Efficient Organic White Light-Emitting Device Utilizing SAlq, A Novel Blue Light-Emitting Material

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Abstract

Efficient organic white light-emitting diodes are fabricated by doping [bis(2-methyl-8-quinolinolato) (tripheny-siloxy)aluminium (III)] (SAlq), a blue-emitting layer, with a red fluorescent dye of 4-dicyanomethylene-2-methyl-6-{2-(2,3,6,7-tetrahydro-1H,5H-benzo[i,j]quinolizin-8-yl)vinyl}-4H-pyran (DCM2). The incomplete energy transfer from blue-emitting SAlq to red-emitting DCM2 enables to obtain a balanced white light-emission. A device with the structure of ITO/TPD (50 nm)/SAlq:DCM2 (30 nm, 0.5 %)/Alq_3 (20 nm)/LiF (0.5 nm)/Al shows emission peaks at 456 nm and 482 nm from SAlq and at 570 nm from DCM2. The white light-emitting device shows an external quantum efficiency of about 2.3 %, a luminous efficiency of about 2.4 lm/W, and the CIE chromaticity coordinates of (0.32, 0.37) at 100 cd/m^2. A maximum luminance of about 23,800 cd/m^2 is obtained at 15 V and the current density of 782 mA/cm^2.

1. Introduction

An organic electroluminescence device (OLED) becomes one of the most promising next-generation large-area flat panel display systems since C. W. Tang and S. A. Vanslyke reported highly efficient OLEDs utilizing tris(8-quinolinolato-N^1,O^3)aluminium (III) (Alq_3) [1]. A variety of materials have been reported for electroluminescence (EL) with various-color emission. However, there are few organic materials and dyes to generate white light-emission.

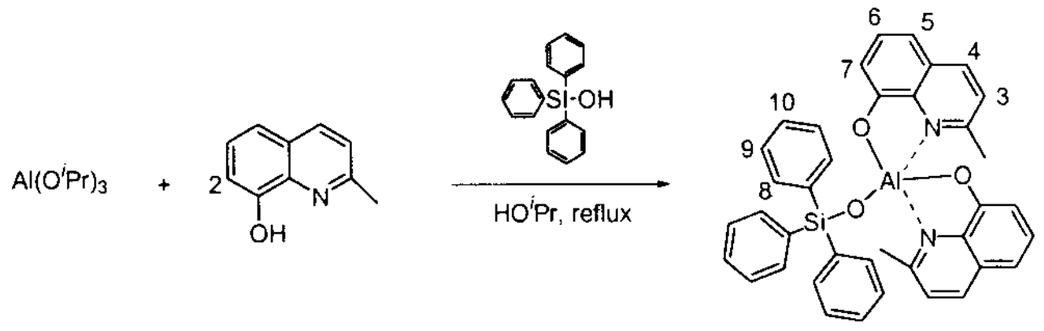
White light-emission is very important for applying OLEDs such as a full-color display combined with a color filter, a backlight of TFT-LCDs, and illumination. J. Kido *et al.* and other researchers have reported various types of white OLEDs [2-10]. Multilayer structures of complementary-color emitting layers or RGB primary-color emitting layers are generally utilized for white light-emitting devices.

White light-emission in doped devices is affected by the complete energy transfer from the host to the dopant as well as charge carrier trapping at dopant molecules. In addition, the low fluorescent efficiency of blue-emitting materials in such devices makes the Commission Internationale d'Eclairage (CIE) chromaticity coordinates deviate from a balanced white point of (0.333, 0.333). Therefore, blue-emitting materials are an important requirement to develop white light-emitting devices. However, blue-emitting materials with high brightness and thermal stability still remain to be developed [11].

In this paper, we report the synthesis of blue-emitting SAlq with a high EL quantum efficiency and the white light-emitting devices with a single-emitting layer of SAlq doped with red-emitting DCM2.

