Office de la Propriété Intellectuelle du Canada
Un organisme d'Industrie Canada

Canadian Intellectual Property Office
An agency of Industry Canada

CA 2412379 A1 2004/05/22

(21) 2 412 379

(12) DEMANDE DE BREVET CANADIEN
CANADIAN PATENT APPLICATION

(13) A1

(22) Date de dépôt/Filing Date: 2002/11/22

(41) Mise à la disp. pub./Open to Public Insp.: 2004/05/22

(51) Cl.Int.⁷/Int.Cl.⁷ H01L 33/00

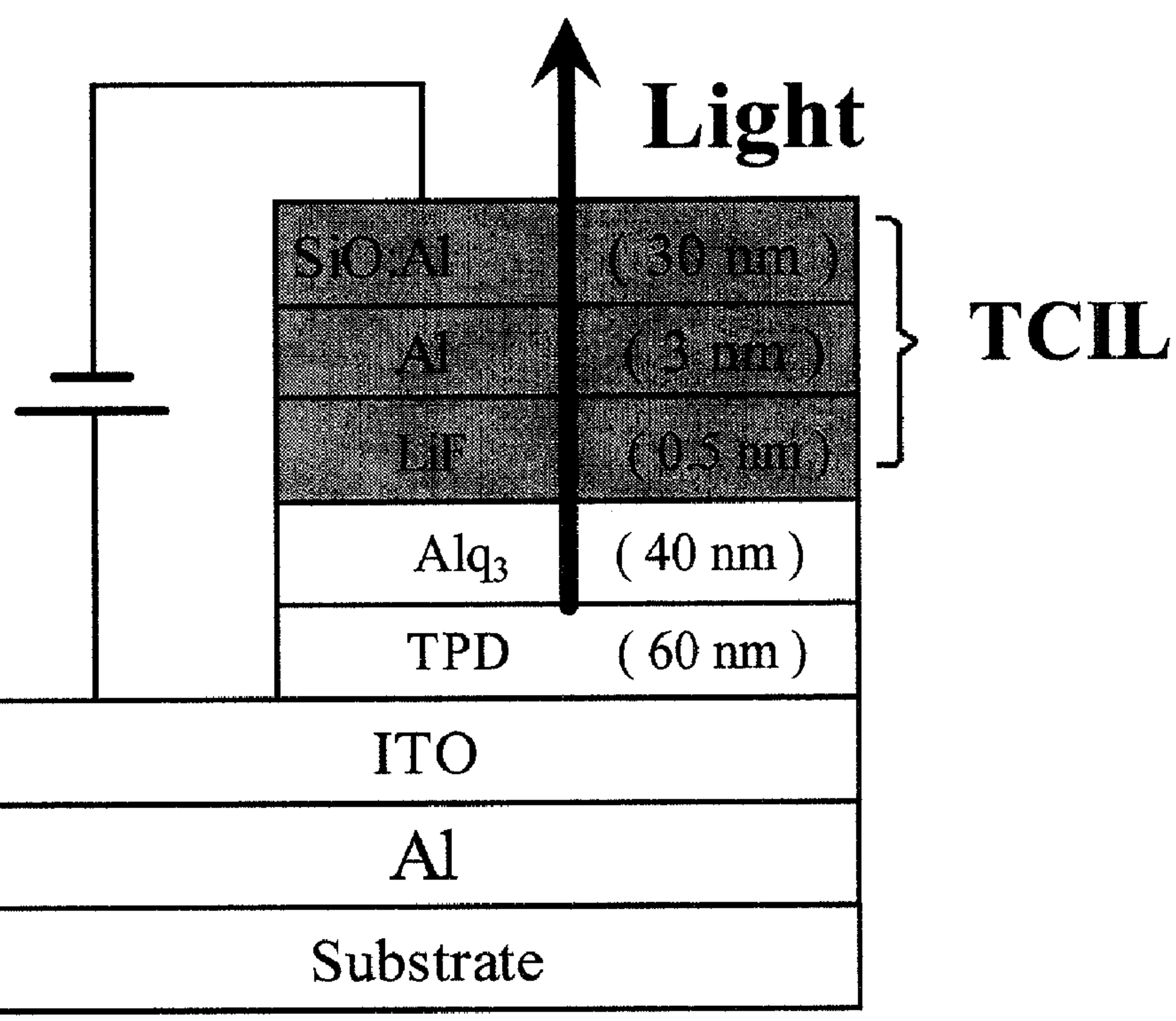
(71) Demandeur/Applicant:
LUXELL TECHNOLGIES INC., CA

(72) Inventeurs/Inventors:
HAN, SIJIN, CA;
FENG, XIADONG, CA;
LU, ZHENGHONG, CA;
WOOD, RICHARD P., CA;
JOHNSON, DAVID J., CA

