White organic electroluminescence from an exciplex based on the novel substituted aluminium quinolate complex

Jung-An Cheng*[a] and Chin H. Chen[b]

A saturated white organic light-emitting device was obtained from an exciplex mechanism by using a novel blue-green fluorescent tris(5-mesitylsulfone-8-hydroxyquinolato)aluminium(III), $Al(MSq)_3$. Its absorption and photoluminescence appear at 3.25 eV and 2.56 eV. In photoluminescence, an exciplex, generated from an $Al(MSq)_3$/TPD blend, was found at 2.10 eV. The lifetime of the $Al(MSq)_3$/TPD blend is 22.3 ns, which is longer than that of either $Al(MSq)_3$ or TPD. Due to the exciplex formation, the orange light generated can be combined with the blue-green light generated from $Al(MSq)_3$ to achieve a white light emission. In an optimized electroluminescence device structure of [glass/ITO (200 nm)/PEDOT:PSS (75 nm)/blend (72 nm)/ BCP (8 nm)/LiF (1 nm)/Al (200 nm)], the saturated white light emission was observed at Commision Internationale De L'Eclairage (CIE) coordinates of (0.35, 0.34). The EL spectrum of the device is also independent of the applied voltage. Its maximum brightness and efficiency are 26 cd m^{-2} at 15.2 V/67 mA cm^{-2}, and 597 µW A^{-1} at 12.5 V/28 mA cm^{-2}, respectively.

Introduction

Organic electroluminescence (EL) is the electrically driven emission of light from non-crystalline organic materials, which was first observed and extensively studied in the 1960s.[1,2] Since Tang discovered the first highly efficient multiplayer organic light-emitting diode (OLEDs) in 1987,[3] electroluminescent devices based on organic semiconductors have gained a great deal of attention due to their high luminance efficiency, low power consumption, wide range of emissive colors, and low cost lighting possibility. Therefore, the growing interest is largely motivated with the employment of this technology in flat panel displays and the lighting source.

Due to the fact that the organic films can be formed by vacuum evaporation or solution casting, the fabrication cost is comparable with that of the liquid crystal displays (LCDs). However, there are few white light-emitting devices owing to the lack of the organic fluorescent dyes with white fluorescence. Several approaches to obtain white EL were reported.[4–9] Up to now, a simple approach for making white electroluminescent devices is still needed. Recently, different methods of obtaining white emission based on singlet exciplexes[10–14] and triplet excimers[15,16] have also been proposed, which is clearly advantageous from the perspectives of both cost and the ease of fabrication. But the chromaticities of devices constituted by photo-complexes are still far from the real and saturated white emission of the 1931 CIE$_{x,y}$ (0.33, 0.33). In this article, we present our experimental study of color tunable blends based on the novel blue-green metal complexes, tris[5-(mesitylsulfone)-8-quinolato]aluminium [Al(MSq)$_3$] synthesized by the molecular engineering of Alq$_3$ to obtain saturated white organic light-emitting devices. The mechanisms of obtaining saturated white were also proposed.

*jacheng.ac89g@nctu.edu.tw

Experimental

General comments

All the reactions were carried out under nitrogen atmosphere. THF was distilled from benzophenone–sodium before using. Mesitylene was distilled from calcium hydride. All reagents were purchased from commercial sources and were used without purification. Melting points were taken on a traditional apparatus without correction. ^{1}H NMR and IR spectra were recorded by a Varian Unity 300 Hz spectrometer and a Nicolet 360 FT-IR, respectively. Elemental analysis was carried out by a HEREAUS CHN-OS RAPID. Optical absorption was measured by a HEWLETT PACKARD 8453 absorption spectrometer. The thermal properties of aluminium complexes were determined by means of thermogravimetric analysis (TGA, SEIKO I TG/DTA 200) and differential scanning calorimetry (DSC, SEIKO SII DSC 2000).

Preparation of 8-hydroxyquinoline-5-mesitylsulfone (3)

8-hydroxyquinol-5-sulfonic acid (10.00 g, 44.44 mmol), and 25.0 ml methanesulfonic acid with 10% P_2O_5 and mesitylene (5.87 g, 48.89 mmol) were placed in a 100 mL round-bottom flask fitted with magnetic stirrer, condenser, and N_2 inlet. The mixture was heated overnight at 80 °C. The reaction mixture was filtered to remove the solid by-product. Solvent was removed by rotary evaporator and the solid was sublimed at 2.0×10^{-2} mmHg at 150 °C. White crystals were collected with a yield of 11.62 g (80%). ^{1}H NMR (300 MHz, CDCl$_3$) δ 8.831 (1 H, d, J = 4.20 Hz); 8.768 (1 H, d, J = 9.0 Hz); 8.046 (1 H, d, J = 8.40 Hz); 7.535 (1 H, q); 7.218 (1 H, dd, J = 8.25 Hz); 6.949 (2 H, s); 2.546 (6 H, s); 2.297 (3 H, s). Mp 203–204 °C. IR (KBr) 3365, 3103, 3052, 2975, 2929, 2914, 1685, 1623, 1603, 1567, 1506, 1465, 1307, 1270, 1209, 1137, 953, 850, 793, 678, 581 cm^{-1}. The product was characterized by mass

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spectrometry which revealed no extraneous peak other than the desired parent m/z = 327 (M$^+$); 328 (M$^+$ + 1) (for $C_{18}H_{17}NO_3S$). Anal. Calcd for $C_{18}H_{17}NO_3S$: C, 66.03; H, 5.23; N, 4.28; O, 14.66; S, 9.79. Found: C, 66.07; H, 5.24; N, 4.12%.