2. Experimental

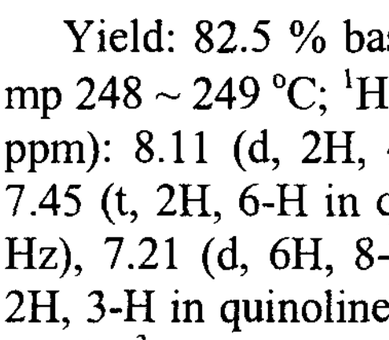
Bis(2-methyl-8-quinolinolato) (triphenylsiloxy) aluminium (III) was synthesized in high yield and high purity through the homogeneous-phase reaction between aluminium *iso*-propoxide and two ligand of 2-methyl-8-quinolinol and triphenylsilanol at reflux condition as shown in scheme 1.



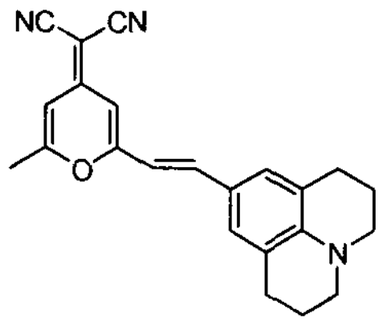
<Scheme 1>

All manipulations were performed under a nitrogen atmosphere using standard techniques. All solvents and chemicals were reagent grade. The purity of SAlq was characterized by ^{1}H NMR, ^{13}C NMR, PL spectra, elemental analysis, and melting test. ^{1}H and ^{13}C NMR spectra were recorded on Varian Unity-400 spectrometer with TMS as internal standard and elemental analysis was performed by EA-1110 of CE Instruments Co., Ltd.

Yield: 82.5 % based on aluminium *iso*-propoxide; mp 248 ~ 249 °C; ^{1}H NMR (400.267 MHz, $CDCl_3$ δ in ppm): 8.11 (d, 2H, 4-H in quinoline, $^3J_{4,3}$= 8.4 Hz), 7.45 (t, 2H, 6-H in quinoline, $^3J_{6,5}$= 8.1 Hz, $^3J_{6,7}$= 7.6 Hz), 7.21 (d, 6H, 8-H in Ph, $^3J_{8,9}$= 7.5 Hz), 7.18 (d, 2H, 3-H in quinoline, $^3J_{3,4}$= 8.4Hz), 7.17 (d, 3H, 10-H in Ph, $^3J_{10,9}$= 7.2 Hz), 7.16 (d, 2H, 5-H in quinoline, $^3J_{5,6}$=8.1 Hz), 7.08 (d, 2H, 7-H in quinoline, $^3J_{7,6}$= 7.6 Hz), 7.02 (t, 6H, 9-H in Ph, $^3J_{9,8}$= 7.5 Hz, $^3J_{9,10}$= 7.2 Hz), 2.61 (s, H, Me); ^{13}C NMR(400.267 MHz, $CDCl_3$ δ in ppm): 157.5, 156.3, 139.3, 138.7, 138.6, 134.7, 128.9, 128.6, 127.1, 124.1, 114.0, 112.5, 23.0; Anal. Calcd for $C_{38}H_{31}O_3N_3AlSi$: C, 73.76; H, 5.05; N, 4.53. Found: C, 73.61; H, 5.11; N, 4.49.