(74) Agent: LANG MICHENER LLP

(54) Titre : CATHODE TRANSPARENTE POUR DIODES ELECTROLUMINESCENTES A EMISSION PAR LE HAUT

(54) Title: TRANSPARENT-CATHODE FOR TOP-EMISSION ORGANIC LIGHT-EMITTING DIODES



(57) Abrégé/Abstract:
A new transparent-charge-injection-layer consisting of LiF/Al/Al-doped-SiO has been developed as (i) a cathode for top emitting organic light-emitting diodes (TOLEDs) and as (ii) a buffer layer against damages induced by energetic ions generated during deposition of other functional thin films by sputtering, or plasma-enhanced chemical vapor deposition. A luminance of 1900 cd/m^2 and a current efficiency of 4 cd/A have been achieved in a simple testing device structure of ITO/TPD (60 nm)/Alq_3 (40 nm)/LiF (0.5 nm)/Al(3 nm)/Al-doped-SiO (30 nm). A thickness of 30 nm of Al-doped SiO is also found to protect organic layers from ITO sputtering damage.

Abstract

A new transparent-charge-injection-layer consisting of LiF/Al/Al-doped-SiO has been developed as (i) a cathode for top emitting organic light-emitting diodes (TOLEDs) and as (ii) a buffer layer against damages induced by energetic ions generated during deposition of other functional thin films by sputtering, or plasma-enhanced chemical vapor deposition. A luminance of 1900 cd/m^2 and a current efficiency of 4 cd/A have been achieved in a simple testing device structure of ITO/TPD (60 nm)/Alq_3 (40 nm)/LiF (0.5 nm)/Al (3 nm)/Al-doped-SiO (30 nm). A thickness of 30 nm of Al-doped SiO is also found to protect organic layers from ITO sputtering damage.

Transparent-cathode for top-emission organic light-emitting diodes

Background of the Invention

Top-emitting organic light-emitting diodes (TOLEDs), unlike conventional ones that emit light through a transparent bottom electrode (ITO) and glass substrate, is of great importance for the integration of OLED devices with electrical drivers. Top emission is desirable for active-matrix OLED displays because all circuitry can be placed at the bottom without any interference from components such as wiring and transistors. TOLEDs are eminently suitable for making microdisplays because of the high level of integration of necessary driver circuits with the matrix structure of OLEDs on a silicon chip. Therefore, design and fabrication of this top transparent cathode is an enabling technology for high-end OLED displays.

Intensive studies on the conventional OLEDs have been well documented. However, there is limited information on the fabrication of TOLED devices. The use of rf sputtered ITO as a top transparent electrode with a buffer layer such as MgAg,[1,2] phthalocyanine (CuPc)[3,4] or 3,4,9,10-perlyenetetracarboxylic dianhydride (PTCDA)[5] films have been reported. However, the energetic ion damages to the underlying organic layer induced by sputtering, as we will discuss later, is a major problem and the yield of devices remains a great concern. It is thus believed that the only possible cathode deposition method has to be based on thermal evaporation.[6] But there is no report on TOLED cathode fabricated based solely on thermal evaporation.

Summary of the Invention

It is therefore an object of the present invention to provide a novel transparent-cathode for top emission OLEDs that obviates or mitigates at least one of the above-identified disadvantages of the prior art. In an aspect of the invention, there is provided a material system, which consists of LiF/Al/Al-doped SiO multilayers, for use as a (a) top electrode and (b) buffer layer against radiation damages for rf sputter deposition of other active and passive over layers.

A new transparent-charge-injection-layer consisting of LiF/Al/Al-doped-SiO has been developed as (i) a cathode for top emitting organic light-emitting diodes (TOLEDs) and as (ii) a buffer layer against damages induced by energetic ions generated during deposition of other functional thin films by sputtering, or plasma-enhanced chemical vapor deposition. A luminance of 1900 cd/m^2 and a current efficiency of 4 cd/A have been achieved in a simple testing device structure of ITO/TPD (60 nm)/Alq_3 (40 nm)/LiF (0.5 nm)/Al (3 nm)/Al-doped-SiO (30 nm). A thickness of 30 nm of Al-doped SiO is also found to protect organic layers from ITO sputtering damage.

Brief Description of the Drawings

Preferred embodiments of the present invention will now be explained, by way of example only, with reference to the attached Figures in which:

Figure 1 is a schematic cross-sectional diagram of a top-emitting OLED structure in accordance with an embodiment of the invention;

Figure 2 is a graph showing Luminance (L)-current density (J)-voltage (V) of (a) OLED and (b) TOLED;

Figure 3 is a graph showing efficiencies of OLED and TOLED; and

Figure 4 depicts electroluminescent spetra of the devices with different thickness of ITO.