Preparation of tris[5-(mesitylsulfone)-8-quinolato-N_1,O_8]aluminium (4)

A 200-mL flask fitted with N$_2$ inlet, septum and stir bar was flame-dried and cooled under N$_2$. Compound **3** (1.00 g, 3.05 mmol) was added along with 60 mL of anhydrous THF. A hexane solution of triethylaluminium (1.11 mL, 15 w/w%) was added using a syringe. A yellow solution with blue-green fluorescence was observed immediately. The reaction was stirred at room temperature for 24 h. Solvent was removed and the volatile impurities present in the remaining yellow residue were removed by vacuum sublimation to give 0.97 g (97%) of **4**. Mp 227–228 °C. T_g = 192.4 °C. T_d = 333.5 °C (weight loss at 5 wt%). ^{1}H NMR (300 MHz, CDCl$_3$) δ 9.00 (1 H, d, J = 9.9 Hz), 8.87 (1 H, dd, J = 9.9 Hz), 8.80 (1 H, s), 8.77 (1 H, d, J = 4.5 Hz), 8.73 (1 H, d, J = 6.0 Hz), 8.10 (2 H, dd, J = 8.7 Hz), 7.99 (1 H, d, J = 8.4 Hz), 7.60 (1 H, dd, J = 8.7 Hz), 7.49 (1 H, dd, J = 8.7 Hz), 7.36 (1 H, d, J = 8.7 Hz), 7.35 (1 H, d, J = 8.7 Hz), 7.24 (1 H, d, J = 1.2 Hz), 7.02 (1 H, d, J = 8.7 Hz), 7.00 (2 H, d, J = 8.4 Hz), 6.93 (6 H, s), 2.55 (9 H, s), 2.52 (9 H, s), 2.28 (9 H, s). IR (KBr) 3088, 3037, 2929, 2842, 1741, 1602, 1567, 1496, 1462, 1383, 1305, 1229, 1198, 1145, 1086, 1045, 978.66, 820, 794, 753, 698, 659, 579, 553, 518, 451, 415 cm^{-1}. Product **4** was characterized by mass spectrometry which revealed no extraneous peak other than the desired parent m/z = 1006. Anal. Calcd for $C_{54}H_{48}N_6O_9S_3Al$: C, 64.46; H, 4.81; N, 4.18; O, 14.31; S, 9.56; Al, 2.68. Found: C, 64.90; H, 4.89; N, 4.35%.

Photoluminescence and lifetime measurement

The photoluminescence of PVK, TPD, Al(MSq)$_3$, and BCP in 1,2-dichloroethane (c = 10^{-3}–10^{-4} mol L^{-1}) were measured, separately. For the PL experiments, the excitation wavelength was set at the absorption maximum of each specimen. The lifetime of each component spin-coated into a thin film containing 55 wt% PVK, 15 wt% TPD and 30 wt% Al(MSq)$_3$ was measured at 298 K by an Edinburgh Instruments FL900. The excitation energy was generated by a nF 900 Nanosecond Flashlamp filled with 99.999% pure hydrogen, and the pulse frequency was 40 KHz. The emitting photons were individually recorded at 3.31, 2.98, 2.56, and 2.10 eV, *via* time-correlated single photon counting (TCSPC). Singlet lifetimes were obtained by a single exponential fit of the emission decay.[17] Similar approaches were also applied for Al(MSq)$_3$, PVK, and TPD in 1,2-dichloroethane, individually.

Device fabrication

The devices [ITO/PEDOT:PSS/Al(SPhq)$_3$ blend/BCP/LiF/Al] were prepared in the following manner for EL measurement. For all structures, the indium tin oxide (ITO) glass with 20 Ω □$^{-1}$ sheet resistance were used as substrates. Poly(3,4-ethylenedioxythiophene)/poly(styrene)-sulfonate (PEDOT:PSS,

