Deep blue organic light-emitting diodes of 1,8-diaryl anthracene

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Abstract. We report on the optimization of organic light emitting diode (OLED) devices using 1,8-di-(4-trifluoromethylphenyl)-anthracene (**CF3-DPA**) as the active emissive layer. **CF3-DPA** emits in the deep blue region with an emission peak at 432 nm in solution which showed a slight red shift in thin films. **CF3-DPA** has high reported fluorescence quantum efficiency, ~67%, as compared to 9,10-diphenyl anthracene (9,10-DPA). We optimized the OLED devices with different hole transporting layers (HTLs). Bilayer devices formed with *N,N'*-di(1-naphthyl)-*N,N'*-diphenyl-(1,1'-biphenyl)-4,4'-diamine (NPD) as the HTL gave a reasonable light output. We observed that trilayer or multilayer devices with the inclusion of poly(3,4-ethylene dioxythiophene)-poly(styrene sulfonate) (PEDOT:PSS) and/or copper phthalocyanine as an additional HTL reduced the turn on voltage by ~5 to 9 V, though the brightness of the light emission also decreased. Including suitable carrier (electron or hole) transporting layers like 2, 2',2''-(1,3,5-Benzinetriyl)-tris(1-phenyl-1-H-benzimidazole) (TPBi) and 4,4'-Bis(*N*-carbazolyl)-1,1'-biphenyl (CBP) increases the efficiency of the devices. From our studies, we conclude that though NPD/**CF3-DPA** interface is crucial for light emission, the performance of the devices is limited by the mismatch of the hole and electron mobilities and the low internal quantum efficiency of **CF3-DPA** in the solid state. Devices having ITO/NPD/**CF3-DPA**/TPBi/LiF-Al geometry were observed to be the most efficient.

Keywords. Anthracene derivative; blue organic light emitting device; hole transporting layer.

1. Introduction

Organic light-emitting diodes (OLEDs) have been a focus of recent research and development due to their potential for use in the new type of large-area, flexible flat-panel displays for mobiles and televisions.¹⁻⁴ However, one of the major concerns in the fabrication of full-colour OLED displays is the equal performance of the three primary colors of red, green and blue. It has been successfully demonstrated that efficient red or green OLEDs can be made by using vacuum-deposition of small molecules of organic electroluminescent materials or spin-coating of conjugated polymers.⁴ However, since blue organic materials have a larger band gap, they have a lower affinity for the electron from the cathode in

OLEDs. Therefore, the performance of blue emitters is usually inferior to that of green or red emitters. Though the pure blue material was commercialized for the first active display in 2004, the external quantum efficiency (EQE) of the blue devices has been showing a steady increase.^{5,6} In spite of being able to obtain a reasonably high EQE, long-term stability and color purity of the blue OLEDs require improvement and efforts are on to find newer blue emitting materials with desired properties.⁷

Anthracene in solution exhibits fluorescence in the blue spectral region with high quantum efficiency making it and its derivatives good candidates for efficient blue emitters.⁸ However, they are prone to aggregation in thin films, owing to their highly planar structure.^{9,10} Aggregation of anthracene in thin films lead to the quenching of the fluorescence and redshift in

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its emission maxima. Among anthracene derivatives, 9,10-diphenylanthracene (DPA) might be thought to be a promising material as a blue emitter since DPA has a photoluminescence (PL) quantum yield close to unity. However, a weak blue electroluminescence (EL) emission, about 0.09 cd/m^2 at 100 mA/cm^2 , from bilayer devices consisting of a DPA emitting layer and a diamine hole transporting layer (HTL) layer have been reported.⁸ Thereafter, it has been shown that using multi-layer geometry for the devices with different combinations of HTL and electron transporting layer (ETL) or doping DPA with bathocuproine (BCP) improves the luminous efficiency marginally. About a couple of years back, a new series of undoped 9,10-DPA-based blue fluorophores was synthesized with bulky substituents which showed much higher EQE of 4.5%¹¹ as compared to unsubstituted DPA (EQE-1.5%) or doped 9,10-DPA (EQE $\sim 2\%$).¹² Subsequently, several attempts were made to improve the emission and thin film properties of anthracene derivatives.^{13–18} It was demonstrated that the film forming properties of DPA can be improved by introducing bulky aryl substituents at the 2- and 6-positions of anthracene (instead of the 9- and 10-positions).¹⁹ Recently, the synthesis of derivatives of the 1,8-diaryl anthracene have been reported.^{20,21} These anthracene derivatives were reported to emit in the blue region and showed high fluorescence quantum efficiency. One of the derivatives also exhibited surfactant-assisted nano-self-assembly forming nano-ribbons and nano-plates. It was proposed that these compounds with their blue emitting properties and compatible HOMO–LUMO energy levels would be suitable to form OLEDs.