Detailed Description of the Invention

Referring now to Figure 1, a cross-sectional diagram of a top-emitting OLED device in accordance with an embodiment of the invention is shown. Devices according to this embodiment were fabricated using a Kurt J. Lesker OLED cluster-tools for 4"x4" substrate. The cluster-tools include a central distribution chamber, a loadlock chamber, a plasma treatment chamber, a sputtering chamber, an organic deposition chamber, and a metallization chamber. N,N'-diphenyl-N,N'-bis(3-methylphenyl)-1,1'-biphenyl-4,4'-diamine (TPD) and tris-(8-hydroxyquinoline) aluminum (Alq_3) were used as a hole transport layer (HTL) and electron transport layer (ETL), respectively. Both OLED and TOLED devices were fabricated on 2"x2" substrates. The device structure of OLED is ITO/TPD/Alq_3/LiF/Al. The schematic structure of TOLED was shown in Fig. 1. After the substrate was treated by oxygen plasma for 10 mins in the plasma chamber, it was transferred to the sputtering chamber where ~50 nm of ITO was deposited by rf sputtering at a power of 45 W and an argon pressure of 8.5 mTorr. A grid shadow mask was used to define ITO anode structures. The sheet resistance of ITO is ~300 Ω/ . TPD, Alq_3, LiF, and Al were sequentially deposited by thermal evaporation in the organic and metallization chambers. Al-doped SiO (Al:SiO) films were deposited through a second shadow mask by co-evaporation of Al and SiO. Additional ITO layers were sputtered onto the Al:SiO on some devices to evaluate its robustness against sputter damages. The devices were finally encapsulated with 100 nm thick of SiO film by thermal evaporation. Luminance-current-voltage (L-I-V) characteristics of the devices were measured using a HP 4140B pA meter and a Minolta LS-110 meter.

Table I summarized the test results on various TOLEDs and OLEDs. The sputtering damages were characterized from the performance of the LEDs and the yield of pixels. The poor yields seen in Table I indicate that sputtering damage is still a serious issue. CuPc films are insufficient to prevent the bombardment of ions in the organic layer during the sputtering at a power of 40 W. Although the damage is somewhat reduced when the rf-power is lowered to 15 W, the few surviving TOLEDs have very low luminance. Regular OLEDs have been fabricated with Al and Al/sputtered ITO cathodes and the results are shown in fourth and fifth rows of Table I. The data show that the performance of device with the structure of Al(30nm)/ITO as the cathode is not as good as the one with Al only. Here the rf condition has been reduced to 8 W at 8.0 mTorr, which resulted in a very slow deposition rate at 0.036 Å/s. The OLED results also suggest that an inorganic buffer layer with a thickness more than 300 Å is desired to reduce the sputtering damages. All metal films of this thickness are optically opaque. It can greatly reduce the light output if a thick metal film is used as a buffer layer for sputtering of ITO.

Fig. 2. shows the L-I-V curves of the fourth (OLED) and sixth device (TOLED). The performance of the conventional OLEDs fabricated using our organic cluster tool is similar to those reported in recent literature.[7,8] At 13.6 V, the luminance of TOLED reaches 100 cd/cm^2, which is a typical minimum requirement for video display. We also obtained 1900 cd/cm^2 at a current density of 922 mA/cm^2. The current efficiency and luminous power efficiency vs voltage were shown in Fig. 3. It is found that current efficiency of TOLED is better than that of OLED, while the power efficiency shows the reverse trend. Several factors contribute to this difference. First, the sputtered ITO anode for TOLED has a much higher resistivity than that of the commercial ITO anode used for

OLED. Second, the Al:SiO cathode for TOLED also has a much higher resistivity than that of Al cathode used for OLED. The overall performance of TOLED is not as good as that of OLED. However, those TOLED performance data shown in Figs. 2 and 3 represents the best when compared with recent published results,[2] The TOLEDs here were fabricated only by thermal evaporation. We have not been able to reproduce TOLED results with sputtered ITO cathode with similar buffer layer structures as described in those literatures.

One interesting phenomena observed in our current TOLED devices is that the EL peak position or color varies significantly depending strongly on ITO thickness. Fig. 4 shows the typical EL spectra (with peak high normalized) recorded on TOLED with ITO thickness of 10, 20 and 50 nm, respectively as labeled. Since those devices are fabricated on the same substrate, with the organic films and top cathode deposited under the identical conditions, it thus excludes other uncertainty in organic layer thickness variation. It is noted that the EL peak position shifts to longer wavelength as the ITO layer thickness is increased. This shift may be attributed to multiple factors including optical microcavity and surface plasmons cross coupling. Dodabalapur et al.[9,10] and Bulovic et al.[11] have reported detailed mechanism of microcavity effects on the optical characteristics in OLEDs. And recently Gifford et al.[12] and Hobson et al.[13] have investigated the role of surface plasmon loss in OLEDs. Our TOLED device is somewhat similar to Gifford's observation. The rough ITO surface of our TOLEDs could play the same role as that of intentionally patterned surface used in Gifford's device. A red-shift was expected when a light beam bounces off a reflective surface with energy loss to excite various surface plasmon modes. It may also explain the rather broad shifted EL spectra, whereas pure microcavity effect would only predict sharp shifted peaks.