obtained from Bayer) aqueous solution was spin-coated on the cleaned ITO substrates and baked at 110 °C for 24 h. Blend Al(MSq)$_3$/TPD was subsequently spin-coated on the PEDOT layer with a typical thickness of 72 nm. BCP (8 nm), lithium fluoride (LiF; 1 nm), and Al (200 nm) were sequentially deposited on the top of Al(MSq)$_3$/TPD layer at 1 × 10^{-6} mmHg. The emitting area of the device was 3 mm × 3 mm. The voltage–current–luminance characteristics and EL spectra were measured by using an optical power meter (PR-650) and digital source meter (Keithley 2400) under ambient atmosphere after encapsulation.
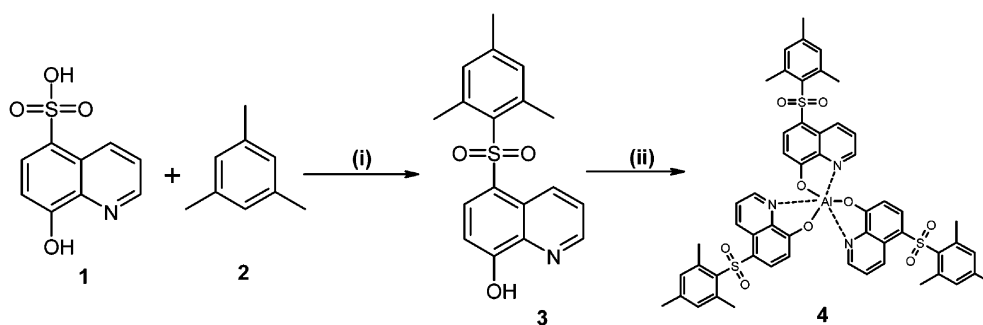
Results and discussion

Synthesis and characterization of Al(MSq)$_3$ complex

An aluminium(III) complex, Al(MSq)$_3$, having three 5-mesitylsulfone-8-hydroxyquinolato as a bidentate ligand, was synthesized as shown in Scheme 1. Commercially available 8-hydroxyquinoline-5-sulfonic acid was reacted with 1,3,5-trimethylbenzene (mesitylene) in methanesulfonic acid with Eaton's reagent[18] by Friedel–Crafts condensation. After purification, bidentate ligands (**3**) were subsequently chelated further with triethylaluminium in THF to give Al(MSq)$_3$. The overall yield was around 77.6%. In solubility test, this complex can be dissolved in commercial organic solvents such as tetrahydrofuran (THF), chloroform, *N,N'*-dimethylformaldehyde (DMF), xylenes, and 1,2-dichloroethane. This complex is also more soluble than other similar complexes species with sulfonamide substituents.[19,20]
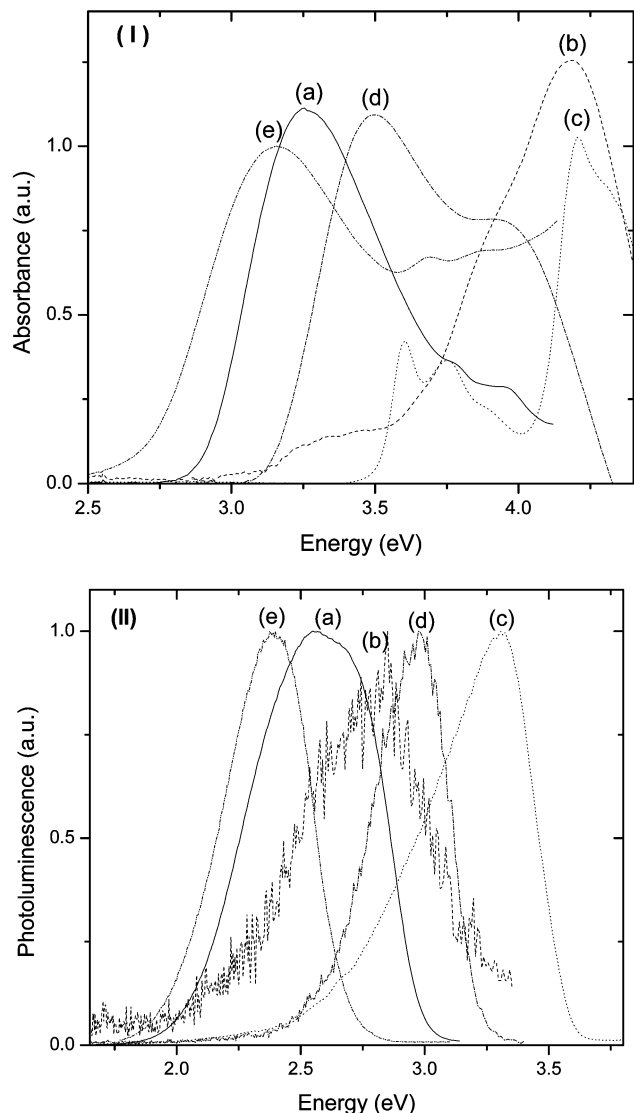
Absorption and photoluminescence (PL)