In this paper, we report on the morphological and electro-optical properties of thin films formed of an anthracene derivative, 1,8-bis-(4-trifluoromethylphenyl)-anthracene (**CF3-DPA**), synthesized according to literature.²⁰ Fluorescence peak at 432 nm, high quantum efficiency ($\sim 67\%$ with respect to 9,10-DPA) and good thin film forming properties of **CF3-DPA** allow us to study it in OLED applications. We fabricated a series of devices with different structures having different HTLs and studied the effect of them on the turn on voltage (V_{ON}) and luminance of the devices. We observed that *N,N'*-Di(1-naphthyl)-*N,N'*-diphenyl-(1,1'-biphenyl)-4,4'-diamine or NPD was the most suited HTL with respect to light output. The device of ITO/NPD/**CF3-DPA**/LiF-Al gave blue light with CIE chromaticity of (0.14, 0.04). However, the turn on voltage (V_{ON}) was quite high, about 13 V. We observed that multilayer devices having a layer of poly(3,4-ethylene dioxythiophene) poly(styrene sulfonate) (PEDOT:PSS) before NPD reduces the V_{ON} considerably, though the

light output is reduced. The efficiency of the devices could be enhanced by using a suitable carrier (electron and/or hole) transporting layers like TPBi and CBP. Devices having ITO/NPD/**CF3-DPA**/TPBi/LiF-Al geometry were observed to be the most efficient at a current density of 50 mA/cm^2 while devices having ITO/PEDOT:PSS/**CF3-DPA**/TPBi/LiF-Al geometry were most efficient at a low current density of 1 mA/cm^2 , though they were not very stable. Thus, we show that 1,8-diaryl anthracene derivatives are good candidates for deep blue OLEDs.

2. Experimental

Trifluoromethyl substituted 1,8-diaryl anthracene was synthesized using the Suzuki–Miyaura coupling reaction of 1,8-dichloroanthracene using the catalyst Pd-PEPPSI-iPr. The details of the synthesis are reported earlier.²¹ The structure of the synthesized compound is shown in Figure 1(a). The photo-physical characterization of the synthesized compound was carried out in solution as well as in solid thin films using SHIMADZU UV180 spectrophotometer and HORIBA FLUOROLOG 3 fluorimeter. The morphology of the thin films was studied by atomic force microscope (AFM).