In summary, TOLEDs on a silicon substrate have been fabricated using a new cathode consisting of a multilayer stack of LiF/Al/SiO:Al. A luminance of 1900 cd/m^2 at 922 mA/cm^2 and a current efficiency of 4 cd/A were achieved. It has been shown that the new TCIL is fairly robust against the radiation damage, which permits deposition of other active and passive films by sputtering or other aggressive plasma processes such as ECR or PECVD. Our data indicates that the metal-doped SiO film is a promising new materials for use as a transparent electrode in TOLED.

The contents of all references identified herein are incorporated herein by reference.

While only specific combinations of the various features and components of the present invention have been discussed herein, it will be apparent to those of skill in the art that desired sub-sets of the disclosed features and components and/or alternative combinations and variations of these features and components can be utilized, as desired.

Table 1. The performance and yield of LEDs with different cathodes. The sputtering power is 8 W unless mentioned in the table.

Device	**Cathode structures**	**Performance**	**Yield**
TOLED	CuPc(7,14,21 nm)/LiF/ITO (RF power 45 W)	Non-functional	0%
TOLED	CuPc(15 nm)/ITO (RF power 10 W)	<50 cd/m^2 at 20 V	< 25%
OLED	LiF/Al (100 nm)	~5000 cd/m^2 at 6.4 V	100%
OLED	LiF/Al (30 nm)/ITO	~5500 cd/m^2 at 11 V	< 70%
TOLED	LiF/Al (3 nm)/Al:SiO (30 nm)/ITO	~1600 cd/m^2 at 25 V	>90%
TOLED	LiF/Al (3 nm)/Al:SiO (30 nm)	~1590 cd/m^2 at 20 V	>90%

References:

1. G. Gu, V. Bulovic, P. E. Burrows, S. R. Forrest and M. E. Thompson, Appl. Phys. Lett. **68,** 2606 (1996).
2. W. E. Howard and O. F. Prache, IBM J. Res. & Dev. **45,** 115 (2001).
3. V. Bulovic, P. Tian, P. E. Burrows, M. R. Gokhale, S. R. Forrest and M. E. Thompson, Appl. Phys. Lett. **70,** 2954 (1997).
4. L.S. Hung, C.W. Tang, Appl. Phys. Lett. 74, 3209 (1999).
5. G. Parthasarathy, P. E. Burrows, V. Khalfin, V. G. Kozlov, and S. R. Forrest, Appl. Phys. Lett. **72,**2138 (1998).
6. L. S. Hung, C. W. Tang, M. G. Mason, P. Raychaudhuri, and J. Madathil, Appl. Phys. Lett. **78,** 54 (2001).
7. C.F. Qiu, H. Y. Chen, Z. L. Xie, M. Wong, and H. S. Kwok, Appl. Phys. Lett. **80,** 3485 (2002).
8. W.P. Hu, K. Manabe, T. Furukawa, and M. Matsumura, Appl. Phys. Lett. **80,** 2640 (2002).
9. A. Dodabalapur, L. J. Rothberg, R. H. Jordan, T. M. Miller, R. E. Slusher and J. M. Phillips, J. Appl. Phys., **80** 12 (1996).
10. A. Dodabalapur, L. J. Rothberg and T. M. Miller, Appl. Phys. Lett., **65** 2308 (1994).
11. V. Bulovic, V.B. Khalfin, G. Gu, P. E. Burrows, D.Z. Garbuzov and S. R. Forrest, Physical Review B. **58** 3730 (1998).
12. D. K. Gifford and D. G. Hall, Appl. Phys. Lett., **80** 3679 (2002).
13. P. A. Hobson, J. A. E. Wasey, I. Sage and W. L. Barnes, IEEE J. on Selected Topics in Quantum Electronics. **8** 378 (2002).

We claim:

A top emitting OLED comprising:

a substrate;

a first aluminum layer deposited above said substrate;

an ITO layer deposited above said first aluminum layer;

a TPD layer deposited above said ITO layer;

an Alq3 layer deposited above said TPD layer; and,

a transparent cathode deposited above said Alq3 layer, said transparent cathode comprising:

an LiF layer deposited above said Alq3 layer;

a second aluminum layer deposited above said LiF layer;

an SiO:Al layer deposited above said second aluminum layer;

wherein when a current is applied between said ITO layer and said transparent cathode light is emitted by said Alq3 layer.

