



Multi-functional highly efficient bipolar 9,9-dimethyl-9,10-dihydroacridine/imidazole-based materials for solution-processed organic light-emitting diode applications



Saripally Sudhaker Reddy¹, Woosum Cho¹, Vijaya Gopalan Sree, Sung-Ho Jin^{*}

Department of Chemistry Education, Graduate Department of Chemical Materials, and Institute for Plastic Information and Energy Materials, Pusan National University, Busan, 609-735, Republic of Korea

ARTICLE INFO

Article history:

Received 14 June 2016
Received in revised form
22 July 2016
Accepted 23 July 2016
Available online 25 July 2016

Keywords:

9,9-Dimethyl-9,10-dihydroacridine/
imidazole
Deep-blue fluorescent emitters
Host
Nondoped OLED
Solution-process

ABSTRACT

A new class of bipolar compounds were designed and synthesized based on 9,9-dimethyl-9,10-dihydroacridine and phenylphenanthroimidazole/triphenylimidazole derivatives with different linking positions. Their thermal, photophysical, electrochemical properties and device characteristics were investigated. All four compounds were fabricated as emitters in solution-processed nondoped OLEDs. In particular, the *N*-phenylacridine 1,4-phenylene linked *N*-phenylphenanthrimidazole analogue displayed high efficiency with a maximum current efficiency (CE_{max}) of 4.6 $cd A^{-1}$ and a maximum external quantum efficiency (EQE_{max}) of 5.1% at CIE coordinates of (0.15, 0.09), which is very close to the NTSC blue standard. In addition, when the *N*-phenylacridine 1,3-phenylene linked triphenylimidazole analogue was used as a host for green phosphor $Ir(mppy)_3$, shows an impressive device performance with a CE_{max} of 62.7 $cd A^{-1}$ and a EQE_{max} of 18.9%. To the best of our knowledge, both results are the best efficiencies as emitter and host for solution-processed nondoped deep-blue OLEDs and green PhOLEDs, respectively.

© 2016 Elsevier Ltd. All rights reserved.

1. Introduction

Organic light-emitting diodes (OLEDs) have drawn much attention owing to their potential applications in flat panel, full-color displays and solid state lightings [1]. In general three primary colors of red, green and blue with color-purity and stability are significant for full-color displays and in particular, emitters with CIE $y < 0.15$ are of high importance. These emitters do not only offer potential for stable deep-blue emission but also can serve as a host for green and red phosphorescent dopants due to their wide energy-gaps [2]. Thus, fluorescent compounds are advantageous over phosphorescent complexes due to their functionality and versatile applications, such dual functional fluorescent compounds are viable to mitigate the manufacturing cost as well as beneficial for white light [3,4]. Several studies demonstrated that the efficient core plays a key role for the development of new fluorescent compounds with desired photophysical properties which

eventually determines the functionality and applicability [5–8]. In this scenario, several cores (pyrene, anthracene, fluorene, chrysenes and diphenylamine) have been chosen and reported as fluorescent emitters and hosts for OLEDs [9–12]. Among them, electron-rich diphenylamine core has been one of the interesting unit with fascinating features triggered the attention of many researchers and developed several fluorescent materials by breaking/making key unit linking C–C bonds. Even though the carbazole (Cz) derivatives have attracted features, the studies have been focused on optoelectronic and biological applications [13,14]. However, choosing hole/electron transporting units are vital for building up of bipolar fluorescent compounds. Currently, 9,9-dimethyl-9,10-dihydroacridine derivatives are drawing great attention for their good hole transporting ability, feasibility of their derivatives and promising results for OLEDs [15,16]. On the other side, electron-deficient imidazole derivatives have been intriguing because of their excellent electron transporting ability [9–12]. In particular, phenylphenanthroimidazole (PPI)/triphenylimidazole (TPI) derivatives are receiving attention due to their blue-emission as well as good electron-injection/transporting ability for OLEDs [17,18].

However, to be either a good fluorescence emitter or a host requires appropriate design strategy in which suitable energy

* Corresponding author.

E-mail address: shjin@pusan.ac.kr (S.-H. Jin).

