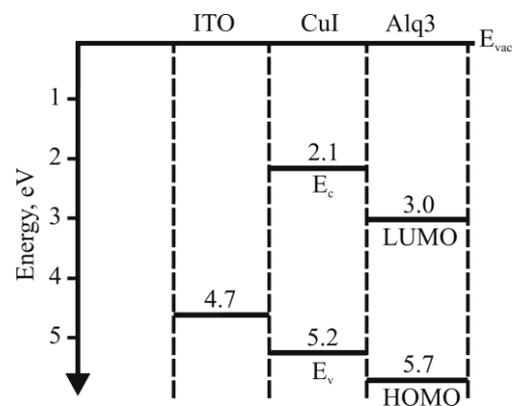
Fig. 4. Energy level diagrams of the ITO/CuI/Alq$_3$/PEGDE/Al structure [14]. Literature values of the ionization potential and electron affinity of ITO, Al, Alq$_3$ [21] and CuI [22] were used.

The efficiency of OLEDs was also improved by introducing of CuI hole injection layers [14]. As shown in Fig. 5, an increase of CuI thickness from 0 to 12nm leads to an increase of luminance from 3000cd/m^2 to 4000cd/m^2 at a current of 200mA/cm^2.
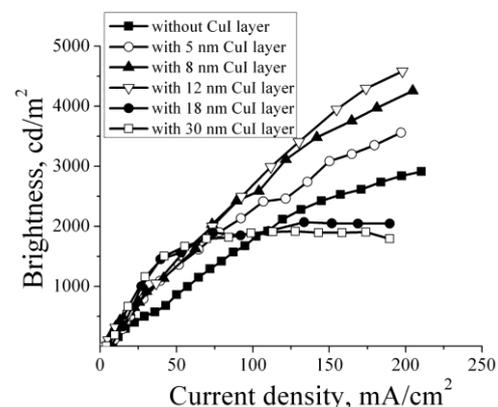
Fig. 5. Brightness of the ITO/CuI/Alq$_3$/PEGDE/Al OLEDs as a function of CuI thickness.

Improving the OLED performance can be explained by the improvement of the electron-hole balance in the active Alq_3 layer. In turn, this increases the distance between the recombination region and the interface with the anode, and decreases non-radiative energy transfer [23]. An increase of CuI thickness beyond 12 nm deteriorates the device performance due to an increase of the series resistance and resulting Joule heating. Therefore, the 12 nm thickness of CuI hole injection layers was found to be optimal for the performance of $ITO/CuI/Alq_3/PEGDE/Al$ organic light-emitting devices.

We found that the CuI thin films prepared by thermal evaporation on substrates held at room temperature are amorphous. We investigated the effect of the thickness of the CuI hole injection layer of electrical and light-emitting properties of the $ITO/CuI/Alq_3/PEGDE/Al$ organic light-emitting devices. J-V curves of the OLEDs were approximated using a trap-charge limited current density model. It was shown that the efficiency of OLEDs can be improved by introducing of CuI hole injection layers up to a thickness of 12 nm. Further increasing of the CuI thickness deteriorates the device performance due to increased series resistance and resulting Joule heating.

References

[1] W. Brütting, S. Berleb, A.G. Mückl, Organic Electronics **2**, 1 (2001).
[2] H. Kubota *et al*., Journ. of Lumin. **87-89**, 56 (2000).
[3] J. Kido, Phys. World **12**, 27 (1999).
[4] M. Stossel, J. Staudigel, F. Steuber, J. Simmer, A. Winnacke, Appl. Phys. A: Mater. Sci. Process **68**, 387 (1999).
[5] T.M. Brown, R.H. Friend, I.S. Millard, P.J. Lacey, J.H. Burroughes, F. Cacialli, Appl. Phys. Lett. **79**, 174 (2001).
[6] B. Choi, H. Yoon, H.H. Lee. Appl. Phys. Lett. **76**, 412 (2000).
[7] K. Fehse, G. Schwartz, K. Walzer, K. Leoa, J. Appl. Phys. **101**, 124509 (2007).
[8] N. Drolet, S. Beaupré, J.F. Morin, Y. Tao, M. Leclerc, J. Optic Pure Appl Optic **4**, S252 (2002).
[9] B.D. Chin, J Phys D: Appl Phys. **41**, 215104 (2008).
[10] S.A. Van Slyke, C.H. Chen, C.W. Tang. Appl. Phys. Lett. **69**, 2160 (1996).
[11] Y. Shen, D.B. Jacobs, G.G. Malliaras, G. Koley, M.G. Spencer, A. Ioannidis, Adv. Mater **13**, 1234 (2001).
[12] C. Qiu, Z. Xie, H. Chen, M. Wong, H.S. Kwok, J. Appl. Phys. **93**, 3253 (2003).
[13] P.Y. Stakhira, V.V. Cherpak, Vacuum **83**, 1129 (2009).
[14] P. Stakhira, V. Cherpak, D. Volynyuk, F. Ivastchyshyn, Z. Hotra, V. Tataryn, G. Luka, Thin Solid Films. **518**, 7016 (2010).
[15] P.M. Sirimanne, T. Soga, M. Kunst, J. Solid. State Chem. **178**, 3010 (2005).
[16] T.H. Lee *et al.*, Adv. Funct. Mater. **18**, 3036 (2008).
[17] T.F. Stoica *et al.*, J. Optoelectron. Adv. Mater. **2**, 684 (2000).
[18] T. Maruyama, K. Fukui, J. Appl. Phys. **70**, 3848 (1991).
[19] Y. Shirota *et al.*, Appl. Phys. Lett. **65**, 807 (1994).
[20] Ch. Jonda, A.B.R. Mayer, U. Stolz, A. Elschner, A. Karbach, J. Mater. Sci. **35**, 5645 (2000).
[21] A.V. Kukhto, J. Appl. Spectrosc. **70**, 165 (2003).
[22] P.M. Sirimanne, M. Rusop, T. Shirata, T. Soga, T. Jimbo, Chem. Phys. Lett. **366**, 485 (2002).
[23] J. Shinar, *Organic light-emitting devices: a survey* (New York, Springer 2004).