

## Efficient green light-emitting diodes based on substituted triphenylamine derivatives

Periyasamy Murugan\*<sup>1</sup> and Kuo Chu Hwang\*<sup>2</sup>

<sup>1</sup>Department of Chemistry, VEL TECH Dr. RR & Dr.SR Technical University, Avadi, Chennai 600 062, India.

<sup>2</sup>Department of Chemistry, National Tsing Hua University, Hsinchu 30043, Taiwan, ROC.

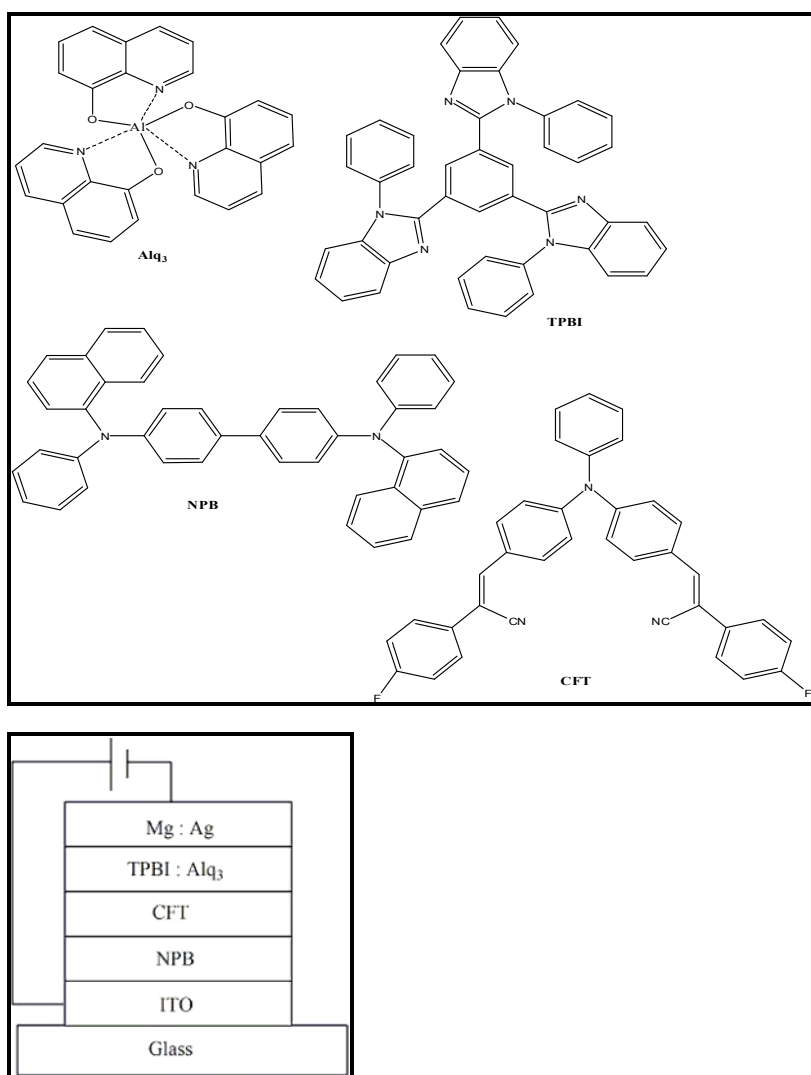
\*Corres.author: pmurugan@veltechuniv.edu.in, kchwang@mx.nthu.edu.tw

**Abstract:** In continuation of our blue light emissive preliminary work, here we report organic electroluminescence (EL) devices based on the green emissive molecule 4,4'-Bis( $\alpha$ -cyano- $\beta$ -fluorostyr- $\beta$ -yl)-triphenylamine(CFT,2). EL devices with a configuration of ITO/NBP(40nm)/CFT(30nm)/TPBI(40nm)/Mg:Ag and ITO/NPB(40nm)/CFT(30nm)Alq(40nm)/Mg:Ag. Where ITO, NPB, TPBI, Alq are indium tin oxide,4,4'-bis[N-(1-naphthyl)-N-phenylamino] biphenyl,2,2',2''-(benzene-1,3,5-triyl)tris [1-phenyl-1H-benzimidazole, tris(8-hydroxy quinolinato) aluminum respectively. Efficient green emission with commission internationale de L' Eclairage (CIE) coordinations ( x = 0.24, y = 0.58) and maximum luminance < 5000 cd/m<sup>2</sup> were obtained from the devices.