SAlq



DCM2

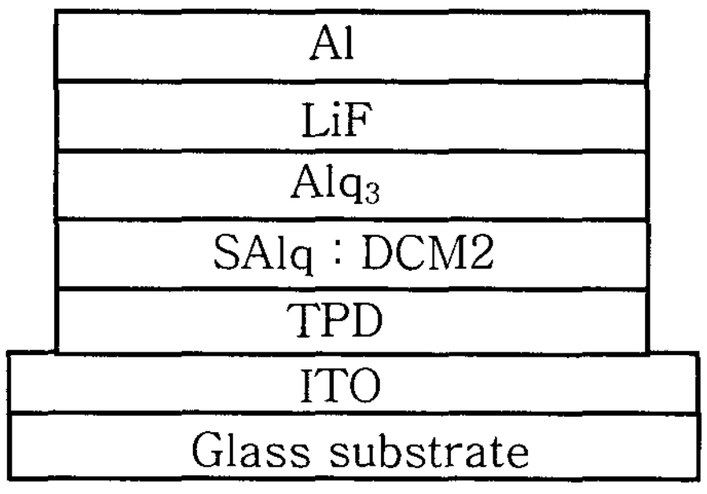


Fig. 1. Molecular structure of bis(2-methyl-8-quinolinolato)$_2$(triphenylsiloxy)aluminium (III) (SAlq) and 4-dicyanomethylene-2-methyl-6-{2-(2,3,6,7-tetrahydro-1H,5H-benzo[I,j]quinolizin-8-yl)vinyl}-4H-pyran (DCM2) and the structure of the device of ITO/TPD/SAlq:DCM2/Alq$_3$/LiF/Al.

Fig. 1 shows the molecular structure of organic materials and the device structure. The device was fabricated by successive vacuum depositions of TPD (50 nm), and SAlq doped with 0.5-% DCM2 (30 nm), Alq_3 (20 nm), LiF (0.5 nm), and Al electrode. ITO substrates with a sheet resistance of about 10Ω/ □ were supplied by Samsung Corning Inc.

The I-V characteristics were measured with a Keithley 236 source-measure unit. The intensity of the EL emission from the devices was simultaneously measured with a Keithley 2000 multimeter equipped with a calibrated Si photodiode or a PMT through an ARC 275 monochromator.

3. Results and discussion

SAlq was synthesized in a high yield of 83 % and a pure form through a homogeneous-phase reaction in *iso*-propanol solution. Because of the strong electrostatic binding force between metal and ligand in SAlq, a good thermal stability is expected. Thermal stability of SAlq is better than blue-emitting materials containing heterocyclic atom such as OXD8 [12] and p-EtTAZ [13]. In general, thermally induced degradation was caused by morphological instability of amorphous organic films. The crystallization or aggregation by intermolecular interaction is a typical type of instability of organic films.