Fig. 1 shows the PL spectra and UV-Vis absorptions of materials used in this experiment. Their specific properties are also shown in Table 1. By regiospecific derivation of Alq$_3$ at one of its LUMO sites of C-5, sulfonyl groups removing electron density from the phenol ring will result in an increase in the energy of the π–π* transition, which hypsochromically shifts the emission of Alq$_3$ to blue.[21–23] In PL spectra, Al(MSq)$_3$ exhibits intense blue-green luminescence in solution. Its absorption and PL maximum appeared at 3.25 eV (λ_{max} at 382.0 nm) and 2.56 eV (λ_{max} at 485.2 nm), respectively. As expected, the introduction of the sulfone substituent to the C-5 of the quinoline ring system leads to a blue-shift of 0.07 eV (8.2 nm) and 0.15 eV (29.0 nm) in the absorption and emission of Al(MSq)$_3$ as compared to those of Alq$_3$, respectively. In the cyclic voltammetry,[24] we also found the HOMO/LUMO energy levels of Al(MSq)$_3$ are −6.02 eV/−3.13 eV. They are 0.40 eV/0.27 eV lower than those of Alq$_3$ (−5.62 eV/−2.80 eV).

In addition, the maximum absorbance of *N,N*-diphenyl-*N,N'*-bis(methylphenyl)-(1,1′-biphenyl)-4,4′-diamine (TPD) and 2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline (BCP) are 3.50 eV (354.6 nm) and 4.19 eV (296.3 nm), respectively. There are two peaks at 3.75 eV (330.4 nm) and 3.60 eV (344.1 nm) in PVK. The corresponding PL peaks for PVK, TPD and BCP were found at 3.31 eV (375.0 nm), 2.98 eV (416.6 nm) and 2.85 eV (435.0 nm), respectively.

Scheme 1 Synthesis of blue emitter Al(MSq)$_3$. (i) 10% P$_2$O$_5$ in methanesulfonic acid; (ii) THF, triethylaluminium.

Fig. 1 Optical spectrum of materials. (I) UV-Vis absorption; (II) photoluminescence (PL): (a) Al(MSq)$_3$; (b) BCP; (c) PVK; (d) TPD; (e) Alq$_3$.

Table 1 Photoluminescence, UV-Vis absorption and HOMO/LUMO energy levels of materials

| Materials | Absorption | | Photoluminescence | | HOMO[a] | LUMO[a] |
	$\lambda_{max}/$ nm	$E_{ABS}/$ eV	$\lambda_{max}/$ nm	$E_{PL}/$ eV	$E_{HOMO}/$ eV	$E_{LUMO}/$ eV
Al(MSq)$_3$	382.0	3.25	485.2	2.56	−6.02	−3.13
BCP	296.3	4.19	435.0	2.85	−6.24	−2.93
Alq$_3$	390.2	3.18	514.2	2.41	−5.62	−2.80
PVK	344.1	3.60	375.0	3.31	−5.81	−2.14
	330.4	3.75				
	294.9	4.21				
TPD	354.6	3.50	416.6	2.98	−5.40	−2.48
	316.9	3.91				

[a] These values are relative to the vacuum energy level.

constituent species. The reason why it is relatively easier to form an exciplex for conjugated organics due to LUMOs is because of the highly delocalized π orbitals.[25] The process of forming an exciplex can be characterized by the following equations. A donor (D) can form an exciting state (D*) after absorbing amount of energy hv_1 as shown in eqn. (1). Combing with an acceptor (A), the excited complex (D/A)* can also be formed as shown in eqn. (2). Emission hv_2 occurs when the excited complex (D/A)* relaxes to their ground state D and A as indicated in eqn. (3).[26]

$$D + hv_1 \rightarrow D^* \tag{1}$$

$$D^* + A \rightarrow (D/A)^* \tag{2}$$

$$(D/A)^* \rightarrow D + A + hv_2 \tag{3}$$
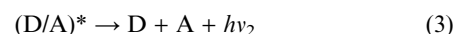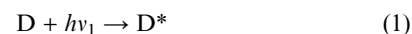
Due to the sulfone, a strong e-withdrawing group was introduced into the quinoline based ligand, so that the Al(MSq)$_3$ turns into a strong e-acceptor. In contrast, TPD is a well-known hole-transporting and strong e-donating material. As a result, a donor–acceptor complex can be easily formed when the two are mixed together in the solid phase. As shown in Fig. 2, we first treated PVK as the matrix, then doped it separately with Al(MSq)$_3$ and TPD (i.e. Fig. 2a and h, respectively). From the spectra, only the emission peaks of Al(MSq)$_3$ (at 2.56 eV) and TPD (at 2.98 eV) were observed. When PVK/TPD/Al(MSq)$_3$ was blended in various ratios, it was found that the peak intensity at 2.56 eV decreased as the TPD concentration increased from 0 wt% to 30 wt%. On the other hand, the intensity of the peak (at 2.10 eV) originating