**Key words :** OLED, Fluorescence ,Triphenylamine, Imidazole, Pyridine

### Introduction

Now a days bright and efficient organic light – emitting devices (OLED) have attracted considerable interest due to their potential application to flat panel display<sup>1,2</sup>. When constructed a light – emitting compounds for OLED devices three main points should be very important : emission color, emission efficiency and device time.In most of the OLED technology, there are two types of compounds : conjugated organic polymers<sup>3-5</sup> and molecular species such as Alq<sup>6,7</sup>. For full-color display applications, efficient blue, green and red emitters with excellent color purity are required. A great effort has been devoted to improve the efficiencies of the electroluminescent (EL) devices through ingenious device configurations, electrode modification and new efficient materials discoveries<sup>8-10</sup>. In this paper, we wish to report a new class of green light- materials based on CFT (2), possessing an electron withdrawing cyano group and electron donating amino phenyl group and reported the optical properties of CFT was determined.



### Device Structure

Fig.1. General device structure and molecular structures of materials used in the devices.

### Experimental

All melting points are uncorrected. IR spectra were recorded on Shimadzu FT-IR 8300 model and BOMEM (Hartmann&Braun). <sup>1</sup>H and <sup>13</sup>C-NMR spectra were recorded on JEOL GMX 400 MHz, Varian 400 MHz, with CDCl<sub>3</sub> and DMSO-*d*<sub>6</sub> as the solvent with tetramethylsilane as the internal standard. Mass spectra were taken using Hewlett-Packard 5985(70eV), Shimadzu QP 1000A. HRMS(High Resolution Mass Spectra) data were recorded on Thermo Finnigan (Model : MAT 95XL). Absorption and emission measurements were carried out using a Hitachi U-3300 absorption spectrophotometer and a Hitachi F-4500 fluorescence spectrophotometer respectively. The thermal data were carried out using a Seiko SII-EXSTAR 600-DSC-6200 differential scanning calorimeter. The redox potentials of the compound (2) were determined by cyclic voltammetry (CV) using an electrochemical analyzer CHI 600A.

### Synthesis

#### 4,4'-Bis(α-cyano-β-fluorostyr-β-yl)-triphenylamine(2)

To a mixture of 4,4'-diformyl triphenylamine<sup>15</sup>(0.5g , 0.166mmole) and KOH (0.2 g excess) in 30 ml of dry EtOH was added 4-fluorophenylacetonitrile(0.44g ,0.33 mmole) under nitrogen atmosphere. The reaction mixture was stirred at room temperature. After the disappearance of the starting material, the separated

green color solid was filtered and the crude product was washed with EtOH and recrystallized two times with Chloroform-Methanol (4:1) to give the product **2**.

Yield:0.62g (70%) ; m.p:180 -183°C ; IR (KBr):2212,1583,1510,1300,832  $\text{Cm}^{-1}$ ;

$^1\text{H-NMR}$ (400MHz,  $\text{CDCl}_3$ ) $\delta$ =7.08-7.36(m,15H),7.58-7.63(m,4H),7.77-7.80(d,4H,J=12Hz);  $^{13}\text{C-NMR}$ (100MHz,  $\text{CDCl}_3$ ) $\delta$ =164.28,148.74,145.81,

141.20,130.88,130.62,129.87, 127.63, 126.47,125.44,122.97,118.33,116.19,

115.90,107.92.Ms:(m/z%) 535(100),419(10),418(20),417(10),310(5),285(8),267(15).

Anal.calcd for  $\text{C}_{36}\text{H}_{23}\text{N}_3\text{F}_2$  :C,80.73; H,4.32; N,7.84.Found: C,80.64; H,4.42; N,8.17

#### 4,4'-Bis( $\alpha$ -cyano- $\beta$ -chlorostyr- $\beta$ -yl)-triphenylamine( **3**)

Synthesis of compound **3**, was carried out by following the procedure for **2**.

Yield:0.75g(80%) ; m.p:174-176 °C ; IR(KBr):2210,1580,1510,1300,800  $\text{Cm}^{-1}$ ;

UV : $\lambda_{\text{max}}$  492nm ; Flu: $\lambda_{\text{max}}$  520nm( $\text{CH}_2\text{Cl}_2$ ) ; Flu: $\lambda_{\text{max}}$ 542nm(solid state);

$^1\text{H-NMR}$ (400MHz,  $\text{CDCl}_3$ ) $\delta$ =7.13-7.40(m,15H),7.56-7.58(m,4H),7.79-7.81(d,4H,J=8.9Hz); $^{13}\text{C-MNR}$  (100MHz,  $\text{CDCl}_3$ )  $\delta$ =148.91,145.78,141.54,134.79,

133.29,130.79,129.92,129.20,127.90,126.99,126.56,125.57,122.99,118.13,107.88.