All OLED devices were grown on ITO coated glass with a surface resistivity of $15\text{--}25 \text{ } \Omega/\text{sq}$ (commercially brought from Sigma Aldrich) which were patterned to make four devices on a substrate of $2 \times 1 \text{ cm}^2$. The ITO substrates were cleaned properly with soap solution, distilled water, propanol and trichloroethylene. Finally, ozone surface treatment was given by keeping the substrates under UV light for 30 min which is known to reduce the work function of ITO.²² Vacuum deposition was used in the preparation of all the OLED devices. Apart from PEDOT:PSS all the other materials were thermally evaporated in a base vacuum of $5 \times 10^{-6} \text{ mbar}$. PEDOT:PSS was spin-coated at 8000 rpm for 40 s and heated at 160°C for 30 min to give a $\sim 40 \text{ nm}$ thick layer.²³ The thickness of each layer deposited by thermal evaporation was measured using an *in situ* quartz crystal monitor. In addition, the thickness of each material was independently calibrated *ex situ* using a surface profilometer. The final electrode or cathode of the devices formed by LiF/Al was deposited using a shadow mask that defined the active area of the OLED devices which were in the range of $6\text{--}25 \text{ mm}^2$. The thickness of LiF was $\sim 1 \text{ nm}$ while that of Al was $\sim 160 \text{ nm}$ for all the devices. The electro-luminance spectroscopy of the devices were measured using a set up consisting of a Bausch and Lomb 350–750 nm monochromator and Hamamatsu R212 photomultiplier tube (PMT) as the detector. The current–voltage (I–V) characteristics were measured using the 2400 Keithley source meter at $\lambda = 439 \text{ nm}$ and the luminance–voltage (L–V) spectra were simultaneously recorded using lock-in detection. The lock-in signal was converted to light units using the conversion factors which were estimated using a calibrated red LED.

3. Results and Discussion

3.1 Characterization of CF3-DPA

Photophysical studies of **CF3-DPA** were carried out in solution and thin films using absorbance and fluorescence spectroscopy. Absorption maxima was obtained at 394 nm and the emission maxima at 432 nm in toluene. The quantum yield in solution was about 67% taking 9,10-DPA as the reference.²⁰ The emission was measured in both spin-coated and evaporated thin films. The emission peak showed a bathochromic shift of 7 nm as compared to the solution (Figure 1(b)). This marginal bathochromic shift indicates little aggregation in thin films of **CF3-DPA**.²⁴

Morphology of thin films of the active emissive layer plays a crucial role for OLED devices. Surface morphology of **CF3-DPA** thin films, which were both spin coated and grown by thermal evaporation, was studied by AFM. The AFM images of these films are shown in Figure 1(c) and (d). The surface roughness of the evaporated films was less than that of the spin-coated ones and was of the order of ~ 2 nm. Thus, thermally evaporated films would be more suited to form OLED devices as smoother interfaces are expected. The HOMO and LUMO energy levels were estimated using the

cyclic voltammetry and were found to be -5.7 and -2.8 eV, respectively. The band gap for **CF3-DPA** was obtained as 2.9 eV which corresponds to 428 nm. The E_{LUMO} is very close to the widely used organic green emitter, tris-(8-hydroxyquinolino)-aluminium (Alq₃) whose OLEDs have been optimized to give a high external quantum efficiency of $\sim 12\%$ at 100 Cd/m^2 .¹²

3.2 Characterization of CF3-DPA OLED devices

PEDOT: PSS is a widely used HTL in OLEDs. Since this has E_{HOMO} closer to that of the plasma treated ITO, hole injection efficiency is expected to be high²⁵ which might result in a low turn on voltage (V_{ON}). Bilayer devices, ITO/PEDOT:PSS/**CF3-DPA**/LiF-Al, whose energy band diagram is shown in Figure 2(a) were fabricated. The thickness of PEDOT:PSS and **CF3-DPA** layers were ~ 50 nm each. Figure 2(c) shows the plot between current density (J) and bias voltage (V).

A non-zero current density (J) was obtained at ~ 2.5 V and a distinct change of slope at ~ 5.0 V while no light output was detected at this bias voltage (Figure 2(d)) which could be due to the non-recombination of the carriers. To improve hole transport, we studied the effect of adding NPD, another HTL, in the device. Interestingly, for these trilayer devices, ITO/PEDOT:PSS/NPD/**CF3-DPA**/LiF-Al (energy band diagram is shown in Figure 2(b)), the J–V curve showed a non-zero value of J at a voltage of 2.5 V (Figure 2(c)). This was similar to the bilayer device. However, a further rise of J was observed at a voltage of 7.5 V at which light was also detected implying that $V_{\text{ON}} = 7.5$ V for this device. At $J = 50 \text{ mA/cm}^2$, the light output was quite low $\sim 2 \text{ Cd/m}^2$ as shown in Figure 2(d).