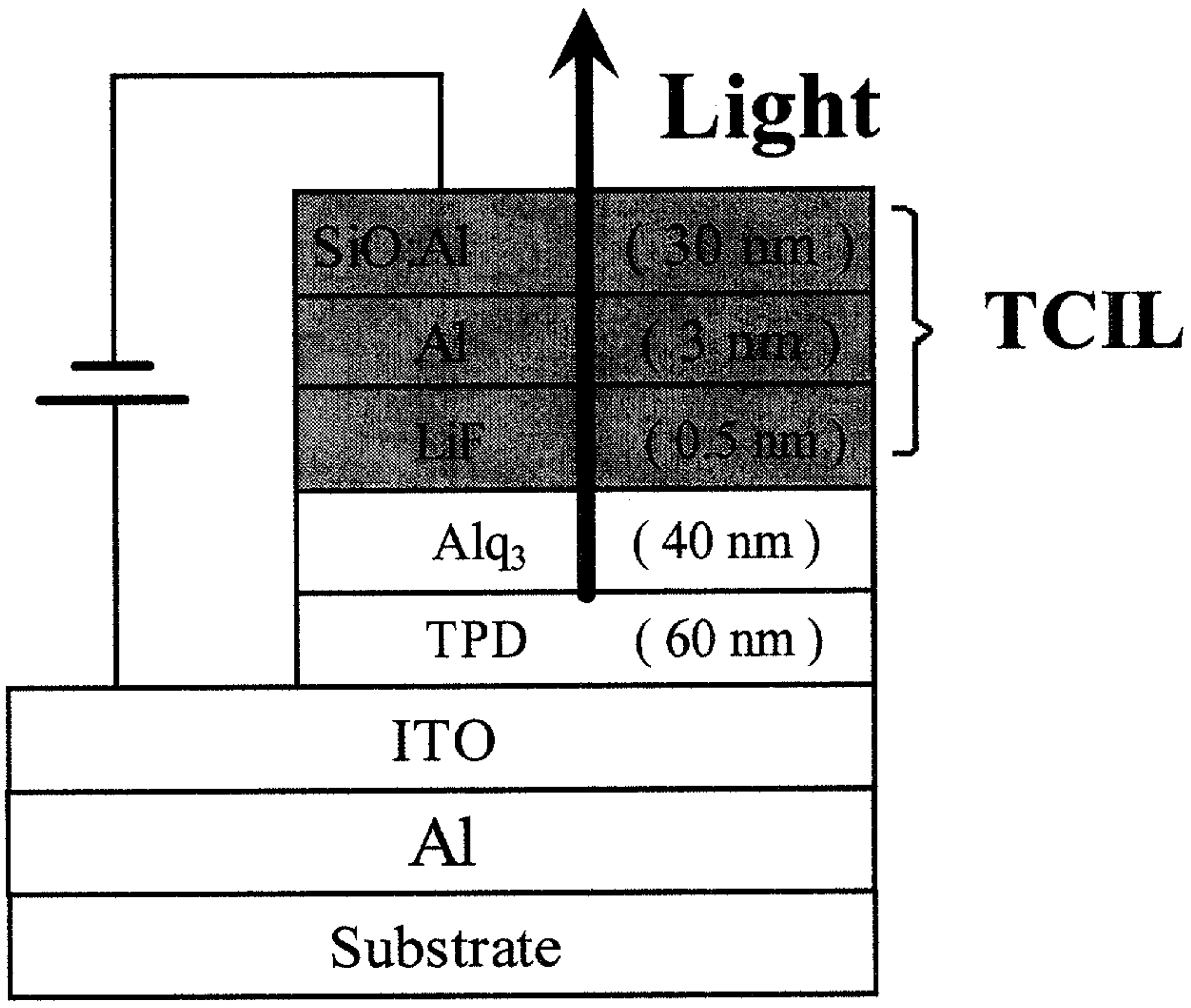


Fig. 1. Schematic cross-sectional diagram of the top-emitting OLED structure. TCIL is a combination of LiF, Al and Al-doped-SiO films.