MS:m/z(%) 570(29),569(15),567(5),282(5),261(50); Anal.calcd for  $\text{C}_{36}\text{H}_{23}\text{N}_3\text{Cl}_2$  :C,76.05; H,4.07; N,7.39. Found: C,76.24; H,4.42; N,6.87.

#### 4,4'-Bis( $\alpha$ -cyano- $\beta$ -bromostyr- $\beta$ -yl)-triphenylamine( **4**)

Synthesis of compound **4**, was carried out by following the procedure for **2**.

Yield:0.75g(61%) ; m.p: 187-189 °C ; IR(KBr): 2210,1580,1510,1300,800  $\text{Cm}^{-1}$ .

UV : $\lambda_{\text{max}}$  490 nm ; Flu: $\lambda_{\text{max}}$  525nm( $\text{CH}_2\text{Cl}_2$ ); Flu: $\lambda_{\text{max}}$ 545nm(solid state); $^1\text{H-NMR}$ (400MHz,  $\text{CDCl}_3$ )  $\delta$ = 7.13-7.40 (m,15H),7.56-7.58 (m,4H),7.78- 7.82(d,4H,J=8.9Hz);  $^{13}\text{C-MNR}$ (100MHz,  $\text{CDCl}_3$ )  $\delta$ =147.91,144.78,140.54,134.69,

133.99,130.09,129.12,129.50,127.90,126.19,126.16,124.57,122.19,117.13,107.88;

Anal.calcd for  $\text{C}_{36}\text{H}_{23}\text{N}_3\text{Br}_2$  : C,65.77; H,3.53; N,6.39.Found: C,65.34; H,3.42; N,6.87

#### 4,4'-Bis(1-methyl-2,4,6-trioxo-hexahydro-pyrimidin-5-ylideno methyl)-triphenylamine(**5**)

Synthesis of compound **5**, was carried out by following the procedure for **2**.

The mixture of 4,4'-diformyltriphenylamine **1**(0.5g, 0.166mmol) and methyl barbutyric acid (0.48g,0.33mmol) in 15 ml of AcOH/Ac<sub>2</sub>O(12ml:3ml) was heated At 110°C for 8hrs, then the separated solid was filtered and washed with acetone in Several times, to give the red color compound **5**.

Yield: 0.65g (71%) ;m.p: >250°C ; IR(KBr):3096,1732,1702,1645  $\text{Cm}^{-1}$  ;

UV:  $\lambda_{\text{max}}$  480 nm ; Flu:  $\lambda_{\text{max}}$  605 nm(MeOH) ; Flu :  $\lambda_{\text{max}}$  624 nm(solid state);

$^1\text{H-NMR}$ (400MHz,  $\text{DMSO}-d_6$ )  $\delta$ =3.14(s,3H,CH<sub>3</sub>),3.16(s,3H,CH<sub>3</sub>),7.07-7.18 (m,7H),7.45-7.54(m,4H),7.77 – 7.91 (m,4H);  $^{13}\text{C-NMR}$ (100MHz,  $\text{DMSO}-d_6$ ) $\delta$ =162.55,

161.55,160.97,154.65,154.04,150.47,149.93,144.56,136.65,130.38,126.76,121.46,121.37,116.02,27.72,27.13;

MS:m/z(%) 549(20),548(18),547(5),408(5),376(6), 292(18),290(10); Anal.calcd for  $\text{C}_{30}\text{H}_{23}\text{N}_5\text{O}_6$  : C,65.56; H,4.21; N,12.74.Found: C,65.24; H,4.42; N,12.25.