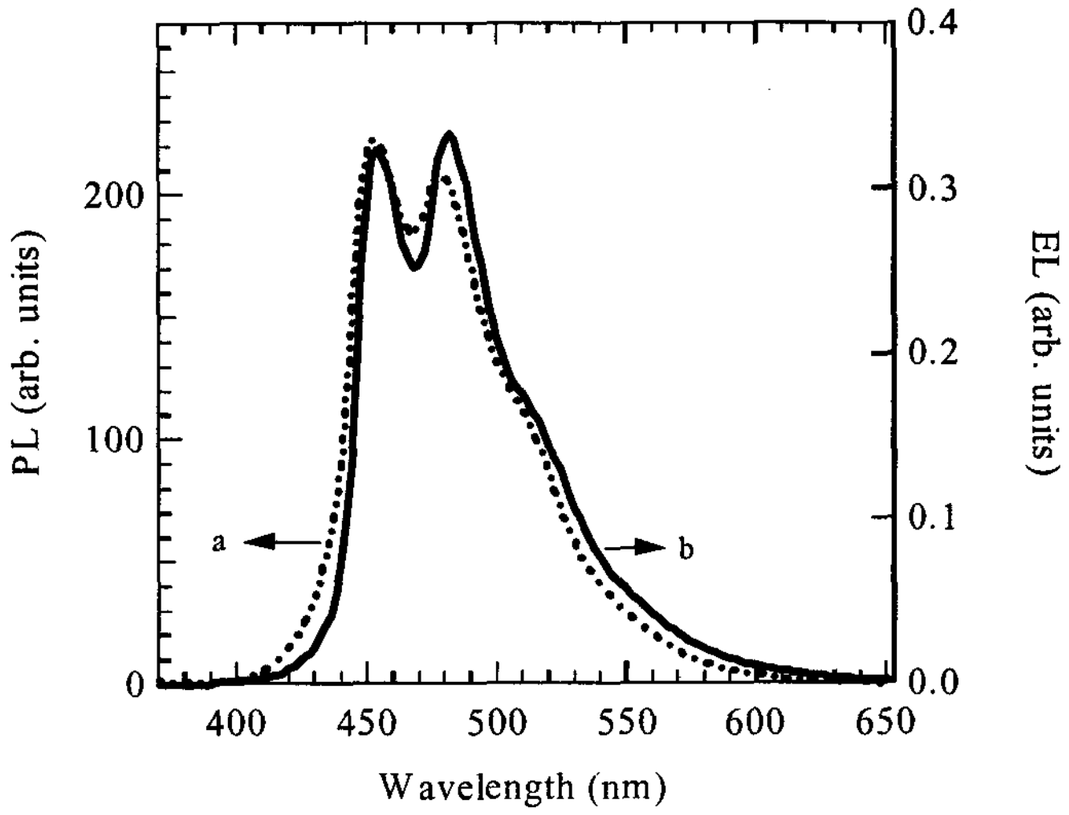


Fig. 2. (a) Photoluminescence spectrum of SAlq film fabricated as the thickness of 100 nm and (b) electroluminescence spectrum for ITO/TPD (50 nm)/SAlq (30 nm)/Alq$_3$ (20 nm)/LiF (0.5 nm)/Al.

Fig. 2 compares the photoluminescence (PL) spectrum of SAlq (100 nm) deposited on quartz substrates and the EL spectrum of ITO/TPD (50 nm)/SAlq (30 nm)/ Alq_3 (20 nm)/LiF (0.5 nm)/Al. Both PL and EL spectra of SAlq show two PL peaks at 452 nm and 477 nm and a shoulder around 510 nm. The brightness and a luminance efficiency of SAlq are higher than those of spirobifluorene [14], BMB-2T [11], and even DPVBi doped with BCzVBi. [15]. The device in Fig. 2 shows an external quantum efficiency of about 3.0 % at 4.3 mA/cm^2 and a luminous efficiency of about 2.5 lm/W, which is one of the highest efficiencies reported in blue-emitting organic EL devices.

Fig. 3 shows the variation of the EL spectra for a device with the structure of ITO/TPD (50 nm)/SAlq:DCM2 (0.5 %, 30 nm)/Alq_3 (20 nm)/LiF (0.5 nm)/Al when the current density is changed from 17 to 174 mA/cm^2. The inset shows the CIE chromaticity diagram with the coordinates of the corresponding EL spectra. It is observed that the device exhibits EL peaks from both SAlq and DCM2. Therefore, the incomplete energy transfer from SAlq to DCM2 offers a possibility to produce white light-emission by control the relative intensity of the EL peaks of SAlq and DCM2.

The relative EL intensities of SAlq and DCM2 were controlled to obtain a balanced white light by adjusting the thickness of SAlq and DCM2 concentration. We find that the device structure shown in Fig. 3 results in relatively stable white light-emission. As the current density increases, the blue part of the EL spectra shown in Fig. 3 increases slightly and the CIE coordinates changes from (0.34, 0.38) at current density of 17 mA/cm^2 to (0.30, 0.36) at 174 mA/cm^2. However, the change in the CIE coordinates with the current density is relatively small compared with the devices of multiple light-emitting layers since the light-emission occurs in a single layer of SAlq:DCM2,

Fig. 4 shows the current-voltage-luminescence (I-V-L) characteristics of the device shown in Fig. 3. The inset shows the external quantum efficiency (QE) and the power efficiency as a function of the current density for the same device. The onset voltage of the light emission is about 3 V. The device has an external QE of about 2.3 % and a luminous efficiency of 2.4 Im/W at luminance of 100 cd/m^2 (bias voltage of 7 V and current density of 1.9 mA/cm^2). The maximum luminance of 23,800 cd/m^2 was achieved at 15 V and 782 mA/cm^2.