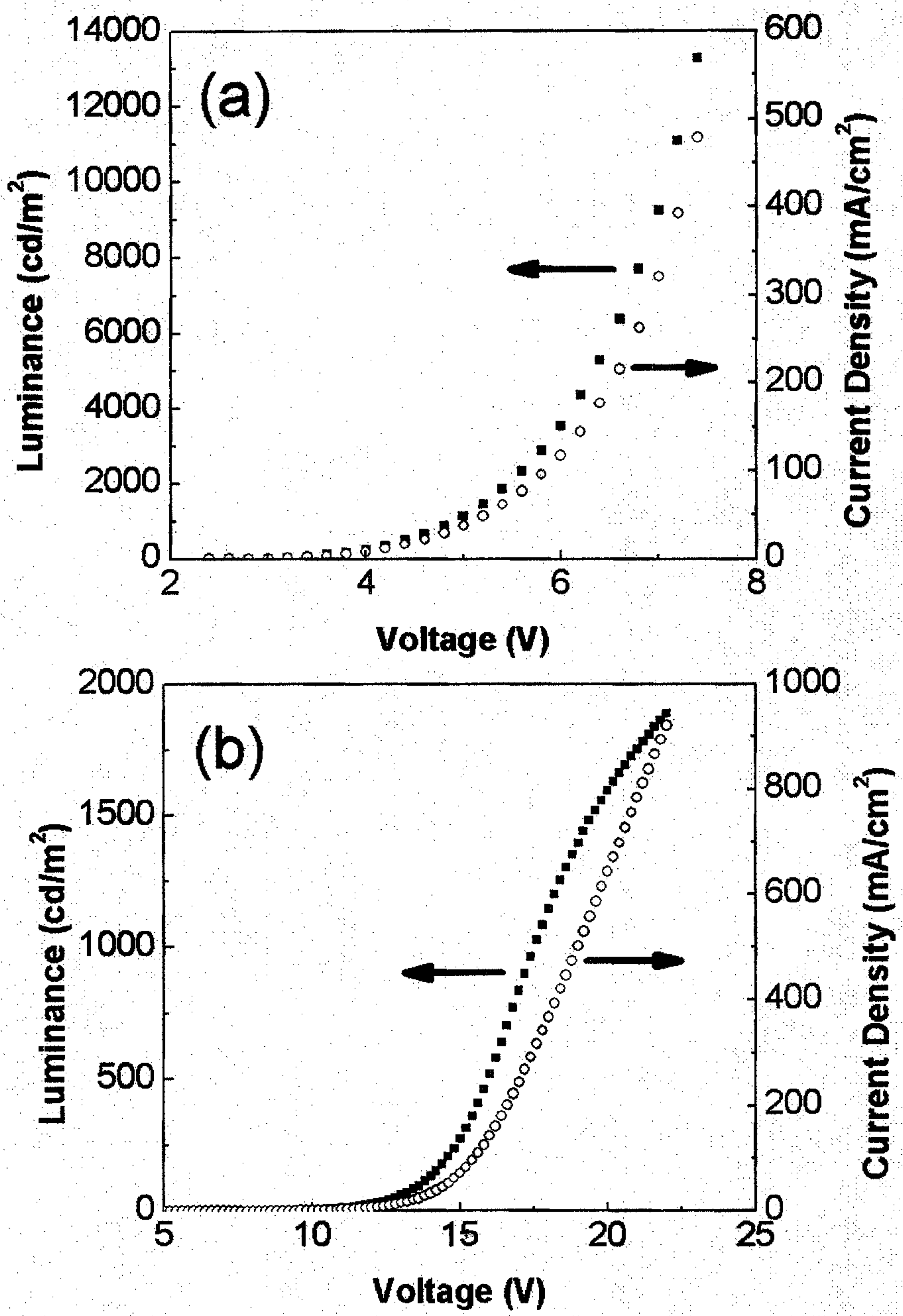


Fig. 2. Luminance (L)-current density (J)-voltage (V) of (a) OLED and (b) TOLED. Filled diamonds and open circles are responsible for the L-V and J-V, respectively.