**Diethyl 4-(2-(1-pyrenyl)-1-ethynyl)-2,6-pyridine dicarboxylate (8)**

A mixture of **6**<sup>14</sup>(1.5g, 0.5 mmol) palladium (II) acetate (23mg, 0.1mmol), triphenyl phosphine (54mg, 0.2mmol), copper (I) iodide (38mg, 0.2mmol) and 1-ethynyl pyrene **7**. (1.1g, 0.5mmol) in dry triethylamine (25ml), dry toluene (20ml) was deaerated with nitrogen and heated at 90-95°C for 9 hours. After the reaction was complete, the mixture was evaporated in vacuo. The residue was dissolved in chloroform (120ml), the solution was washed with water (3x20ml) and dried with anhydrous sodium sulphate. Removal of solvent by evaporation yielded a crude material which was purified by column chromatography on silica using hexane / chloroform 1:4 as eluent to give compound **8** in 1.2g (54% yield); m.p : 210-212°C ; IR (KBr) : 1598, 1721, 2196 cm<sup>-1</sup> ; <sup>1</sup>HNMR(400 MHz, CDCl<sub>3</sub>) δ = 1.47-1.50 (t, 6H, CH<sub>3</sub>, J = 5.9 Hz), 4.51-4.55 (q, 4H, CH<sub>2</sub>, J = 5.3Hz), 8.02-8.07(m, 2H), 8.13-8.16(m, 2H), 8.21-8.28(m, 4H), 8.48(s, 2H), 8.59-8.61(d, 1H, J=8Hz); <sup>13</sup>CNMR(100MHz, CDCl<sub>3</sub>) δ = 14.26, 62.58, 91.01, 96.15, 124.13, 124.40, 124.60, 124.99, 126.17, 126.24, 126.52, 127.17, 129.10, 129.41, 130.13, 131.14, 132.42, 134.48, 148.93, 164.40; MS : (m/z %) : 447(100), 375(50), 327(25), 301(80), 250(20), 224(5), 223(6); HRMS: 447.4904 C<sub>29</sub>H<sub>21</sub>NO<sub>4</sub> found 447.1455; Anal. calcd for C<sub>29</sub>H<sub>21</sub>NO<sub>4</sub> C : 77.83 ; H: 4.72 ; N : 3.12 ; Found C : 77.41 ; H : 4.36 ; N : 3.57.

**Diethyl 4-(2-(9-anthranyl)-1-ethynyl)-2,6-pyridine dicarboxylate (9)**

Synthesis of compound **9** was carried out by following the procedure for **8**.

1.2g (54% yield); m.p : > 250°C ; IR (KBr) : 1598, 1721, 2196 cm<sup>-1</sup> ;

<sup>1</sup>HNMR(400 MHz, CDCl<sub>3</sub>) δ = 1.47-1.50 (t, 6H, CH<sub>3</sub>, J = 5.9 Hz), 4.51-4.55 (q, 4H, CH<sub>2</sub>, J = 5.3Hz), 7.60-7.62(d, 4H, J = 8Hz), 7.86(d, 2H, J = 2H) , 7.95-7.97(d, 2H, J = 8Hz), 8.13-8.16(m, 2H), 8.54(s, 1H); <sup>13</sup>CNMR(100MHz, CDCl<sub>3</sub>) δ = 14.26, 62.58, 91.01, 96.15,

124.42, 124.65, 125.58, 125.90, 127.30, 129.46, 131.37, 131.68, 132.36, 135.10, 138.18, 144.65; HRMS: 423.1471 C<sub>27</sub>H<sub>21</sub>NO<sub>4</sub> found 423.4532.

**2-Anthranyl-4,5-diphenyl-1-propyl imidazole (13)**

The mixture of imidazole **11**<sup>11</sup>(0.5g, 1.2mmol), n-propyl bromide (0.15g, 1.2mmol), NaH(0.05g, 2.0mmol) and dry DMF (10ml) was heated (70-75 °C) under nitrogen atmosphere for 20 hours, then cool the reaction mixture, poured in to ice water, the separated solid was filtered, dried and recrystallized from CHCl<sub>3</sub>-MeOH (1:1).

Yield: 0.310g (56%). M.p: 208-210 °C . UV: λ<sub>max</sub> 385 nm (MeOH) ; Flu: λ<sub>max</sub> 460 nm (MeOH); <sup>1</sup>H-NMR (400MHz, CDCl<sub>3</sub>): δ = 8.58(s, 1H), 8.08-

8.01(m, 2H), 7.78-7.81(m, 2H), 7.65-7.56(m, 2H), 7.55-7.24(m, 8H), 7.23-7.11(m, 4H), 3.47-3.43(t, 2H, J=8Hz), 1.08-1.00(m, 2H), 0.29-0.25(t, 3H, J=8Hz); <sup>13</sup>C-NMR(100MHz, CDCl<sub>3</sub>) δ = 144.69, 138.18, 135.01, 132.35, 131.97, 131.60, 131.37, 129.46, 129.23, 128.91, 128.81, 128.32, 127.05, 126.93, 126.47, 126.19, 125.65, 125.42, 46.61, 23.97, 11.11; Anal calcd for C<sub>32</sub>H<sub>26</sub>N<sub>2</sub> : C, 87.43 ; H, 6.19 ; N, 6.37. Found C, 87.58; H, 6.35 ; N, 6.43.