¹ S. S. Reddy and W. Cho contributed equally to this work.

levels, better charge carrier mobility, high thermal stability and high solubility for solution-processable OLEDs, especially high triplet energy (E_T) required for host material to prevent back energy transfer from dopant to host [12,19]. In this regard, despite many fluorescent compounds have been designed and synthesized for nondoped deep-blue fluorescent OLEDs, the materials with more than 5% external quantum efficiency (EQE) are very rare [20,21]. Recently, a bipolar host material was reported by utilizing Cz and benzimidazole for solution-processed green EL devices with an EQE of 13.2% [22]. Although numerous attempts have been made on various kinds of materials with different linking topology based on carbazole, benzimidazole and phenylphenanthroimidazole derivatives as host materials for efficient PhOLEDs, the EQE is limited to 13–18% [22–29]. Moreover, most of the devices were fabricated by a vacuum-process, which can enhance the overall production cost, while solution-processed OLEDs are economically feasible to large-area and low-cost devices that are very significant for commercialization. Therefore, it is very challenging task to design new fluorescent building blocks and understanding the structure-property relationship for the development of efficient fluorescent emitters as well as high performance hosts for solution-processed OLEDs.

Herein, we designed and synthesized four 9,9-dimethyl-10-phenyl-9,10-dihydroacridine and phenylphenanthroimidazole/triphenylimidazole derivatives by different linkage to investigate the impact of position on thermal, optical, electrochemical properties, and further applied as fluorescent emitters and also host for highly efficient solution-processed OLEDs. All four compounds show deep-blue emission, suitable energy levels, high mobility, good thermal stability and high E_T . Four compounds have shown impressive device performance when fabricated as emitters in solution-processed nondoped OLEDs. In particular, compound **I** displayed high performance of a CE_{max} of 4.6 cd A^{-1} and a EQE_{max} of 5.1% with the CIE coordinates of (0.15, 0.09). Furthermore, using **IV** as a host for green phosphor showed an excellent device performance with a CE_{max} of 62.7 cd A^{-1} and a EQE_{max} of 18.9%. As of now, these are the best efficiencies for solution-processed nondoped OLEDs as emitters as well as host for green PhOLEDs.

2. Experimental section

2.1. Materials and measurements

Chemicals and reagents were used without further purification; THF was purified by distillation over sodium/benzophenone under a N_2 atmosphere and other solvents were used without purification. Merck-precoated aluminium thin layer chromatography (TLC) plates were used to monitor the reactions. The melting points were recorded on a Buchi Melting Point M-560 and the IR spectra were recorded on the Agilent technologies Cary 630 FTIR. 1H and ^{13}C NMR spectra were recorded on a Varian Mercury Plus 300 MHz spectrometer in $CDCl_3$ using tetramethylsilane (TMS) as an internal standard and the chemical shifts were reported in ppm relative to the singlet's of $CDCl_3$ at 7.26 and 77 ppm for the 1H and ^{13}C NMR, respectively. These abbreviations were used to explain the multiplicities in 1H NMR spectra: s = singlet, d = doublet, t = triplet, q = quartet, m = multiplet. HRMS (FAB) mass spectrometry was received from the Korea Basic Science and Institute Daejeon Center. JASCO V-570 and Hitachi F-4500 fluorescence spectrophotometers were used to record the UV-vis absorption and fluorescence spectra, respectively. Quantaurus-Tau Hamamatsu compact fluorescence lifetime spectrometer C11367 was used to measure decay time. Thermal analyses (thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC)) were performed on a Mettler Toledo TGA/SDTA 851e, DSC 822e analyzer under a N_2