We tried to find the most optimal choice of HTL so that the V_{ON} is minimized and the light output is maximized for the devices. As the light output was quite low in PEDOT:PSS/NPD based devices, we studied the effect of another well-known HTL, copper phthalocyanine (CuPc) on the device performance which is also known to enhance hole injection efficiency from ITO.²⁶ Since, the NPD/**CF3-DPA** interface was important for light output, we made trilayer and multilayer devices retaining this interface. Devices with the following geometries ITO/CuPc/NPD/**CF3-DPA**/LiF-Al and ITO/CuPc/PEDOT:PSS/NPD/**CF3-DPA**/LiF-Al (energy band diagrams given in Figure 3(a) and (b), respectively) were made where the thickness of CuPc was kept at ~ 40 nm. The thicknesses of PEDOT:PSS and **CF3-DPA** were kept at ~ 50 nm as the previous devices. Figure 3(c) shows the J–V for both these devices and Figure 3(d) shows the L–V for the same two devices. Multilayer devices (Figure 3(b)) with PEDOT:PSS had

Figure 1. (a) The structure of trifluoromethyl substituted 1,8-diaryl anthracene (**CF3-DPA**); (b) Emission spectra of **CF3-DPA** in solution (black square), evaporated thin film (blue star) and spin coated thin film (red circle); (c) and (d) AFM images ($4.8 \mu\text{m} \times 4.8 \mu\text{m}$) of the spin-coated and evaporated thin films, respectively.

Figure 2. (a) Energy band diagram of ITO/PEDOT:PSS/CF3-DPA/LiF-Al device; (b) Energy band diagram of ITO/PEDOT:PSS/NPD/CF3-DPA/LiF-Al device; (c) Current density (J) vs Voltage (V) plot of ITO/PEDOT:PSS/CF3-DPA/LiF-Al (red circles) and ITO/PEDOT:PSS/NPD/CF3-DPA/LiF-Al (blue stars) devices; (d) Luminance (L) vs Voltage (V) plot of ITO/PEDOT:PSS/CF3-DPA/LiF-Al (red circles) and ITO/PEDOT:PSS/NPD/CF3-DPA/LiF-Al (blue stars). Devices showed higher light output in PEDOT:PSS/NPD based devices as compared to PEDOT:PSS based devices.

low $V_{ON} = 4.0$ V compared to $V_{ON} = 10$ V for the trilayer devices without PEDOT:PSS (Figure 3(a)). We observed that all CuPc-based devices had low current density. The light output for the multilayer device was 6 Cd/m^2 at 10 mA/cm^2 . The reason for the decrease in current density in CuPc based OLEDs has been discussed in the literature.²⁷ It is believed that oxygen is released from ITO at a higher bias which diffuses into the CuPc layer. For higher CuPc thicknesses, the diffused oxygen results in a higher number of trap states for the holes. This reduces the hole mobility resulting in charge imbalance reducing light output as well as the current density.

As observed in all the devices studied so far, the presence of PEDOT:PSS reduces V_{ON} as well as the light output. In addition, NPD/CF3-DPA interface was found to play a crucial role in light output. Therefore, we decided to fabricate bilayer devices with only NPD as the HTL and CF3-DPA as the emitting layer.

See schematic of the device in Figure 4(a) and the corresponding energy band diagram in Figure 4(b). The thickness of NPD was kept ~ 45 nm. As the E_{HOMO} and E_{LUMO} of CF3-DPA were close to that of Alq3, devices with similar geometry but with Alq3 as the emitting layer were fabricated for comparison. The thickness of CF3-DPA was kept at ~ 50 nm which was optimized to give the maximum light output in this geometry. The thickness of Alq3 was ~ 60 nm. Figure 4(c) shows the J–V plots of two separate devices with CF3-DPA and Alq3 as emitters. The V_{ON} was observed to be higher in CF3-DPA than Alq3. Besides, the light output at 50 mA/cm^2 was almost one order of magnitude lower than that of Alq3 (Figure 4(d)). The electroluminescence spectra of Alq3 and CF3-DPA were recorded (Figure 4(e) and (f)) showing the emission peak at 514 nm and 439 nm, respectively. Deep blue emission was observed from the CF3-DPA devices (see an image of the device in the inset of Figure 4(f)). The 1931 CIE

