Organic Electronics 15 (2014) 2148–2157

Contents lists available at ScienceDirect

Organic Electronics

journal homepage: www.elsevier.com/locate/orgel

Tetrasubstituted-pyrene derivatives for electroluminescent application

Rossatorn Muangpaisal [a,b,c], Ming-Chi Ho [b,d], Tai-Hsiang Huang [b], Chih-Hsin Chen [b], Jiun-Yi Shen [b], Jen-Shyang Ni [b], Jiann T. Lin [b,*], Tung-Huei Ke [e], Li-Yin Chen [e], Chung-Chih Wu [e,*], Chiitang Tsai [d,*]

[a] Taiwan International Graduate Program, Taiwan
[b] Institute Chemistry, Academia Sinica, Taipei 11529, Taiwan
[c] Department of Chemistry, National Tsing Hua University, 101 Section 2 Kuang-Fu Road, Hsinchu 30031,Taiwan
[d] Department of Chemistry, Chinese Culture University, Taipei 111, Taiwan
[e] Department of Electrical Engineering, Graduate Institute of Electro-optical Engineering and Graduate Institute of Electronics Engineering, National Taiwan University, Taipei 106, Taiwan

ARTICLE INFO

Article history:
Received 21 February 2014
Received in revised form 21 April 2014
Accepted 2 June 2014
Available online 21 June 2014

Keywords:
Pyrene
Star-shaped materials
Organic light-emitting diode
Ambipolar transport

ABSTRACT

Tetrasubstituted-pyrenes containing peripheral diarylamines (**1**–**4**) or fluorenes (**5**–**6**) have been synthesized. These compounds are highly fluorescent and possess high morphological stability and thermal stability. Compounds containing peripheral arylamines (**1**–**3**) can be used as the hole-transport and green-emitting materials for two-layered electroluminescent devices. Compounds with peripheral fluorenes (**5**–**6**) are efficient blue emitters and exhibit ambipolar carrier-transport characteristics with high electron mobilities (10^{-3}–10^{-2} $cm^2/V\,s$) and high hole mobilities (>10^{-3} $cm^2/V\,s$). Non-doped blue-emitting devices with promising electroluminescent performance (i.e., high efficiency and narrow/saturated emission) can be achieved using fluorene–substituted pyrenes as either the hole-transport/emitting layer or the electron-transport/emitting layer in the two-layered devices.

1. Introduction

Two decades after Kodak's [1] and Cambridge's [2] seminal work on organic electroluminescence (EL), organic light-emitting diodes (OLEDs) have become widely used in practical appliances such as car stereos, mobile phones, and digital cameras. In OLEDs, balanced electron and hole mobilities are extremely important in order to achieve good EL performance. Through proper choice of carrier-transport and emitting materials, and engineering of device structures, better confinement of charge recombination can generally be achieved in a multilayer device. Moreover, longevity of EL devices is also highly demanded for commercial applications. Therefore, the durability, e.g., thermal and morphological stability of deposited organic films, cannot be overlooked. Unlike polymeric materials which can easily form films via the spin-coating technique, films of small molecules normally are vacuum-deposited. Consequently, the compounds must be able to tolerate the deposition temperature, which frequently exceeds 250 °C. Amorphous compounds possessing high glass transition temperatures (T_g) are highly desired as crystallization can be avoided and film morphology can be retained at elevated temperatures during device operation [3–6].

Molecules possessing a non-planar structure and different conformers have a great tendency to form amorphous glasses [5]. Among these, star-shaped π-conjugated molecules allowing incorporation of several bulky and

* Corresponding authors. Tel.: +886 2 27898522.
E-mail addresses: jtlin@chem.sinica.edu.tw (J.T. Lin), chungwu@cc.ee.ntu.edu.tw (C.-C. Wu), tsaictom@faculty.pccu.edu.tw (C. Tsai).