atmosphere at a heating rate of $10 \text{ }^\circ\text{C min}^{-1}$. Cyclic voltammetry (CV) studies were carried out with a CHI 600C potentiostat (CH Instruments), which was equipped with a platinum disc as the working electrode, platinum wire as the counter electrode, and Ag/AgCl as the reference electrode, at a scan rate of 100 mV s^{-1} in a 0.1 M solution of tetrabutylammonium perchlorate (TBAP) as the supporting electrolyte in methylene chloride (CH_2Cl_2) solution. The potentials were referenced to the ferrocene/ferrocenium redox couple (Fc/Fc^+), and it was assumed that the redox potential of Fc/Fc^+ had an absolute energy level of -4.8 eV to vacuum. The potential of Fc/Fc^+ was measured under the same conditions and located at 0.09 V relative to the Ag/Ag $^+$ electrode. All electrochemical experiments were performed under an air atmosphere at room temperature. AFM images were measured by utilizing XE-100 (park system corp., Korea) at tapping mode. Density functional theory (DFT) was used to investigate the molecular geometries and electron density distribution. B3LYP correlation functional and the basis set of 6-31G* in the Gaussian 03 package were used for theoretical calculations.

2.2. Device fabrication and measurement

The hole-injection material poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS), hole-transporting/electron-blocking material tris(4-carbazoyl-9-ylphenyl)amine (TCTA) and electron-transporting material 2,2',2''-(1,3,5-benzinetriyl)-tris(1-phenyl-1-H-benzimidazole) (TPBI) were commercially available. PEDOT:PSS was spin-coated on the UV-Ozone-treated ITO-coated glass substrate, followed by annealing in air for 15 min at $150 \text{ }^\circ\text{C}$. For the nondoped devices the emitting layer (EML) was then spin-coated on top of PEDOT:PSS and annealed at $120 \text{ }^\circ\text{C}$ for 20 min in inert N_2 atmosphere. Whereas for PhOLEDs, a 40 nm layer of TCTA was spin-coated on top of PEDOT:PSS and the EML was then spin-coated onto TCTA (IV as a host doped with 8 wt% of the tris-(2-(4-(4-tolyl)phenyl)pyridine) iridium(III) ($Ir(mppy)_3$). Chlorobenzene was used as the solvent for the EML. TPBI, a cathode composed of LiF and Al were sequentially deposited over the EML layer for both nondoped and PhOLEDs by vacuum evaporation process. The current density-voltage-luminescence (J-V-L) characteristics of the devices were measured with a Keithley 236 source measurement unit and CS1000 spectrophotometer. All measurements were carried out under ambient condition at room temperature. The thickness of each layer was measured by an Alpha-step IQ surface profiler (KLA Tencor, San Jose, CA).

2.3. Synthesis

2.3.1. Synthesis of 9,9-dimethyl-10-phenyl-9,10-dihydroacridine (**2**)

To a solution of 9,9-dimethyl-9,10-dihydroacridine (**1**) (1.00 g, 4.78 mmol) in dry toluene (20 mL) was added sodium *tert*-butoxide (NaOt-Bu) (0.92 g, 9.56 mmol) and bromobenzene (0.83 g, 5.26 mmol). The reaction mixture was purged with argon for 30 min, and then palladium(II) acetate ($Pd(OAc)_2$) (22 mg, 0.10 mmol) and tri-*tert*-butylphosphine ($P(t-Bu)_3$) (48 mg, 0.24 mmol) was added. The resulting reaction mixture was stirred at $110 \text{ }^\circ\text{C}$ for 12 h. Upon reaction completion, the reaction mixture was diluted with ethyl acetate (EtOAc) and water, extracted organic layer, washed with water and brine solution. Finally, the organic layer was dried under reduced pressure. The crude compound was purified by column chromatography on silica gel eluted with EtOAc/hexanes (0.1:9.9 to 0.2:9.8) to afford **2** as a white solid. Yield: 1.30 g (95%). $R_f = 0.6$ (silica, EtOAc/hexanes 1:9), mp: $122\text{--}125 \text{ }^\circ\text{C}$. FTIR (neat, cm^{-1}) 2962, 1583, 1434, 1316, 1266. 1H NMR (300 MHz, $CDCl_3$, δ): 7.66–7.61 (m, 2H), 7.53 (d, $J = 6.9 \text{ Hz}$, 1H), 7.47 (d, $J = 7.5 \text{ Hz}$, 2H),

Download English Version:

<https://daneshyari.com/en/article/175265>

Download Persian Version:

<https://daneshyari.com/article/175265>

[Daneshyari.com](https://daneshyari.com)