**2-Pyrenyl-4,5-diphenyl-1-propyl imidazole (14)**

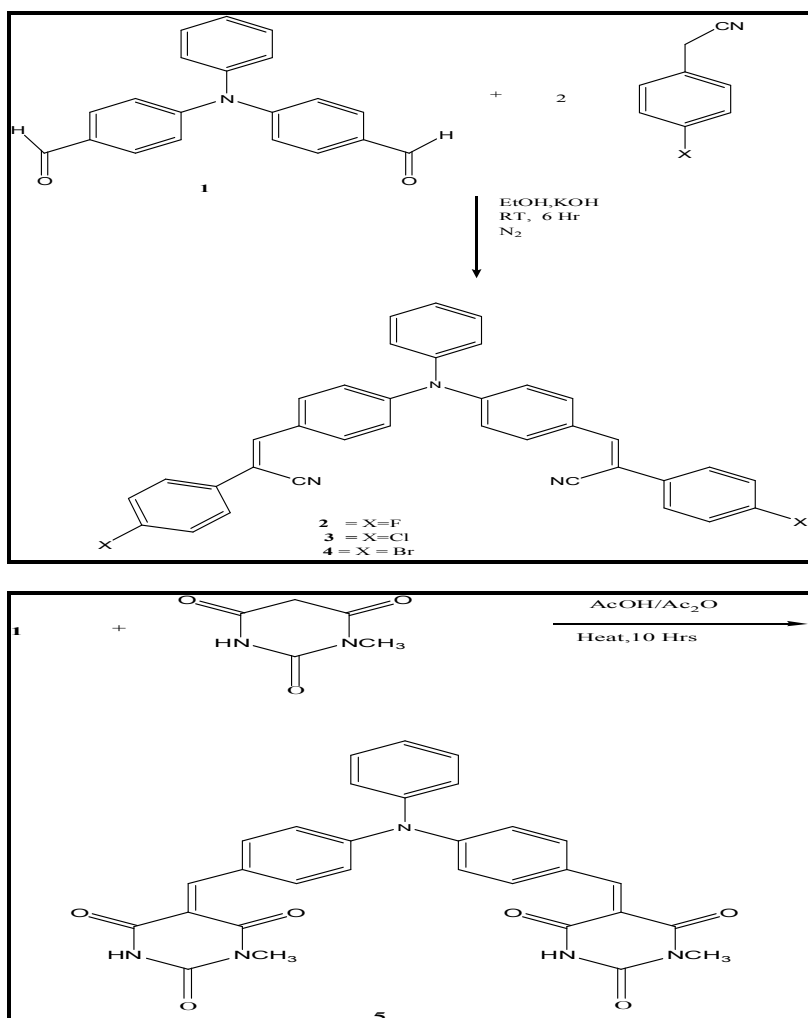
The mixture of imidazole **12**<sup>11</sup>(0.5g, 1.2mmol), n-propyl bromide (0.15g, 1.2mmol), NaH(0.05g, 2.0 mmol) and dry DMF (10ml) was heated (70-75 °C) under nitrogen atmosphere for 20 hours, then cool the reaction mixture, poured in to ice water, the separated solid was filtered, dried and recrystallized from CHCl<sub>3</sub>-MeOH(1:1).

Yield: 0.310g (56%); M.p: >250 °C ; UV: λ<sub>max</sub> 350 nm (MeOH); Flu: λ<sub>max</sub> 457 nm (MeOH); <sup>1</sup>H-NMR (400MHz, CDCl<sub>3</sub>) δ = 8.30-8.24(m, 4H), 8.10-8.01(m, 5H), 7.59-7.47(m, 5H), 7.21-7.15(m, 5H), 3.47-3.43 (t, 2H, J=8Hz), 1.08-1.00(m, 2H), 0.29-0.25(t, 3H, J=8Hz); <sup>13</sup>C-NMR(100MHz, CDCl<sub>3</sub>) δ = 146.41, 137.56, 134.82, 131.66, 131.39, 130.98, 130.79, 130.48, 129.11, 128.83, 128.36, 128.24, 128.03, 128.00, 127.82, 126.58, 125.98, 125.93, 125.89, 125.25, 125.16, 124.69, 124.59, 124.28, 46.64, 23.47, 11.11; Anal calcd for C<sub>32</sub>H<sub>27</sub>N<sub>2</sub> : C, 87.43 ; H, 6.19 ; N, 6.37. Found C, 87.58; H, 6.35 ; N, 6.4

