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LED based on alternating benzene-furan oligomers

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ABSTRACT

A series of conjugated-chain compounds which contain two furan units and more than three other aromatic ring units were synthesized by two different methods, their UV–visible absorption coefficient (ϵ), maximum absorption wavelengths (λ_a), fluorescence emission wavelengths (λ_e), stokes shift and quantum yields (Φ) were determined, the relationships between the molecular structures and the effects of spectral behaviors were discussed. Meanwhile, the excitation spectrum and emission spectrum of **2b** and **2d** in solid and in ethyl acetate were respectively recorded, the thermal stability of **2b** and **2d** was evaluated, and the fluorescence emission behaviors of two light emitting diodes, fabricated with **2b** and **2d** as phosphors, were investigated. The results showed that most of target compounds can perform high fluorescence emission ability, compounds **2b** and **2d** can perform higher thermal stability under 350 °C and they are suitable for making light emitting diode as phosphors, the light emitting diodes fabricated with **2b** and **2d** can show higher fluorescence emission ability. Therefore, those compounds are worthy of further being developed as fluorescent emission materials.

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1. Introduction

Chemists recently paid a lot of attention to synthesize and investigate the optical behaviors of organic conjugated-chain materials, because these compounds are very useful in many fields, such as in twophoton [1–4] and three-photon absorption [5], organic second-order nonlinear optics [6,7] and organic light-emitting device [8–11]. Among these conjugated-chain materials, the compounds containing thiophene [12-17], thiazole, imidazole [18] or furan unit [19-26] could perform very interesting optical property. To the best of our knowledge, there are some reports about the synthetic methods [27-36] and optical property [19-23,36] of alternating benzene- furan oligomers, however, the reports about the relationships between the structure and optical property of these oligomers are rather sparse, and especially, the reports about the applications of these oligomers for OLED as light emitting diode was not found. Therefore, a series of 1,4-bis-(3,5-diphenylfuran-2-yl)benzene (Fig. 1.1), 1,4-bis-(4,5-diphenylfuran-2-yl)-benzene (Fig. 1.2) and 1,3-bis(3,5-diphenylfuran-2-yl)benzene (Fig. 1.3) analogues were designed and synthesized in order to investigate their

* Corresponding authors. *E-mail address:* yjhxhc@mail.sysu.edu.cn (H. Song). optical behaviors and their applications for OLED. The synthetic pathway and structures of target compounds were outlined in the following (Fig. 2. and Fig. 3).

2. Experimental

2.1. Instruments and reagents

All melting points were determined on a $\times 4$ melting point microscope. ¹H and ¹³C NMR spectra were run on a Bruker AVANCE-300 NMR spectrometer or AVANCE-300 NMR spectrometer. Element analyses were taken with an Elementar Analysensysteme GmbH Vario EL. Ultra- violet spectra were recorded on a Shimadzu UV-1601 spectraphotometer. Fluorescence spectra were taken with a Shimadzu RF-5301PC spectrofluorophotometer. Mass spectra and high resolution mass spectrometer were taken respectively with a Shimadzu LCMS-2010A and Thermo MAT95XP. Thermogravimetric analyses were taken with a Netzsch TG209 F1 Thermogravimetric analyzer. Color coordinates of emission spectra of LEDs were determined by Minolta CS-100A Chroma Meter with Minolta DP-101 data processor. All the reagents were commercially available and they were used without further purification. All the solvents were dried using standard methods before use. All reactions were performed in flame-dried glassware under argon.



Fig. 1. Typical structures of 1,4-bis-(3,5-diphenylfuran-2-yl)benzene (1), 1,4-bis-(4,5- diphenylfuran-2-yl)benzene (2) and 1,3-bis(3,5-diphenylfuran-2-yl)benzene (3).