Figure 3. (a) Energy band diagram of ITO/CuPc/NPD/CF3-DPA/LiF-Al device; (b) Energy band diagram of ITO/CuPc/PEDOT:PSS/NPD/CF3-DPA/LiF-Al device; (c) J–V plot of ITO/CuPc/NPD/CF3-DPA/LiF-Al (red circles) and ITO/CuPc/PEDOT:PSS/NPD/CF3-DPA/LiF-Al (blue stars). (d): L–V plot of ITO/CuPc/NPD/CF3-DPA/LiF-Al (red circles) and ITO/CuPc/PEDOT:PSS/NPD/CF3-DPA/LiF-Al (blue stars).

chromaticity (x, y) was (0.14, 0.04). Considering that the spectral response of the human eye in blue is much lower than that of green (see standard photopic curve), the light output seems reasonable though not very promising. This could be attributed to primarily two factors: (i) Low internal quantum efficiency of CF3-DPA in the solid state;²⁸ (ii) low probability of recombination in the emissive layer. In order to overcome the second problem, it was decided to introduce an ETL or a hole blocking layer (HBL) after CF3-DPA or introduce an electron blocking layer (EBL) before the CF3-DPA. Bathophenanthroline (BPhen) with a HOMO of -6.4 eV and LUMO of -3.0 eV²⁹ seemed to be a good choice for an HBL when placed after CF3-DPA. However, devices with ITO/NPD/CF3-DPA/BPhen/LiF-Al geometry did not help to improve the device performance. TPBi is also known to be an efficient HBL and ETL.³⁰ Devices with the geometry ITO/NPD/CF3-DPA/TPBi/LiF-Al were fabricated with the thickness of thermally evaporated TPBi being ~ 15 nm. The devices based on this geometry gave a high current density (J)

as well as high light output (L, Figure 5(b) and (c)) resulting in a luminance of 54 Cd/m^2 @ 50 mA/cm^2 . Since the LUMO of TPBi is just 0.1 eV higher than that of CF3-DPA (Figure 5(a)), it efficiently transports the electrons to the emissive layer. In addition, it has a HOMO of -6.2 eV which helps to block the holes thereby increasing the efficiency of carrier recombination in the emissive layer. 4,4'-Bis(*N*-carbazolyl)-1,1'-biphenyl (CBP) is known to have ambipolar charge transport properties³¹ and has been mostly used as a host material for phosphorescent dopants. It has a HOMO of -5.9 eV and a LUMO of -2.6 eV. Thus, we made devices where we used PEDOT:PSS and CBP as double HTLs with the following device geometry, ITO/PEDOT:PSS/CBP/CF3-DPA/TPBi/LiF-Al. In these devices, a 40 nm thick CBP layer was thermally evaporated over spin coated ITO/PEDOT:PSS. There was a weak blue emission from the devices with low currents. To possibly increase the efficiency of these devices, the area of contact of CBP with the active layer was increased. A blend of CF3-DPA and CBP (1:1

Figure 4. (a) 3D Schematic of ITO/NPD/CF3-DPA/LiF-Al OLED device; (b) Energy band diagram of the same device; (c) Current density (J) vs Voltage (V) plot of **CF3-DPA** (blue circles) and **Alq3** (green squares); (d) Luminance (L) vs Voltage (V) plot of **CF3-DPA** (blue circles) and **Alq3** (green squares); (e) Electroluminescence spectra of **Alq3** showing emission peak at 515 nm (inset: image of the device emitting green light); (f) Electroluminescence spectra of **CF3-DPA** showing emission peak at 439 nm (inset: image of the device emitting blue light).

weight ratio) was made in chloroform and it was spin coated on ITO/PEDOT:PSS at 5000 rpm for 40 s resulting in a thickness of 45 nm. Devices with the geometry ITO/PEDOT:PSS/(CBP + **CF3-DPA**)/TPBi/LiF-Al were fabricated which showed low V_{ON} (~5.5 V) and a very sharp increase of light (Figure 5(b) and (c)).