## Exciplexes and white light emission

According to the theory of photo-physics, exciplex generation is favored where there is significant spatial overlap between the lowest unoccupied molecular orbitals (LUMOs) of the
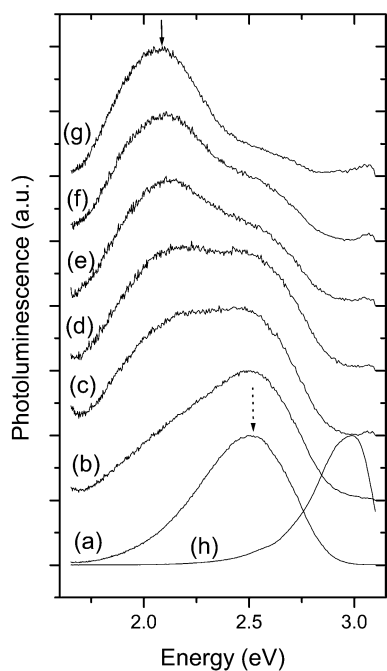
Fig. 2 Photoluminescence of PVK/TPD/Al(MSq)$_3$ thin solid films. The weight ratios of PVK/TPD/Al(MSq)$_3$ mixtures are as follows: (a) 70/0/30; (b) 65/5/30; (c) 60/10/30; (d) 55/15/30; (e) 52/18/30; (f) 50/20/30; (g) 40/30/30; (h) 0/50/0.

Table 2 Integral components and their corresponding 1931 CIE$_{x,y}$ coordinates in solid films

	Weight ratio of components (wt%)			1931 CIE coordinates	
Entry	PVK	TPD	Al(MSq)$_3$	x	y
(a)	70	0	30	0.24	0.40
(b)	65	5	30	0.31	0.40
(c)	60	10	30	0.35	0.40
(d)	55	15	30	0.36	0.41
(e)	52	18	30	0.39	0.41
(f)	50	20	30	0.41	0.42
(g)	40	30	30	0.43	0.42
(h)	50	50	0	0.16	0.05

from the exciplex increased. In the corresponding chromaticity values shown in Table 2, the 1931 CIE$_{x,y}$ coordinates were changed from blue-green (0.24, 0.40) [originating from Al(MSq)$_3$; Table 2(a)] to orange (0.43, 0.42) [generated from TPD/Al(MSq)$_3$ complex; Table 2(g)]. In the balanced weight ratio of PVK/TPD/Al(MSq)$_3$ at 60/10/30, a saturated white light emission with (0.35, 0.40) was obtained [Table 2(c)].

To verify the exciplex formation mechanism and energy transfer, a series of absorption spectra of thin films were measured as shown in Fig. 3. Fig. 3a and b are the absorptions of Al(MSq)$_3$ and TPD in the solid phase, respectively. Their absorptions appeared at 3.25 eV and 3.50 eV, separately. When TPD was blended into Al(MSq)$_3$, as shown in Fig. 3c, we can observe not only the absorption peak from TPD at 3.25 eV but also a shoulder at low energy generated from Al(MSq)$_3$. For further exploration, excitation was also measured to trace back the exciplex energy as shown in Fig. 3d. Clearly, this spectrum is very similar to Fig. 3c. It implies that
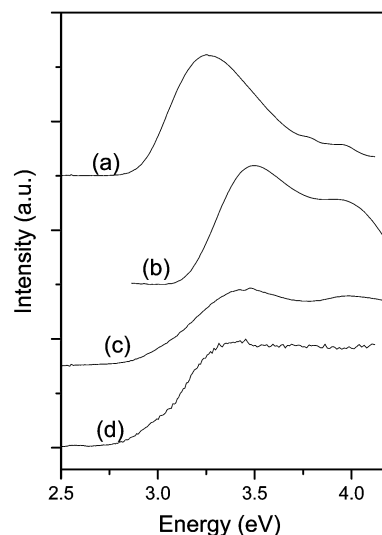
Fig. 3 Absorption spectra of thin films: (a) Al(MSq)$_3$, (b) TPD, and (c) Al(MSq)$_3$ + TPD. Here, (d) is the photoluminescence excitation spectrum of Al(MSq)$_3$ + TPD.

the energy source for the exciplex formation is not only from TPD but also extracted from Al(MSq)$_3$. This result also suggests that blend thin films excited by the absorption energy of either TPD or Al(MSq)$_3$ will form exciplex emission.

From the schematic energy diagram shown in Fig. 4, we can suggest that the electron of TPD (donor) will hop from its HOMO to LUMO (path A) after excitation. By intersystem crossing, the electron will go through path (B) and transfer to the LUMO side of Al(MSq)$_3$. Consequently, the electron in the LUMO of Al(MSq)$_3$ will decay radiatively to the HOMO of TPD by path (C). Therefore, we could observe a yellow-orange emission after excitation. Similar phenomena were also explored by Mazzeo and Thompson et al.[11,27] In addition, a blue-green emission will also be observed while is Al(MSq)$_3$
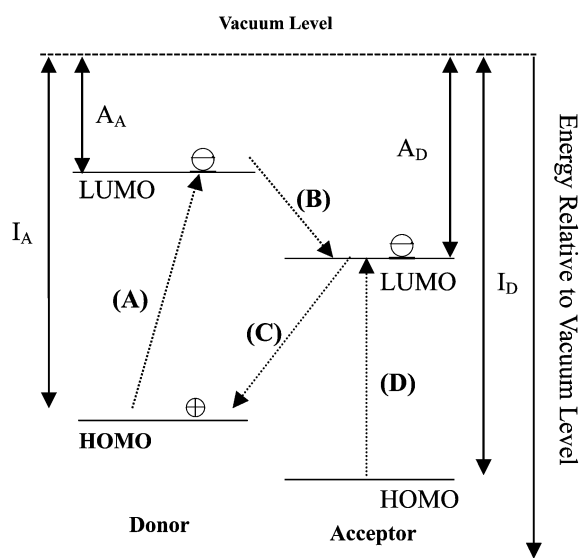
Fig. 4 Mechanism of exciplex formation and decay. A_D, I_D and A_A, I_A are the electron affinities and ionization potentials of the donor and acceptor molecules, respectively.