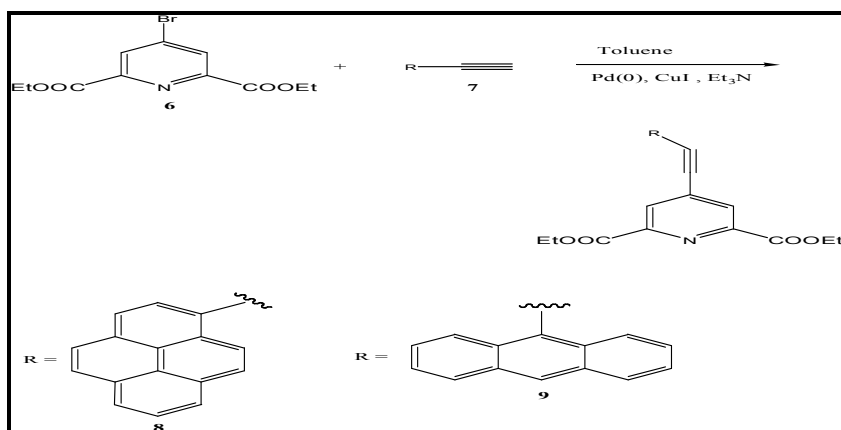
## Results and Discussion

The reaction of 4,4'-diformyl triphenyl amine(1) and halo substituted phenyl aceto nitrile with KOH led to green color compounds 2- 4, similarly the dialdehyde and methyl barbutyric acid was refluxed with mixture of AcOH/Ac<sub>2</sub>O to furnish red color compound 5 (Scheme I). Sonogashira coupling reaction of 6 with the acetylenic compounds 7 was carried out to obtain the tridentate pyridine compounds 8, 9 in moderate yield (Scheme – II). Compound 10 was treated with aromatic aldehydes in the presence of acetic acid and ammonium acetate to furnish the substituted imidazoles<sup>11</sup> 11, 12, further treatment of compounds 11, 12 with propyl bromide was done in presence of sodium hydride in DMF to afford the imidazole derivatives 13, 14 (Scheme – III). The structure of the compounds was confirmed by IR, <sup>1</sup>H and <sup>13</sup>C NMR, Mass and elemental analysis. All the melting points are not corrected. The thermal data of CFT (2) was obtained from a Seiko SII-EXSTAR 600-DSC-6200 differential scanning calorimeter. CFT (2) was heated at 20°C/min under helium atmosphere until complete melting and then cooling rapidly. A second heating scan was performed at the same rate to detect the glass transition temperature (*T<sub>g</sub>*), the crystallization temperature (*T<sub>c</sub>*) and the melting temperature (*T<sub>m</sub>*)<sup>12</sup>.

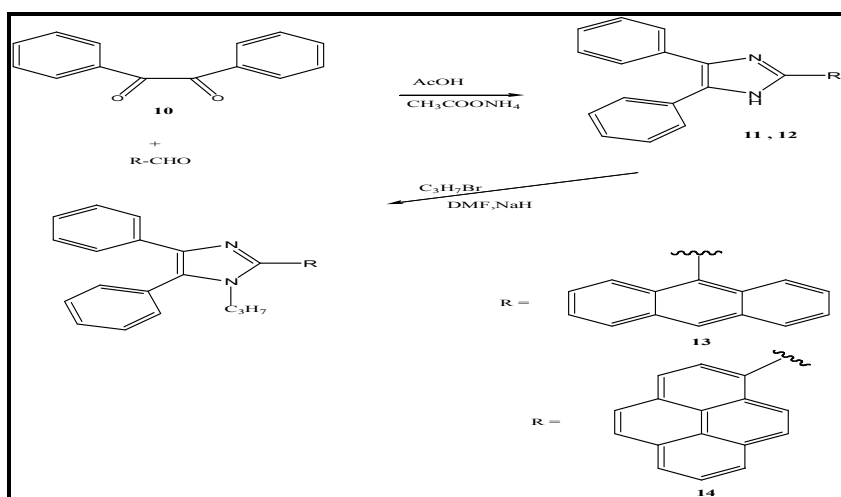
Fluorescence quantum yield ( $\Phi_f$ ) of the CFT (2) was determined by comparing with that of coumarin -1 in ethyl acetate<sup>13</sup> ( $\Phi_f = 0.31$ ) at ambient temperature. The redox potentials of the compound (2) were determined by cyclic voltammetry (CV) using an electrochemical analyzer CHI 600A. CV measurements were carried out in dry Dichloromethane solutions of the compounds with 0.01 M tetra-n-butyl ammonium Hexafluorophosphate as the electrolyte medium. A conventional three – electrode Cell system was used with glassy carbon, platinum wire and Ag/AgCl as working, Counter and reference electrodes, respectively. The HOMO level was calculated from the oxidation-reduction potentials, which were obtained from the cyclic voltammetry curves. The LUMO level was estimated based on the HOMO energy levels and the lowest – energy absorption edge of the UV-Vis absorption spectrum<sup>14</sup>.