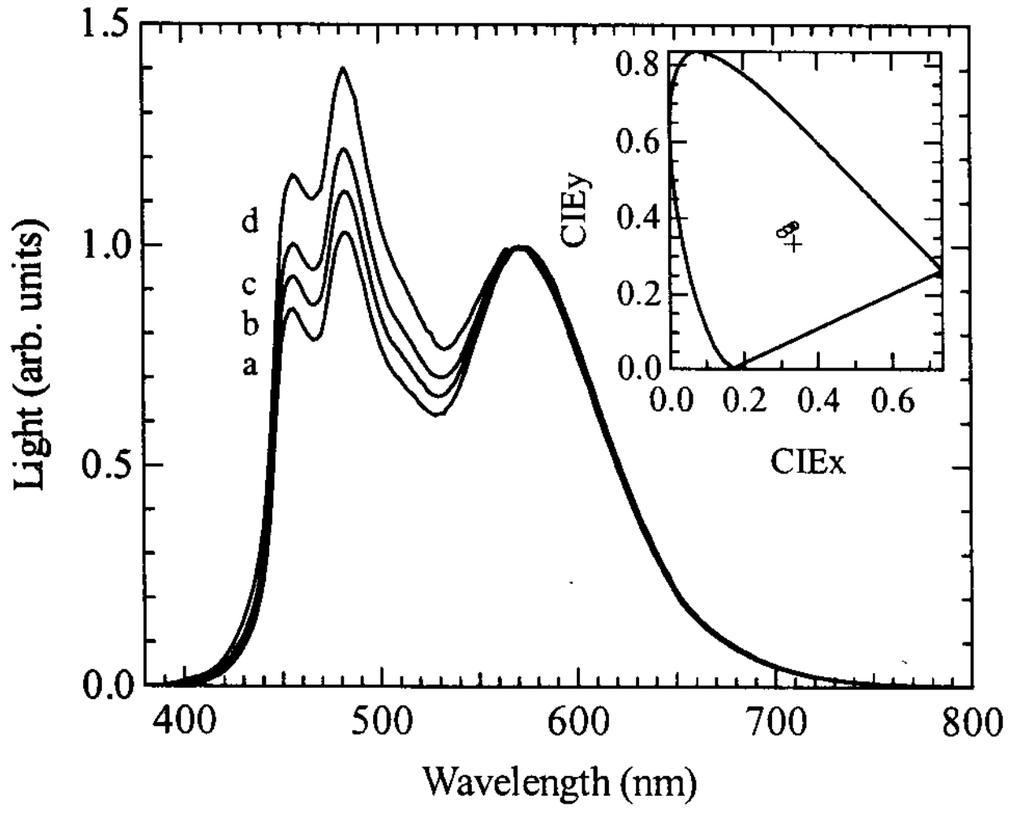


Fig. 3. Variation of the EL spectra with current density for ITO/TPD (50 nm)/SAlq:DCM2 (0.5 %, 30 nm)/Alq_3 (20 nm)/LiF (0.5 m)/Al at 17 (a), 35 (b), 70 (c), and 174 mA/cm^2 (d). The inset shows the CIE chromaticity diagram with the coordinates of the corresponding EL spectra.