Table 3 PL Lifetime value of organic materials measured by time-resolved PL at room temperature

Materials	Observed energy/eV	PL lifetime[a]/ns	PL lifetime in white light emission/ns
Al(MSq)$_3$	2.56	16.3	16.5
PVK	3.31	12.1	—[b]
TPD	2.98	3.4	—[b]
Exciplex	2.10	—[b]	22.3

[a] They were measured in 1,2-dichloroethane individually. [b] The values cannot be detected during excitation.

excited and radiated recoverably. According to their corresponding energy levels in Table 1, the energy of the exciplex emission is very close to the difference between the LUMO side of the Al(MSq)$_3$ and the HOMO side of the TPD.

In this experiment, the PL lifetime measurement was used to verify the relation between the exciplex and the blend. The individual fluorescence lifetimes of Al(MSq)$_3$, PVK, and TPD are shown in Table 3. Their corresponding lifetimes are 16.3 ns, 12.1 ns, and 3.4 ns, respectively. The lifetimes of Al(MSq)$_3$ in Al(MSq)$_3$/PVK/TPD blended thin film and exciplex were measured to be 16.5 ns and 22.3 ns. It is obvious that the exciplex shows a longer lifetime than other materials in Table 3.

Furthermore, we cannot detect the photons emitted from PVK and TPD. Based on the above verification, it is reasonable to suggest that the white-light spectrum of PVK/TPD/Al(MSq)$_3$ is contributed to by two emissions of the exciplex TPD/Al(MSq)$_3$ (yellow-orange) and Al(MSq)$_3$ itself (blue-green).

Electroluminescence (EL)

To observe the white emission in EL, devices based on the blend were fabricated. All of the device architectures and material structures are shown in Fig. 5. In the testing devices of the structure [glass/ITO/PEDOT:PSS/blend/LiF/Al] as shown in Fig. 5(a), PEDOT is used as a hole-injection layer. Since TPD possesses excellent hole-transporting ability,[28] the band gap between the anode and the emitting layer can effectively be reduced. As the result, the turn-on voltage of the device with TPD can be lowered in comparison with that without TPD. Their EL spectra were also similar to that in Fig. 2. From the rough data, however, those devices showed poor efficiency. This could be due to the mixed organic layers, which cannot efficiently confine recombination of holes and electrons in the emitting layer so that some excitons may be generated at the electrodes and relaxed non-radiatively as illustrated in Fig. 6(a). In this study, we have inserted BCP as the hole
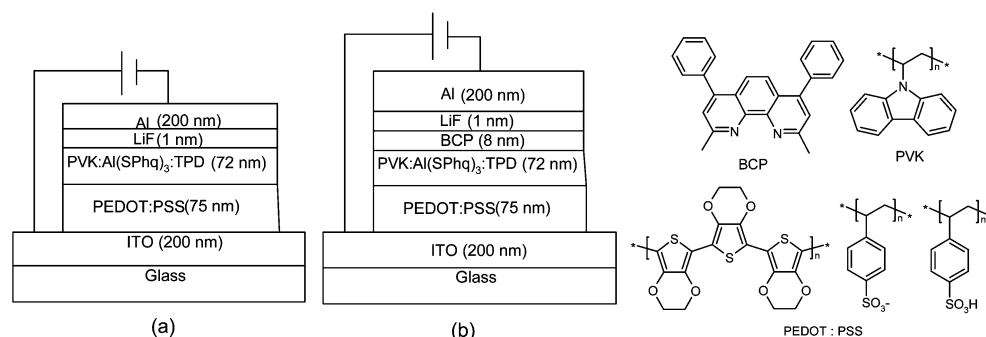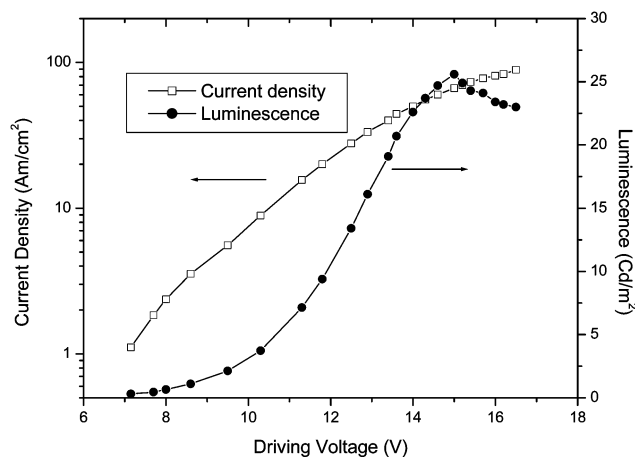

