



Molecular Crystals and Liquid Crystals

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Synthesis and Electroluminescent Properties of Fluorene- And Anthracene-Derivatives Containing Novel Tetraphenylbenzene Moiety

Kwang-Yol Kay^a, Jung Kim^a, Hyun Cho^b & Jong-Wook Park^c

^a Department of Molecular Science and Technology, Ajou University, Suwon, Korea

^b Materials and Devices Research Center, KIST, Seoul, Korea

^c Department of Chemistry/Center for Nanotechnology Research, The Catholic University, Pucheon, Korea

Version of record first published: 18 Oct 2010

To cite this article: Kwang-Yol Kay, Jung Kim, Hyun Cho & Jong-Wook Park (2004): Synthesis and Electroluminescent Properties of Fluorene- And Anthracene-Derivatives Containing Novel Tetraphenylbenzene Moiety, *Molecular Crystals and Liquid Crystals*, 424:1, 167-172

To link to this article: <http://dx.doi.org/10.1080/15421400490506126>

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SYNTHESIS AND ELECTROLUMINESCENT PROPERTIES OF FLUORENE- AND ANTHRACENE-DERIVATIVES CONTAINING NOVEL TETRAPHENYLBENZENE MOIETY

Kwang-Yol Kay and Jung Hoon Kim
Department of Molecular Science and Technology,
Ajou University, Suwon 442-749, Korea

Hyun Nam Cho
Materials and Devices Research Center,
KIST, Seoul 130-650, Korea

Jong-Wook Park
Department of Chemistry/Center for Nanotechnology Research,
The Catholic University, Puccheon 420-734, Korea

2,7-Bis[(2,3,4,5-tetraphenyl)phenyl]-9,9-diethylfluorene(BTPDF) and 2,7-Bis[(2,3,4,5-tetraphenyl)phenyl]-9,10-anthracene(BTPA), which consist of a diethylfluorene and an anthracene with two tetraphenylbenzene moieties, were synthesized by Diels-Alder reaction and characterized to investigate electroluminescent(EL) behavior.

BTPDF and BTPA showed violet and blue PL spectra at 400 nm and 456 nm. The device of m-MTDATA(600 Å)/NPB(150 Å)/BTPDF or BTPA(300 Å)/Alq₃(300 Å)/LiF(10 Å)/Al(2000 Å) showed turn-on voltage of 9 V and 13 V and blue and green EL spectrum at 466 nm and 504 nm, respectively.

Keywords: anthracene; electroluminescence; fluorene; highly phenyl substituted molecules

Received 1 October 2003; accepted 5 February 2004.

The author (K. Y. K.) wishes to acknowledge the financial support from BK21 project in 2003, and this work was supported by grant No. R01-2002-000-00347-0 from the interdisciplinary research program of the KOSEF.

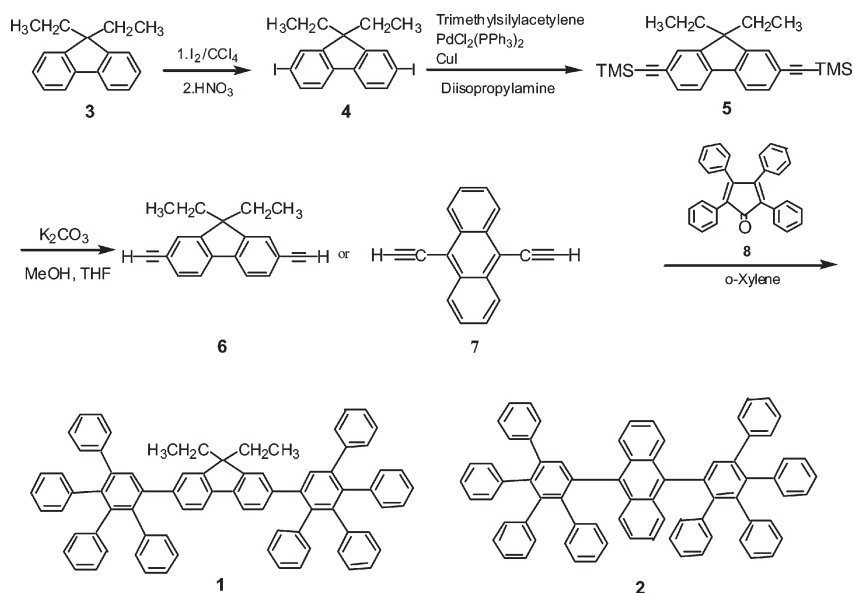
Address correspondence to Jong-Wook Park, Department of Chemistry/Center for Nanotechnology Research, The Catholic University, Puccheon 420-734, Korea. Tel.: +82-32-340-3821, Fax: +82-32-340-3764, E-Mail: hahapark@catholic.ac.kr

1. INTRODUCTION

Organic light-emitting devices (OLEDs) have attracted a great attention due to their promising applications in electroluminescent displays since the first report of efficient emission from amorphous molecular materials by Tang and VanSlyke [1–3]. Thereafter, considerable efforts have been focused on obtaining amorphous glassy materials with high EL efficiency, which have stable nonplanar molecular structures bearing bulky and heavy substituents such as polyphenylene [4,5] or triarylamine [6] moieties. In line with these aspects we report here the first synthesis and characterization of 2,7-bis[(2,3,4,5-tetraphenyl)phenyl]-9,9-dioctylfluorene (BTPDF, **1**) and 2,7-bis[(2,3,4,5-tetraphenyl)phenyl]-9,10-anthracene (BTPA, **2**) having highly phenyl-substituted benzene moieties through Diels-Alder reaction. Diels-Alder reaction for synthesis of OLEDs material could be expected to have several merits such as highly pure material and high yield because of no catalyst reaction.