## Scheme I



## Scheme –II



## Scheme – III

## Fabrication and measurement of light – emitting devices

The OLED devices containing CFT was fabricated by vacuum deposition. The substrate was an indium tin oxide (ITO)-coated glass with a sheet resistance 25Ω per square. The ITO – coated glass was pretreated according to a regular chemical cleaning using detergent, deionized water and alcohol in sequence, followed by ozone cleaning. The thermal evaporation of organic materials was carried out at chamber pressure of 10<sup>-6</sup>Torr. The dopant and host materials were co-evaporated from different heating sources. The cathode consisting of Mg:Ag(10:1;55nm) was deposited by co-evaporation of Mg and Ag metals at a deposition rate of 5-7 and 0.5-0.7 Å/s, respectively. A thick silver –capped layer (100nm) was deposited over the cathode by evaporation of silver metal with a rate of 3-4 Å/s. The deposition rate of organic compounds is in between 1 and 2 Å/s. The effective area of the emitting diode was 9.00mm<sup>2</sup>. Device were constructed with 40-, 30-, 40-nm thickness of NPB,CFT,TPBI or Alq<sub>3</sub> respectively. Current-Voltage-luminescence (*I-V-L*) measurements were made simultaneously using a Keithly 2400 source meter and a Newport 1835-C optical meter equipped with a Newport 818-ST silicon photodiode. The electroluminescence spectra were recorded on a Hitachi F-4500 fluorescence spectrophotometer. The measurements were at room temperature under ambient conditions.

The physical properties of CFT shown in Table-I. The strong fluorescence exhibited by the CFT dye both in solution and solid state. A three- layer device configuration was studied, where the CFT was used as the emission layer and NPB was the hole transporter layer. Both Alq and TPBI was used as the electron transporter. Both the configuration of devices ITO/NPB(40nm)/CFT(30nm)/TPBI(or Alq)(40nm)/Mg:Ag, were gave the bright green emission. Fig –I depicts the normalized EL spectra of above two devices. The CFT gave a emission maximum at 521 nm in the case of TPBI as electron transporter. In the case of Alq , the CFT gave a emission maximum at 525 nm. The *I-V-L* plots for both ITO/NPB(40nm)/CFT(30nm)/TPBI(or Alq)(40nm)/Mg:Ag are shown in Fig-III and Fig –IV. Efficient green emission with commission internationale de L' Eclairage (CIE)

coordinations ( $x = 0.24$ ,  $y = 0.58$ ) and maximum luminance  $< 5000 \text{ cd/m}^2$  were obtained from the devices (Fig-V).

**Table- I**  
**Physical properties of CFT**

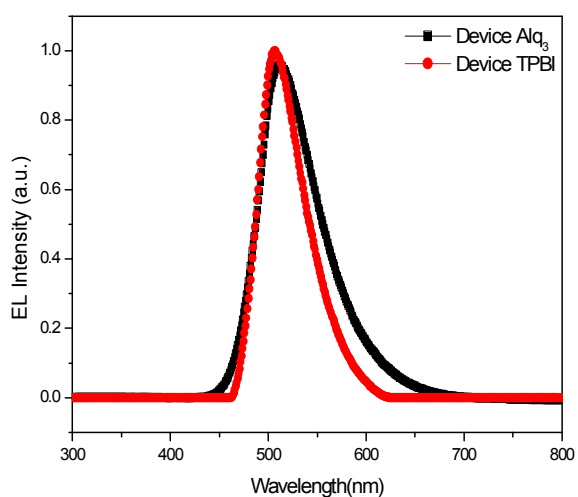
Compound	$T_m/T_g(^{\circ}\text{C})^a$	$\lambda_{\text{abs}}^b(\text{nm})$	$\lambda_{\text{em}}^b(\text{nm})$	$\lambda_{\text{em}}^c(\text{nm})$	$\phi_f^d$	HOMO/LUMO( $\text{e}^{\text{V}}$ )
CFT	182/95	490	510	540	0.31	5.51/3.12

<sup>a</sup> from DSC measurements.