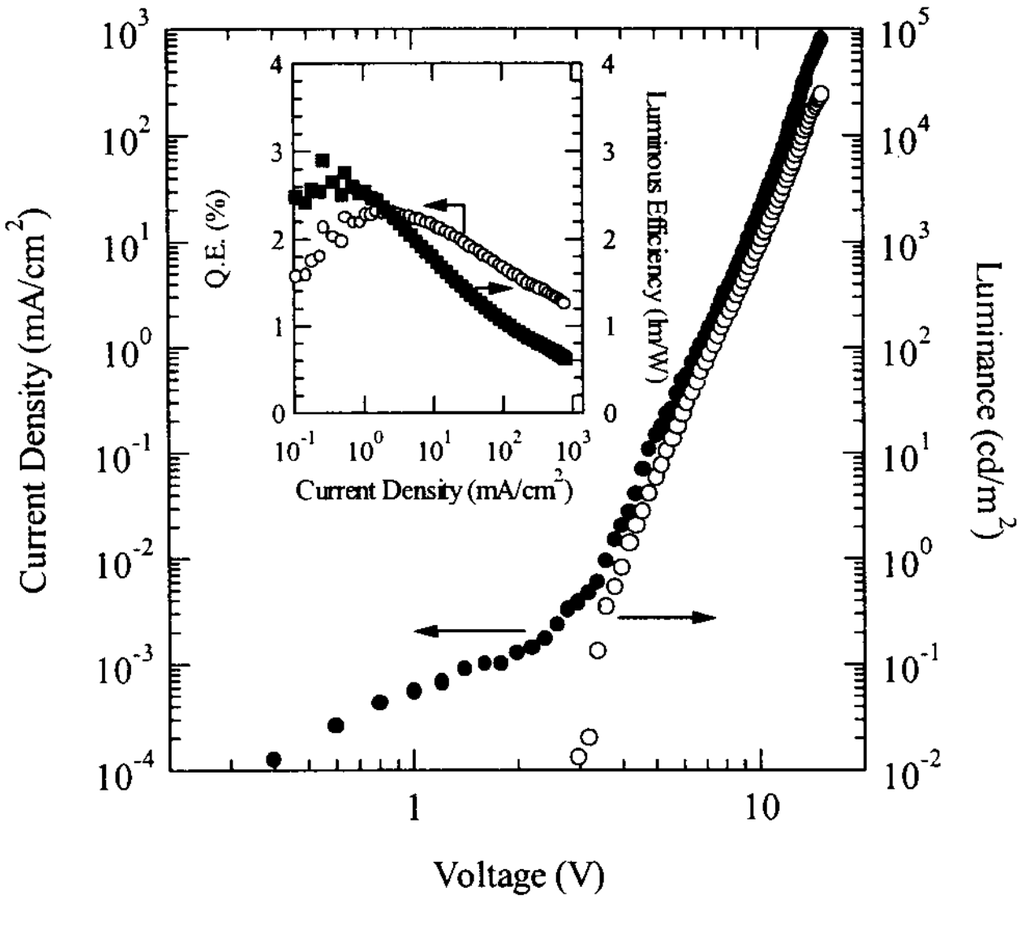


Fig. 4. The current-voltage-luminescence characteristics of ITO/TPD (50 nm)/SAlq:DCM2 (0.5 %, 30 nm)/Alq_3 (20 nm)/LiF (0.5 m)/Al. The inset shows the external quantum efficiency (QE) and the power efficiency as a function of the current density.

4. Conclusion

The blue-emitting material of SAlq was synthesized in high purity and high yield by one-step reaction. The SAlq showed high melting point of 248 ~ 249 °C. The device using SAlq as an active layer showed an external quantum efficiency of about 3.0 % at 4.3 mA/cm^2 and a luminous efficiency of about 2.5 lm/W, which is one of the highest efficiencies reported in blue-emitting organic EL devices. We have shown that the device of ITO/TPD (50 nm)/SAlq:DCM2 (0.5 %, 30 nm)/Alq_3 (20 nm)/LiF (0.5 nm)/Al exhibits white EL emission with an external QE of about 2.3 %, a luminous efficiency of about 2.4 lm/W, and the CIE chromaticity coordinates of (0.32, 0.37) at luminance 100 cd/m^2. The maximum luminance of 23,800 cd/m^2 was achieved at 15 V and 782 mA/cm^2.

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5. Acknowledgements

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