Fig. 5 The device architecture and molecular structures. The emitting layer was constructed with PVK/TPD/Al(MSq)$_3$ of 55/15/30 weight ratio.
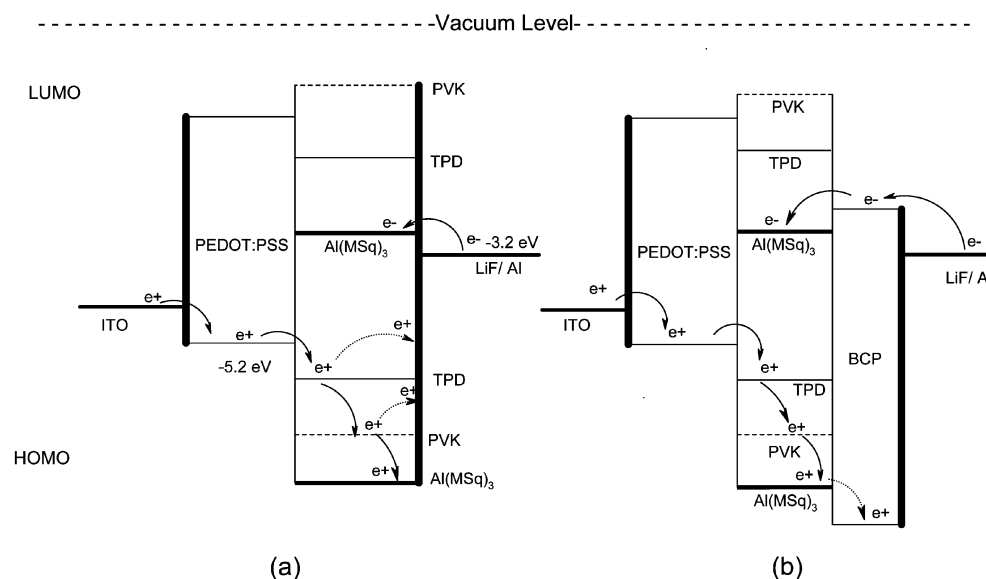
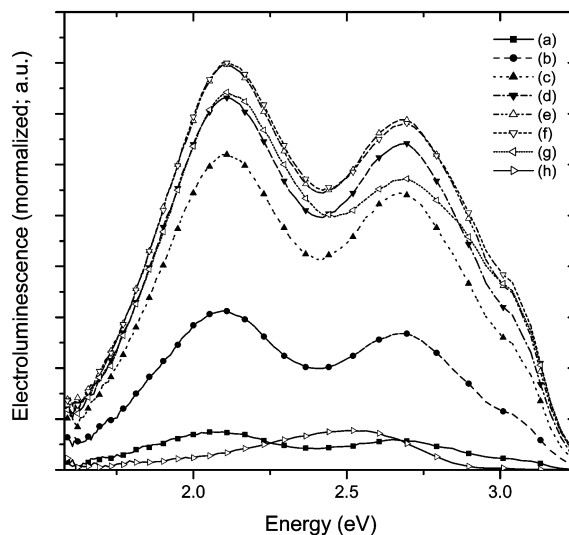**Fig. 6** Energy diagrams: (a) double-layer; (b) multi-layer with BCP.

blocking layer between the cathode and the blended layer to impede holes from diffusing into the cathode, therefore, enhancing EL efficiency. The modified device architecture is also shown in Fig. 5(b). Their corresponding voltage–current–brightness characteristics are depicted in Fig. 7.

In the EL device characterizations, the turn-on voltage is 7.1 V, which corresponds to a current density of 1.1 mA cm^{-2}. The maximum brightness and efficiency are 26 cd m^{-2} at 67 mA cm^{-2}/15.2 V and 597 μW A^{-1} at 28 mA cm^{-2}/12.5 V, respectively. We have observed that both current density and luminance increase with forward bias in Fig. 7. When the current density goes over 67 mA cm^{-2}, the brightness drops gradually. Its efficiency also drops from 47 μW A^{-1} to 31 μW A^{-1}. This could be attributed to the imbalance of electrons and holes injected into the emitting layer, which results in the quenching effect. In comparison, the white device based on Al(MSq)$_3$ showed higher brightness and efficiency than that of the 2,5-bis(5-*tert*-butyl-2-benzoxazolyl)thiophene (BBOT)/poly(3-hexylthiophene) (P3HT) based device.[14]