However, these devices were highly unstable and the light emission sharply decreased at a J of 5 mA/cm². We tried to increase their stability, by changing the ratio of CBP and **CF3-DPA** (0.2:1 to 1:1 by weight), the thickness of the layer (40–60 nm) and doing post-annealing of the **CF3-DPA**+CBP spin-coated layer at

Figure 5. (a) Energy band diagram of ITO//NPD/CF3-DPA/TPBi/LiF-Al device; (b) J–V plot of ITO/NPD/CF3-DPA/TPBi/LiF-Al (red circles) and ITO/NPD/CF3-DPA+CBP/TPBi/LiF-Al (blue stars). (c) L–V plot of ITO/NPD/CF3-DPA/TPBi/LiF-Al (red circles) and ITO/NPD/CF3-DPA+CBP/TPBi/LiF-Al (blue stars).

Figure 6. (a) L–J plot of the devices with all different geometries studied; (b) Efficiency (Cd/A) vs J plot for the four best devices. Red circles are for the bilayer device with NPD as HTL, blue triangles are for the tri-layer device with PEDOT:PSS/NPD as the HTL, dark green diamonds are for the trilayer device with NPD as HTL and TPBi as the ETL and the light green polygons are for the device where the active emissive layer is formed with a blend of CF3-DPA and CBP.

different temperatures (100–125 °C). The stability of the devices could not be improved appreciably. However, these results indicate that CBP behaves as an efficient carrier transporter/blocker.

To compare devices with all geometries, we plotted L vs J for all of them which is shown in Figure 6(a). It is clear from the plot that the best device geometry is ITO/NPD/CF3-DPA/TPBi/LiF-Al as it gives

the highest efficiency at 50 mA/cm² (Figure 6(b)). However, it is worth noting that at low J (~1 mA/cm²), the devices ITO/PEDOT:PSS/(CBP+CF3-DPA)/TPBi/LiF-Al seemed to be the most efficient thereby showing the utility of CBP as an ambipolar charge transporter.

4. Conclusions

We have synthesized trifluoromethyl substituted 1,8-diaryl anthracene (CF3-DPA) which absorbs at 394 nm and emits at 432 nm in solution. It shows a quantum efficiency of 67% when compared to 9,10-DPA thereby making it a promising candidate for blue-emitting OLEDs. Furthermore, in solid state thin films, it shows a small red shift in emission. The morphology of the films shows a low surface roughness indicating the possibility of the formation of smooth interfaces with HTLs in OLED devices. OLED devices with different geometries containing different HTLs were fabricated by vacuum thermal deposition. The EL spectra obtained on the working devices showed a peak at 439 nm. The turn on voltages and luminance values were compared for the devices with different geometries. Bilayer devices with PEDOT:PSS as the HTL gave no light output. Including a layer of NPD in the devices made them emit light indicating that the NPD/CF3-DPA interface was important for light output. In addition, the turn on voltage was also low ~7.5 V with PEDOT:PSS. The performance of CuPc based devices was poor. However, bilayer devices with the following geometry of ITO/NPD/CF3-DPA/LiF-Al gave reasonable light output though the turn on voltage was slightly higher at 13 V. Furthermore, introducing charge carrier blockers/transporters like TPBi and CBP helped to increase the efficiency of the devices. Devices having ITO/NPD/CF3-DPA/TPBi/LiF-Al geometry were observed to be the most efficient with a V_{ON} ~ 8.5 V. From our studies, we conclude that though a derivative of 1,8-DPA can be used as a blue emitter in OLED devices, the performance of the devices is limited by the mismatch of the hole and electron mobilities and the low internal quantum efficiency of CF3-DPA in solid state. Optimized device geometry using novel, suitable carrier blockers might increase the efficiency of 1,8-di-(4-trifluoromethylphenyl)-anthracene based blue emitters.

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