http://dx.doi.org/10.1016/j.orgel.2014.06.003
1566-1199/

rigid substituents are of particular interest. Previously we developed several series of star-shaped compounds which readily formed glasses once isolated. These high T_g materials were successfully used to fabricate efficient electroluminescent (EL) devices [7,8]. In our continuation of developing high T_g materials for EL applications, we selected 1,3,6,8-tetrabromo-pyrene as the building block for new star-shaped compounds based on the following reasons: (1) according to our previous studies and other literature reports, incorporation of the pyrenyl unit was beneficial in raising the thermal stability of the compounds [9,10], (2) tetraphenylpyrene was reported to be strongly emissive [11] and (3) pyrenyl unit can be readily functionalized and its emission color can be tuned [12,13]. Compounds containing pyrenyl units have found useful applications in liquid crystals [14,15] and field-effect transistors [16,17]. Besides, the pyrenyl unit and its excimer was found useful in sensing [18–20]. However, formation of excimers normally quenches the fluorescence and deteriorate the performance of EL devices [21]. Star-shaped pyrenyl derivatives obtained by replacing four bromine atoms of 1,3,6,8-tetrabromopyrene with appropriate peripheral substituents are expected to encapsulate the pyrenyl moiety and prevent the formation of excimers. Moreover, incorporation of carrier-transporting peripheral substituents is possible. There have been reports on using 1,3,6,8-substituted pyrenyl derivatives as the light emitting materials in OLEDs [19]. Liu et al. developed pyrene derivatives containing four peripheral oligofluorenes of varied chain lengths. Single-layered OLED based on these materials exhibited a luminance efficiency of 1.28 cd A^{-1} with CIE coordinates at (0.19, 0.32) and 1.75 cd A^{-1} with CIE coordinates at (0.20, 0.32) for bifluorene and tris(fluorene) derivatives, respectively [22]. Sonar et al. developed pyrene-based compounds capable of emitting various colors by incorporation of different moieties. Among these, only the deep blue-emitting 1,3,6,8-tetrakis-(4-butoxyphenyl)pyrene was subjected to OLED fabrication, and high luminance efficiency of 2.56 cd A^{-1} with CIE coordinates at (0.15, 0.18) was achieved [23]. Recently, Thomas et al. reported blue to yellow light-emitting materials based on tetrasubstituted-pyrene core with acetylene unit as the linker to the peripheries, and these compounds exhibited red-shifted emission compared to the monosubstituted derivatives. However, these materials need to be doped in a host for OLEDs, possibly due to lack of amorphous morphology [24].

Herein, we report light-emitting 1,3,6,8-substituted pyrene derivatives possessing high quantum efficiency and carrier mobility. EL devices fabricated from these star-shaped materials will be also presented.

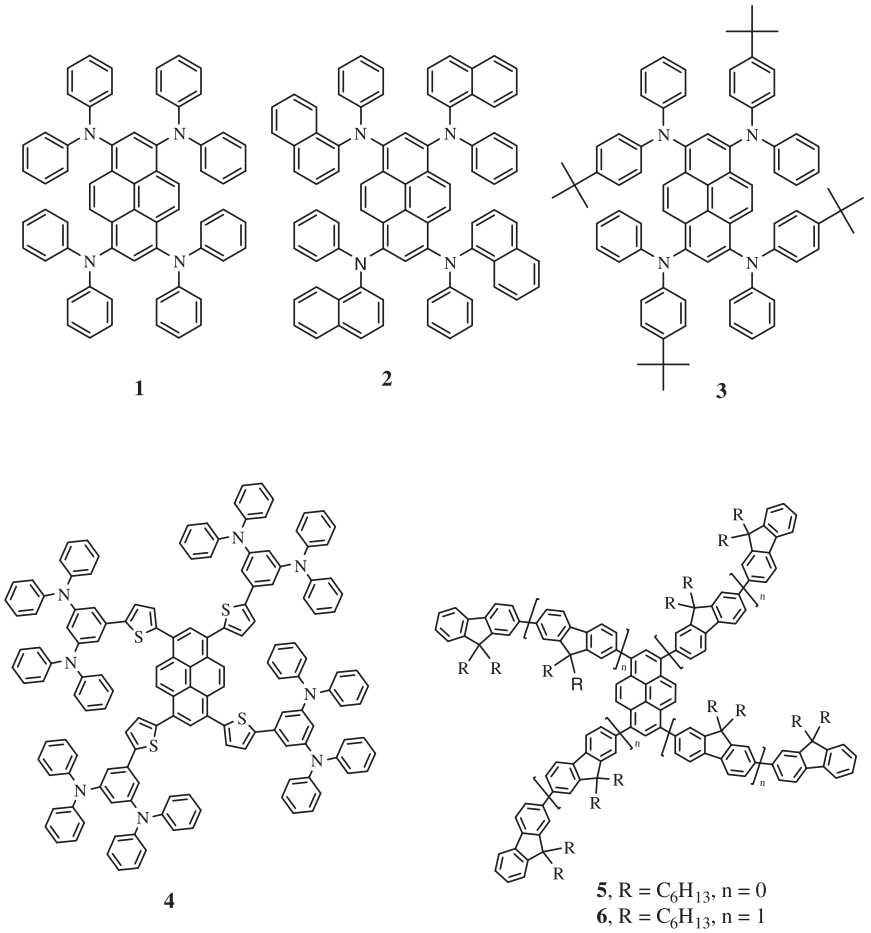

Chart 1. Structure of compounds **1–6**.