2.2. Synthesis of samples

2.2.1. Synthesis of intermediate compound 1a-1d

2.2.1.1. Intermediate compounds **1a-1c**. Intermediate compounds 2-(biphenyl-4-yl)-2-(phenylethynyl)-1,3-dithiolane(**1a**), methyl 4-(2-(hex-1-ynyl)-1,3-dithiolan-2-yl)benzoate(**1b**) and 2-(naphthalen-2yl)-2- (phenylethynyl)-1,3-dithiolane(**1c**) were prepared according to reference [7a].

2.2.2. 2,3'-(1,4-Phenylene)bis-(2-(phenylethynyl)-1,3-dithiolane) (1d)

According to reference [7a], intermediate **1d** was obtained with a yield of 71.0% from ethane-1,2-dithiol and 1,1'-(1,4-phenylene)bis-(3-phenylprop-2-yn-1-one) which was prepared by oxidation reaction of 1,1'-(1,4-phenylene)bis-(3-phenylprop-2-yn- 1-ol) with activated MnO₂. ¹HNMR (300 MHz, CDCl₃, δ): 3.77–3.79 (m, 8H), 7.23 (s, 4H), 7.50–7.59 (m, 6H), 8.17 (d, J = 10.5 Hz, 4H). ESI-MS m/z: 487.1 (M + H)⁺.

2.2.3. Synthesis of 2a-2e

General annulation procedure **A** for the synthesis of compound **2a–2e** is in the following. Into a 50 mL three-neck flask 4.2 mmol of compound **1a** (or **1b**, **1c**) was added, the system was washed with

argon for several times to remove O₂ and water, and reaction was carried out under argon atmosphere. Then dried THF was injected into the mixture. Compound 1a (or 1b, 1c) was dissolved by stirring the mixture, when the mixture was cooled to -78 °C, 4.41 mmol (1.76 mL) BuLi was added. The mixture was stirred for 1.5 h at -78 °C, 2.0 mmol of aromatic dialdehyde in 10-30 mL of dried THF was added. After the reaction mixture was stirred at this temperature for another 40 min, the mixture was allowed to reach to room temperature and the stirring was continued for another 30 min. 0.3 mL of trifluoroacetic acid was added, after being stirred for 10 h 3 mL of saturated ammonium chloride was added into the mixture to quench the reaction and the organic layer was evaporated under reduced pressure to get black viscous mixture. The residue was isolated by chromatography on a silica-gel column and the solid product was recrystallized from chloroform to obtain pure compounds 2a-2e.

2.2.4. 1,3-Bis(5-([1,1'-biphenyl]-4-yl)-3-phenylfuran-2-yl)benzene (**2a**)

0.52 g Of **2a** was obtained with a yield of 38.6% from 4.2 mmol (1.50 g) of compound **1a** and 2.0 mmol (0.27 g) of 1,3benzenedicarboxaldehyde according to procedure **A**. m.p. 230–232 °C. ¹H NMR (300 MHz, CDCl₃, δ): 6.82 (s, 2H), 7.20–7.25 (m, 1H), 7.36– 7.51 (m, 18H), 7.63–7.66 (m, 8H), 7.75 (d, *J* = 8.4 Hz, 4H), 8.01 (s,



1a $Ar^{1} = C_{6}H_{5}$, $Ar^{2} = C_{6}H_{5}C_{6}H_{5}$; **1b** $Ar^{1} = C_{4}H_{9}$, $Ar^{2} = 4-C_{6}H_{4}CO_{2}CH_{3}$; **1c** $Ar^{1} = C_{6}H_{5}$, $Ar^{2} = 2-C_{10}H_{7}$.







2c Dimethyl 4,4'-([1,1'-biphenyl]-4,4'diylbis(4-butylfuran-5,2-diyl))dibenzoate





2d 4,4'-Bis(5-(naphthalen-2-yl)-3phenylfuran-2-yl)-1,1'-biphenyl



2e 4,4'-Bis(5-([1,1'-biphenyl]-4-yl)-3-phenylfuran-2-yl)-1,1'-biphenyl

Fig. 2. Synthesis and structures of compounds 2a-2e.

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