The intensity of EL spectra are shown in Fig. 8, and the trends of the curves were consistent with those of Fig. 7. From the EL spectra, two emission bands are observed. One appeared at 2.11 eV with an orange-yellow emission, and the other one located at 2.67 eV with a blue light. According to their PL in the blended film, the peak appearing at 2.11 eV was suggested to be generated from the exciplex of TPD/Al(MSq)$_3$. But the peak located at 2.67 eV seems to be higher than that of Al(MSq)$_3$. According to the energy diagram shown in Fig. 6(b), hole-transporting materials were dispersed in the emitting layer followed by BCP deposition so that holes cannot be confined within the emitting layer completely. Holes come from the anode will migrate toward BCP by passing through the blend layer. Therefore, these holes will recombine with electrons in the interface of the BCP layer and the emission energy will be close to that of BCP (2.85 eV). Here, we also want to mention that Al(MSq)$_3$ self-emits in the EL spectrum. A blank device was fabricated without doping TPD, and its EL spectra is shown in Fig. 7(h). However, since the EL intensity of Al(MSq)$_3$ is much weaker than that of BCP, the peak generated from Al(MSq)$_3$ could not be clearly observed in the blend device.
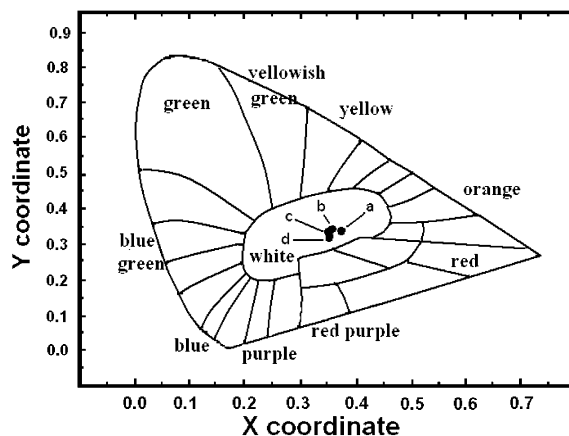

Fig. 8 Electroluminescence (EL) spectra of the double-layer devices. Their corresponding driving voltages (V) and current densities (mA cm^{-2}) are: (a) 9.5/6; (b) 11.8/20; (c) 13.4/40; (d) 14.0/50; (e) 14.6/60; (f) 15.2/70; (g) 16.5/89; (h) 16.8/89.

The 1931 CIE$_{x,y}$ coordinates of EL are shown in Fig. 9. The EL spectrum of the device and chromaticity were almost independent of the applied voltage. Due to the partial light generation from BCP, the CIE y value dropped, which resulted in the shift of white emission to CIE$_{x,y}$ (0.35–0.37, 0.34–0.35) from the original white emission of CIE$_{x,y}$ (0.35, 0.34) measured by photoluminance. This also makes the emission spectrum closer to the saturated white light of CIE$_{x,y}$ (0.35, 0.34) [Fig. 9(c)] than those of earlier studies.[11,13,14]

Conclusion

We have synthesized a novel blue-green organic electroluminescent material based on the substituted aluminium quinolate complex, Al(MSq)$_3$. Due to the blending of the strong e-withdrawing character of Al(MSq)$_3$ and an e-donating molecule of TPD, an exciplex emission of yellow-orange color

Fig. 7 The current density–voltage and brightness–voltage curves of the double-layer device.

Fig. 9 Electroluminescence 1931 CIE$_{x,y}$ coordinates for different applied bias: (a) 9.5 V at (0.37, 0.35); (b) 11.8 V at (0.36, 0.34); (c) 14.0 V at 0.35, 0.34); (d) 16.5 V at (0.36, 0.35).

was observed from Al(MSq)$_3$/TPD interaction during excitation in the solid phase. By adjusting the ratio of Al(MSq)$_3$ and TPD, a blue-green emission, from Al(MSq)$_3$, and a yellow-orange emission, from Al(MSq)$_3$/TPD, could be obtained simultaneously. This phenomenon was also observed in their corresponding EL processes. Devices comprised of the three components, i.e. PVK, TPD, and Al(MSq)$_3$, as the active region were produced by a solution process. A saturated white emission with (0.35, 0.34) was observed in the EL spectra, and their 1931 CIE$_{x,y}$ values were closer to the saturated white light (0.33, 0.33) than those reported in the literature while an additional hole-blocking layer of BCP was inserted into the device. Although the exciplex formation decreases the quantum efficiency of the device, we confirmed that it is a possible way for fabricating a white light EL device.

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Jung-An Cheng*[a] **and Chin H. Chen**[b]
[a]Department of Applied Chemistry, National Chiao Tung University, Hsinchu, Taiwan, ROC, 30050. E-mail: jacheng.ac89g@nctu.edu.tw; Fax: +886-3-5737681; Tel: +886-3-5712121 ext 56587
[b]Display Institute, Microelectronics & Information System Research Center, National Chiao Tung University, Hsinchu, Taiwan, ROC, 30050

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J. Mater. Chem., 2005, **15**, 1179–1185 | 1185