



## Solution processable truxene based blue emitters: Synthesis, characterization and electroluminescence studies



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### ABSTRACT

Truxene (10,15-dihydro-5H-diindeno[1,2-a;1',2'-c]fluorene) derived blue emitting materials were designed and developed utilizing Suzuki and Buchwald-Hartwig type cross-coupling reactions. The target molecules T<sub>1</sub>, T<sub>2</sub> and T<sub>3</sub> bearing N-carbazolyl, 1-pyrenyl and N-phenyl-N-(pyren-1-yl) substituents, respectively at 2,7,12- positions, were synthesized under mild conditions by palladium-catalyzed reactions in good yields. To improve the solution processability of the material, n-hexyl chains were used as substituents at 5,5', 10,10', 15,15' positions. From UV–vis measurements, the absorption maxima were found to be at 330 nm for T<sub>1</sub>, 356 nm for T<sub>2</sub> and 412 nm for T<sub>3</sub>. The materials were found to be blue emitting with their emission maxima at 385 nm, 425 nm and 490 nm for T<sub>1</sub>, T<sub>2</sub> and T<sub>3</sub>, respectively. The excited state lifetimes were investigated using time correlated single photon counting and were found to be 10 ns, 1.4 ns and 5.4 ns for T<sub>1</sub>, T<sub>2</sub> and T<sub>3</sub>, respectively. The highest photoluminescence quantum yield was observed in the case of T<sub>2</sub> corresponding to a value of 0.97. The compounds T<sub>2</sub> and T<sub>3</sub> were used as active materials for the fabrication of solution processed, single layer blue light emitting diodes, with low turn-on voltage (2–3.3 V) and Commission Internationale de l'éclairage (CIE) coordinates corresponding to (0.16, 0.23) and (0.17, 0.36) for T<sub>2</sub> and T<sub>3</sub>, respectively.

### 1. Introduction

Blue emitting materials have attracted considerable scientific and industrial interest since electroluminescence using small organic molecules was first demonstrated by Tang and VanSlyke [1]. This was soon extended to polymers by Friend and co-workers and led to the development of Organic Light Emitting Diode (OLED) technology [2]. Since OLED technology has the advantages of flexibility, low cost, and low power consumption, it has garnered significant interest due to its applications in flexible and large area full-colour displays and energy-efficient solid-state lighting [3]. To realize full-colour displays, highly efficient red, green and blue emitting materials of equal stability and colour purity are required [4,5]. However, the design and development of highly pure and stable blue emission is still a challenging task [6]. High-performance blue emitters are difficult to generate because of the presence of intrinsic wide band gap (> 3 eV), which makes it hard to inject charges into emitters sometimes leading to high drive voltages in devices [7,8]. Additionally, the aggregation in solid state leads to formation of excimers resulting in colour impurity [9]. Some of the prerequisites for blue emitting materials are appropriate energy levels, high quantum yields, good thermal, morphological stabilities and

appropriate molecular mass for solution processability. The  $\pi$ -conjugated organic molecules with their easy chemical tailoring can incorporate the aforesaid desirable properties. Previously, a lot of  $\pi$ -conjugated fluorophores have been reported with anthracene [10,11], pyrene [12,13], carbazole [14,15] and triarylamines [16] as the building blocks. Based on their architecture,  $\pi$ -conjugated blue emitting materials can be classified as either small molecules or polymers. The polymeric materials can be easily solution processed but often with large polydispersity index and the presence of oligomers, their purity becomes questionable leading to decreased efficiency in devices. On the contrary, purification of small molecules is relatively easy with techniques like column chromatography or recrystallization from solution, but they cannot be solution processed and must be vacuum deposited on the substrates. This poses as a major disadvantage for the small molecules since solution processing methods such as spin-coating and inkjet printing are much less wasteful and cost effective techniques [17]. Truxene based molecules belonging to the class of  $\pi$ -conjugated star shaped aromatic materials possess the virtues of both small molecules and polymers. With their precise chemical structures, high purity and solution processability, they have shown great potential as fluorescent blue emitting materials. Generally, the presence of a core and

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side moieties as building blocks is integral to their design [7,18]. Thus, development of new and efficient fluorescent blue emitting materials remains a challenge and an active area of research.