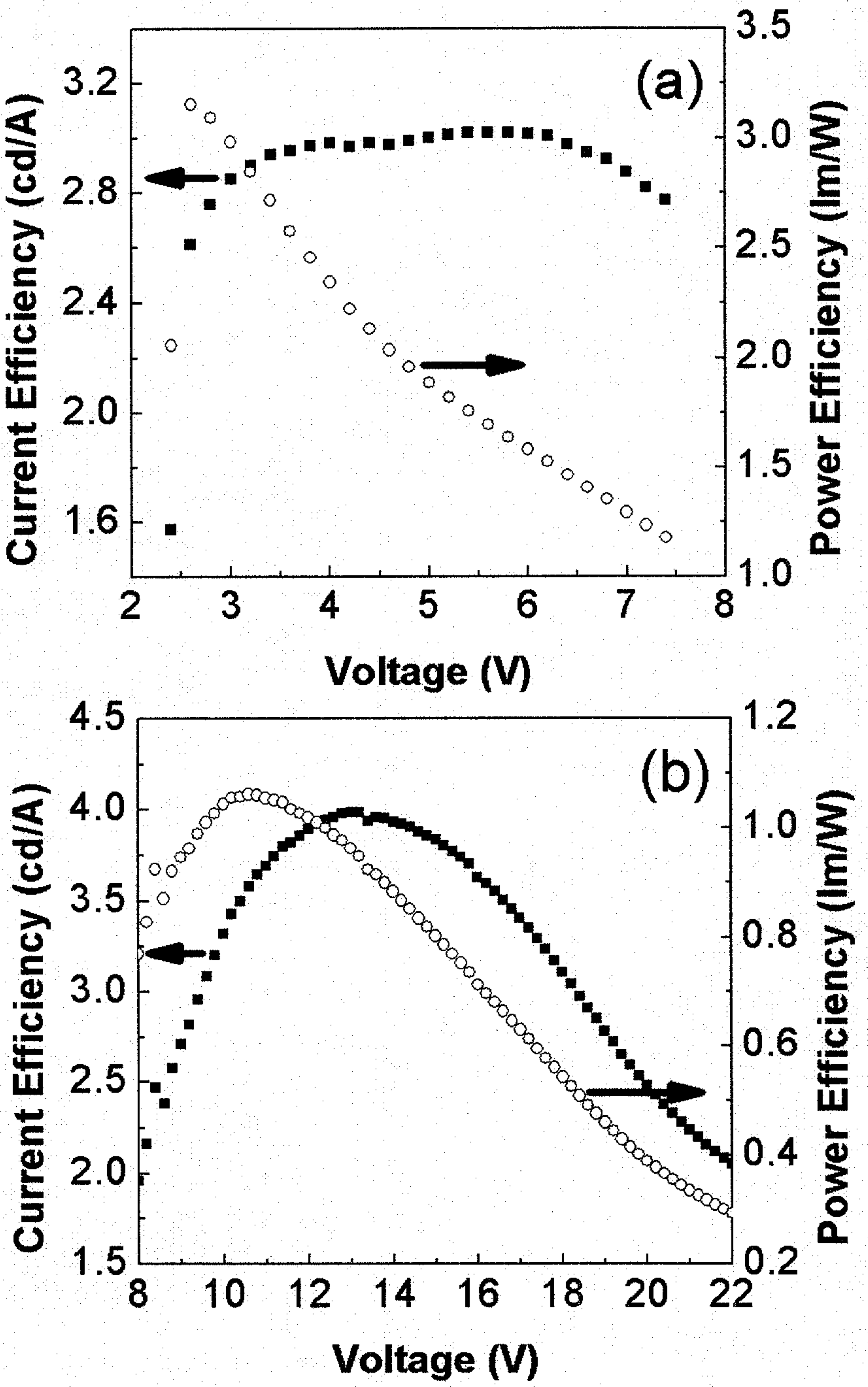


Fig. 3. Efficiencies of OLED and TOLED. Filled diamonds shows the independence of Current efficiency (L/J) vs. Voltage. And open circles shows the independence of Luminous power efficiency vs. Voltage.