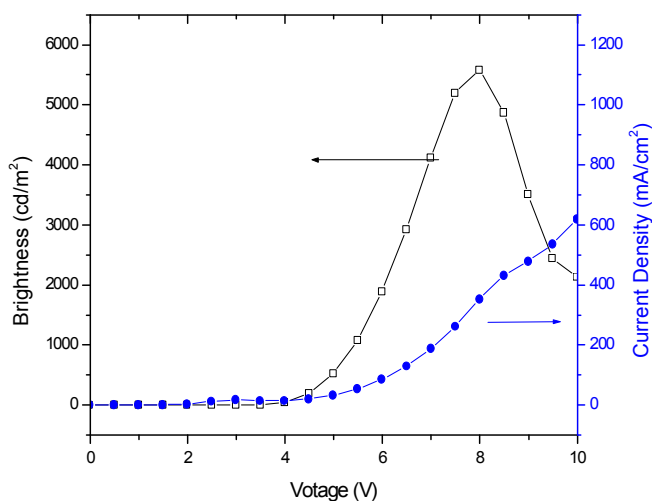
<sup>b</sup>In  $\text{CH}_2\text{Cl}_2$

<sup>c</sup> In the solid state.

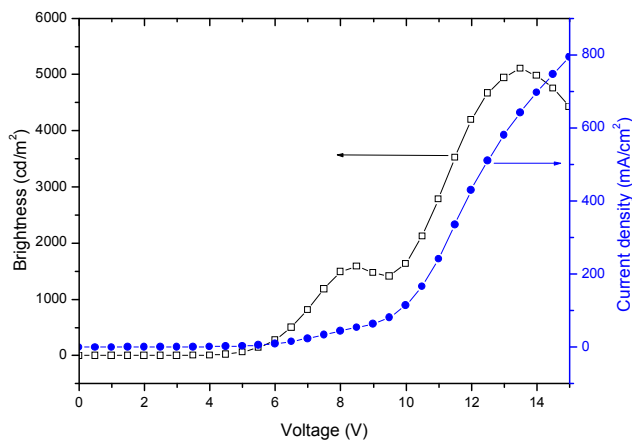
<sup>d</sup>PL quantum yield measured in a Ethyl Acetate solution, Coumarin -1 as a reference standard.



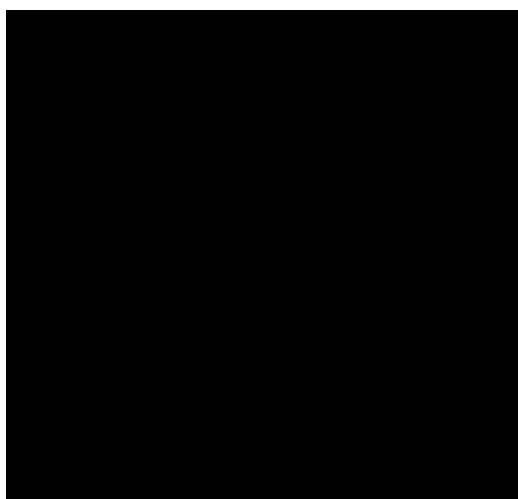
**Fig – II** : Normalized EL spectra of CFT in ITO/NPB(40nm)/CFT(30nm)/TPBI(or  $\text{Alq}_3$ )(40nm)/Mg:Ag at 6-15 V



**Fig -III:** *I-V-L* measurement for devices for CFT device ITO/NPB(40nm)/CFT(30nm)/TPBI(40nm)/Mg:Ag (10:1; 55nm)/Ag(100nm)



**Fig - IV :** *I-V-L* measurement for devices for CFT device ITO/NPB(40nm)/CFT(30nm)/Alq<sub>3</sub>(40nm)/Mg:Ag (10:1; 55nm)/Ag(100nm)



**Fig -V:** The Commission Internationale de L'Eclairage (CIE) chromaticity coordinates of devices CFT.

## Conclusion

In summary, we report simple and straightforward synthesis of different substituted triphenyl amine derivatives **2-5**, Pd- catalyst mediated synthesis of tridentate pyridine compounds **8,9** and highly substituted imidazole derivatives **13,14**. Compound **2**(CFT) shows efficient green light emission (EL) with commission internationale de L' Eclairage (CIE) coordinations ( $x = 0.24$ ,  $y = 0.58$ ) and maximum luminance  $< 5000$  cd/m<sup>2</sup> were obtained from the devices.

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