Truxene has been used for various applications including OLEDs [19–22], photovoltaic solar cells (PSC) [23–25], organic field emitting transistors (OFETs) [26], organic lasers [27], chemo sensors [28], in non-linear optics [29–31], two-photon absorption studies [32–35] and as building blocks for fullerenes [36]. This  $C_3$  symmetric molecule is planar in nature and possesses large  $\pi$ -conjugated system with multiple reactive sites, making it an attractive substrate for designing molecules catering to various applications. With truxene, solution processability can be achieved by attaching *n*-alkyl or branched alkyl chains at positions 5,5', 10,10', and 15,15'. Though the molecule is planar in nature, bulky side groups can be easily introduced at 2-, 7- and 12- positions to introduce solubility into the molecular system. This can also help to prevent the formation of aggregates and retain the colour purity. Also, the introduction of non-coplanarity subdues the crystallinity [37]. A distinct feature of truxene based compounds is their high photoluminescence quantum yield (PLQY) due to their rigid structures which translates into high electroluminescence (EL) efficiency in OLEDs [38]. Due to all these favourable properties, truxene has been used as a core, side moiety and even truxene based polymers have been developed as blue fluorescent emitting materials [39]. On the contrary, only a few of these truxene based blue emitting materials with various moieties such as triphenylamine, spiro-based, oligo(*p*-phenylene), have been used in OLED applications. To the best of our knowledge, the turn-on voltage ( $V_{ON}$ ) has been found in the range of 3–9 V, which results in high drive voltage to reach optimum luminance (800–1000's  $cd\ m^{-2}$ ) [19,21,38]. When compared with their counterparts created from vacuum deposition, their EL efficiencies are still low [38]. Thus, it is imperative to explore the prospect of design and development of novel truxene based materials.

Here, we report on the synthesis, photophysical and device studies on three truxene based blue emitters. The target molecules  $T_1$ ,  $T_2$ , and  $T_3$  (Fig. 1) were synthesized under mild conditions utilizing Suzuki and Buchwald-Hartwig cross-coupling reactions. The photophysical properties of the synthesized materials as well as their OLED device properties are presented and discussed.

## 2. Experimental section

### 2.1. Materials and methods

All the chemicals used for synthesis were obtained from Sigma-Aldrich, Merck and Spectrochem and were used without further

purification unless mentioned.  $^1H$  NMR spectra were recorded on a Bruker 400 MHz spectrometer and were referenced to internal standard, tetramethylsilane (TMS).  $^{13}C$  NMR spectra were recorded at 75 MHz on a Bruker 300 MHz NMR spectrometer. Thin layer chromatography (TLC) was performed on Merck silica gel 60 sheets. Column chromatography was performed on silica gel (Merck 60–120 mesh). The steady-state UV–vis spectra were recorded on a T90 + UV–visible spectrophotometer in tetrahydrofuran (THF). Photoluminescence (PL) spectra were obtained from Agilent Cary Eclipse fluorescence spectrophotometer in THF and as spin coated film on quartz substrate. The PLQY were recorded by relative method in dichloromethane (DCM) at room temperature using anthracene as a reference ( $\Phi_F = 0.27$ , in ethanol). For solid-state PLQY, 10 mg  $mL^{-1}$  solution of  $T_2$  and  $T_3$  in 1,2-dichlorobenzene were spin coated on a pre-cleaned quartz substrate. After exciting the samples with 355 nm laser, the spectrum was collected by an Integrating Sphere (Allied Scientific Pro) and scanned using Research India spectrometer. The time-correlated single photon counting (TCSPC) was done in THF solution on Horiba Jobin Yvon IBH, UK with the time resolution of 7 ps. The Instrument Response Function (IRF) of the three incident sources for 335 nm, 375 nm, and 340 nm was 590 ps, 750 ps, and 280 ps. The fitting of the PL decay was done using the monoexponential decay equation on IBH das v6.2 software. Cyclic voltammograms were recorded on a Biologic-SP 150 electrochemical quartz crystal microbalance (EQCM) at a constant scan rate of 50  $mV\ s^{-1}$  at room temperature using 0.1 M tetrabutylammonium perchlorate in DCM as the supporting electrolyte. The experiments were performed in a three electrode cell, in which a calomel reference electrode in saturated KCl solution, with platinum wire counter electrode and glassy carbon working electrode, were used for measurements. The mass spectra of the synthesized compounds were recorded by utilizing matrix-assisted laser desorption ionization time of flight mass spectrometry (MALDI-TOF MS) on AutoFlex from Bruker Daltronics using dithranol as a matrix.