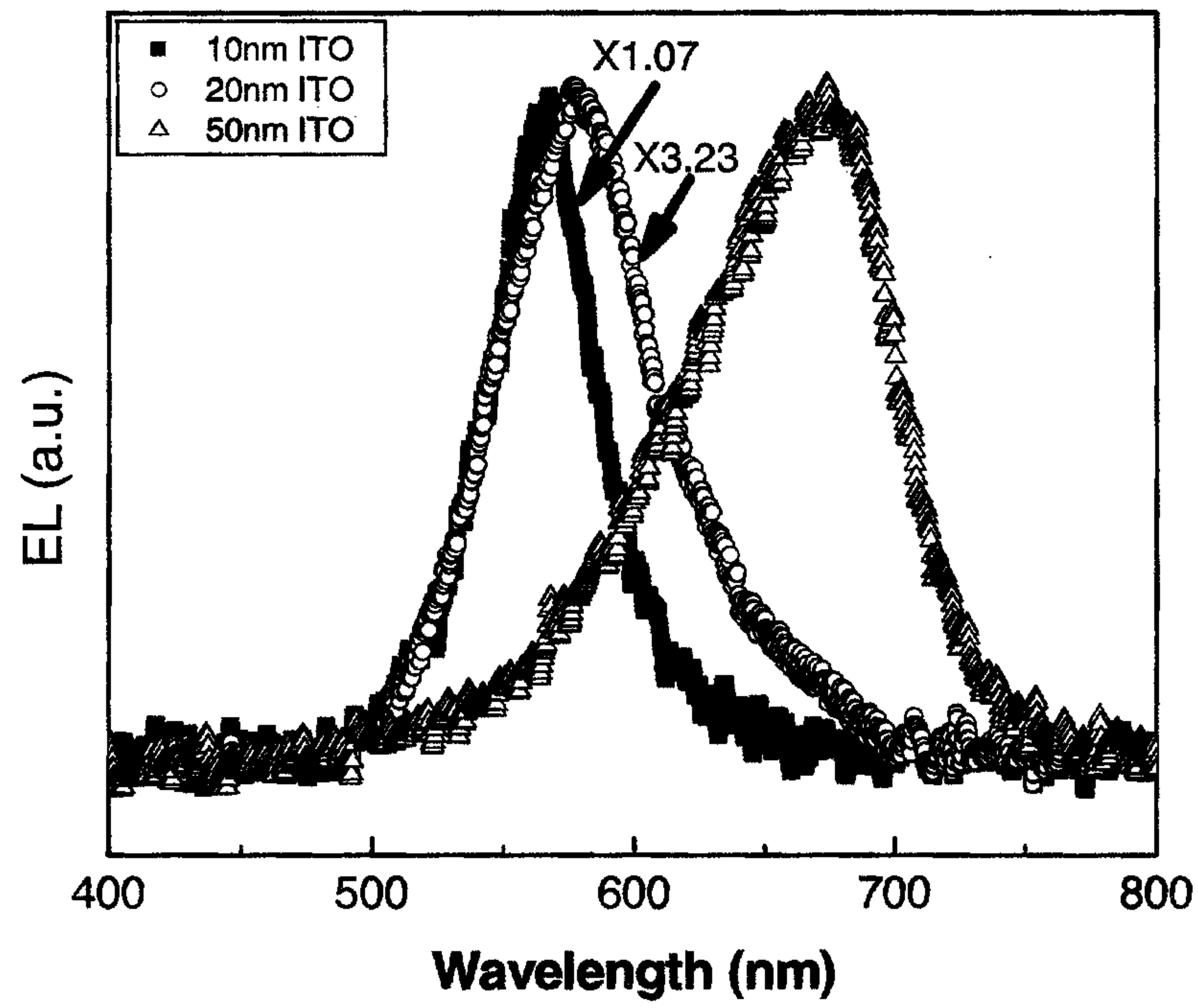
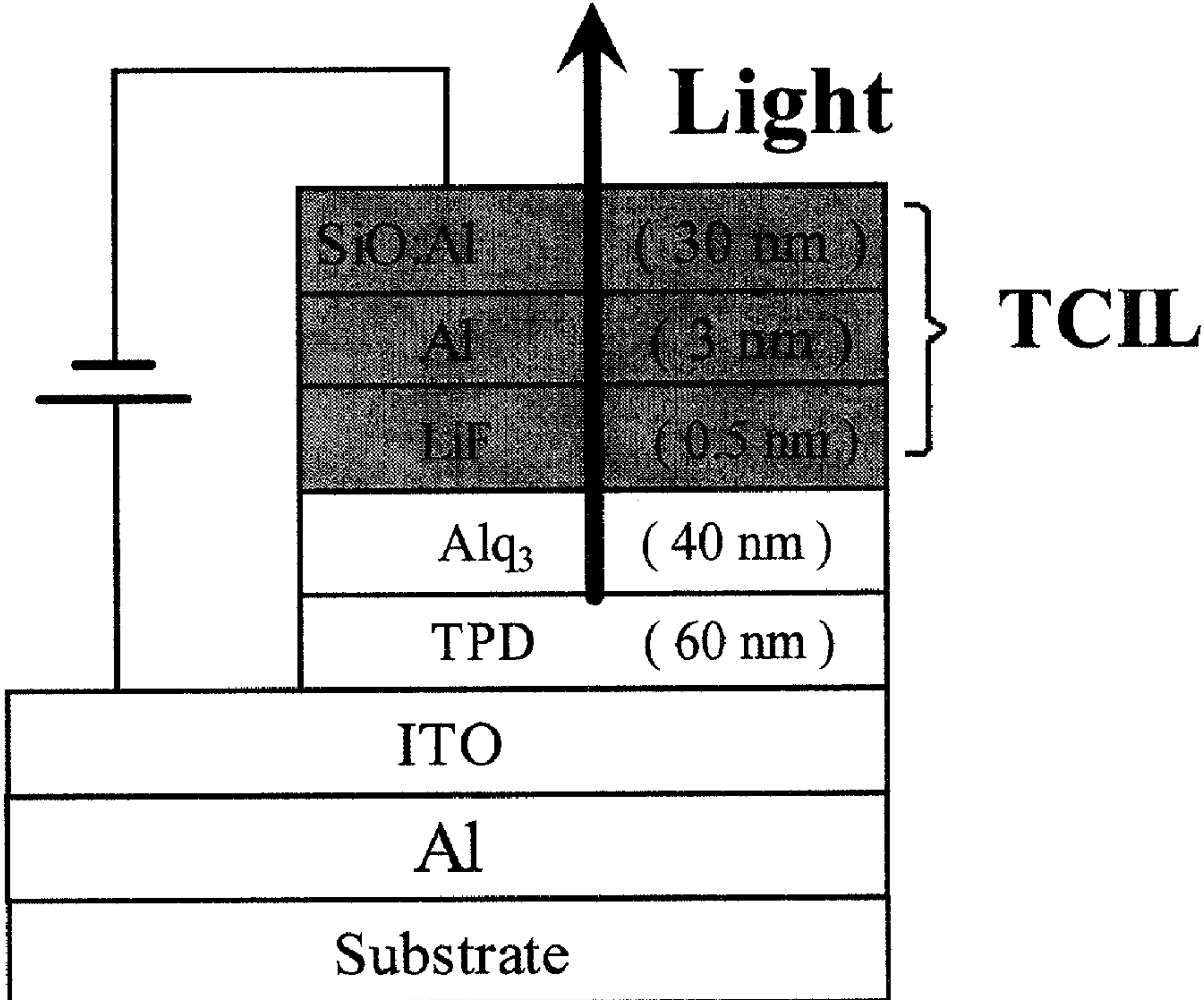


Fig. 4. Electroluminescent spetra of the devices with different thickness of ITO. The numbers in the graph indicate the enlarging factor during normalization.


Light
SiO:Al (30 nm)
Al (3 nm)
LiF (0.5 nm)
TCIL
Alq3 (40 nm)
TPD (60 nm)
ITO
Al
Substrate