### 2.2. Synthetic procedures

The compounds 1-bromopyrene (1) [40], 1-(4,4',5,5'-tetramethyl-1,3,2-dioxaborolan-2-yl)pyrene (2) [41], *N*-phenylpyren-1-amine (3) [42], truxene (4) [43], 5,5',10,10',15,15'-hexahexyltruxene (5) [44,45], and 5,5',10,10',15,15'-hexahexyl-2,7,12-tribromotruxene (6) [35,45] were synthesized according to literature.

#### 2.2.1. 9,9',9''-(5,5,10,10,15,15-hexahexyl-10,15-dihydro-5H-diindeno [1,2-*a*:1',2'-*c*]fluorene-2,7,12-triyl)tris(9H-carbazole) ( $T_1$ )

To a solution of 2,7,12-tribromo-5,5',10,10',15,15'-

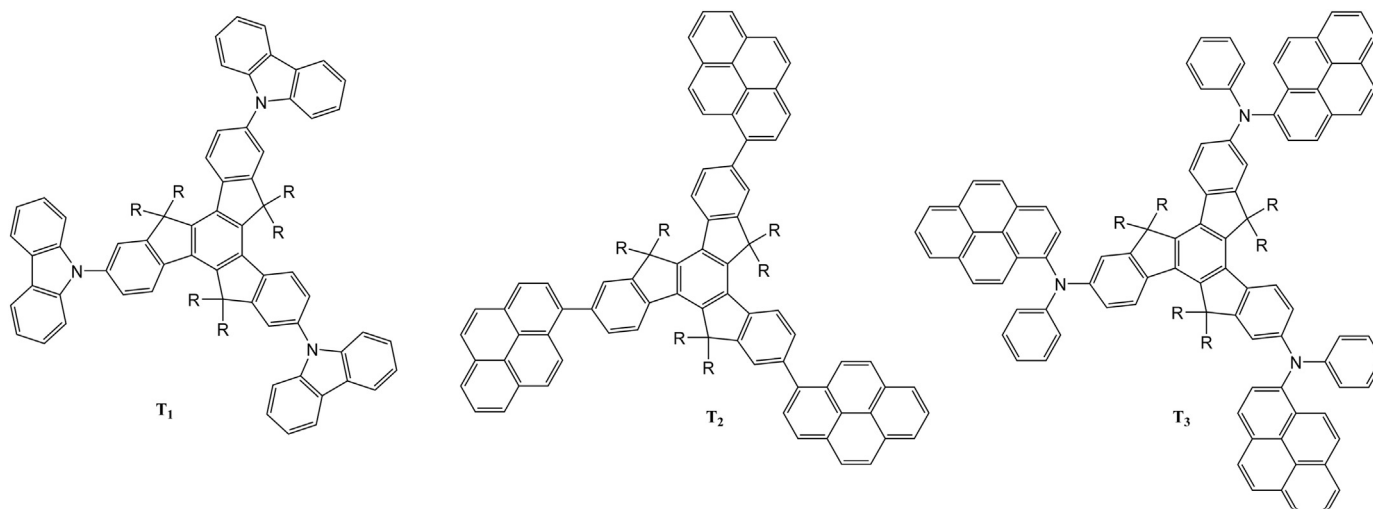


Fig. 1. Structures of the target molecules.

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