2. EXPERIMENTS

Synthesis of BTPDF, **1** was described in Scheme 1.



SCHEME 1 Synthesis of BTPDF(**1**) and BTPA(**2**).

2.1. 2,7-Bis[(2,3,4,5-tetraphenyl)phenyl]-9,9-diethylfluorene (BTPDF) (1)

To a solution of 2,7-diacetyl-9,9-diethylfluorene (1.01 g, 3.74 mmol) in o-xylene (150 ml), tetraphenyl-2,4-cyclopentadiene-1-one (4.13 g, 10.74 mmol) was added. The mixture was refluxed under nitrogen atmosphere for 3 d. After cooled to 80°C, the mixture was added to MeOH (200 ml) to give yellow solids. The crude products were filtered and recrystallized from MeOH/CH₂Cl₂ (1:1) to afford a solid product (1.63 g, 49.3%) (m.p. 96–98°C).

¹H-NMR(CDCl₃, 400 MHz): δ = 7.70 – 6.70 (m, 48H, Ar-H), 1.50 (q, 4H, CH₂), 0.80 (t, 6H, CH₃). FT-IR(KBr): 3054, 2962, 2917 cm⁻¹.

2.2. 2,7-Bis[(2,3,4,5-tetraphenyl)phenyl]-9,10-anthracene (BTPA) (2)

BTPA was synthesized by same synthetic procedure as BTPDF's (0.92 g, 25.0%). ¹H-NMR (CDCl₃, 400 MHz) δ = 7.81 ~ 6.40 (m, 50H, Ar-H). FT-IR (KBr): 3020, 2925 cm⁻¹. MS(MALDI-TOF): 938(M⁺), 523(base peak).

3. RESULT AND DISCUSSION

Figures 1 and 2 show the UV-absorption and photoluminescence (PL) spectra of the two emission layers in thin films on quartz plates. The maximum absorption of BTPDF(**1**) appeared at 328 nm, and the absorption edge of BTPDF(**1**) at 367 nm. For BTPA(**2**), the maximum absorption appeared at 408 nm and the absorption edges at 426 nm.

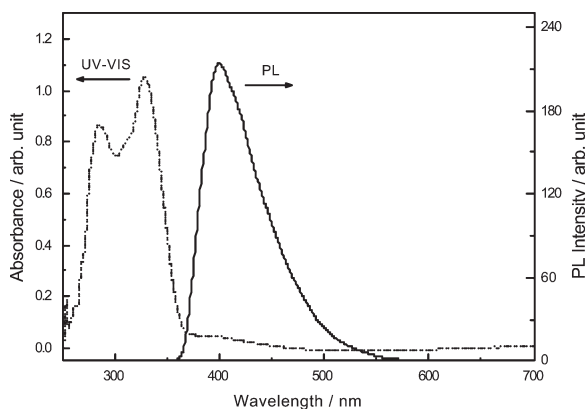


FIGURE 1 UV-Visible spectrum and PL spectrum of BTPDF (excitation wavelength = 328 nm).

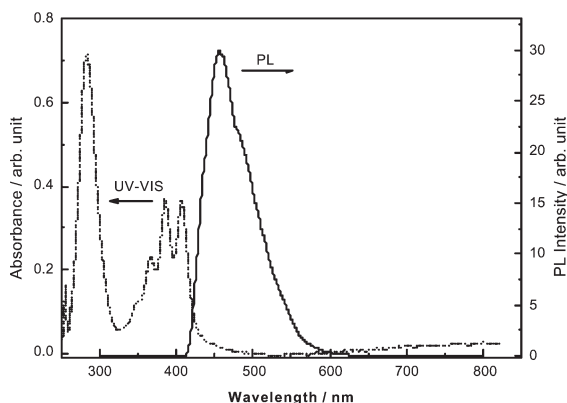


FIGURE 2 UV-Visible spectrum and PL spectrum of BTPA (excitation wavelength = 408 nm).

The energy band gap of BTPDF and BTPA was 3.50 eV and 2.97 eV according to UV absorption edge calculation, a plot of (hv) vs. $(\alpha hv)^2$.

Also, BTPDF and BTPA showed violet and blue PL spectra at 400 nm and 456 nm, respectively. This PL difference might be due to the different interaction between highly phenylated moieties and fluorene moiety and anthracene moiety. More studies on aggregation effect of thin films by through PL experiments are underway.

We measured I-V-L characteristics and EL spectrum of related EL devices. The device structure was ITO 150 nm/m-MTDATA 60 nm/ α -NPD 15 nm/BTPDF or BTPA 30 nm/Alq₃ 30 nm/LiF 1 nm/Al 200 nm.

Figure 3 shows forward-biased current-voltage(I-V) intensity curves for the undoped organic EL device. These curves were obtained when the devices were operated in a continuous direct current (dc) mode under forward bias (positive voltage applied to the ITO contact). These devices exhibit diode characteristics, that is, forward current increases with increasing forward bias voltage. The turn-on voltages of BTPA and BTPDF devices are about 9 V and 13 V, respectively.

Figure 4 shows the EL spectra of BTPDF and BTPA devices. BTPA device showed real blue EL spectrum at 466 nm and BTPDF showed green EL spectrum at 504 nm which might be originated from Alq₃ layer or interface layer between BTPDF and Alq₃. It could be due to electron and hole recombination at Alq₃ layer or excimer or electroplex emission between BTPDF and Alq₃. For making recombination at BTPA inside layer, the thickness of Alq₃ layer could be decreased or the thickness of BTPA layer could be increased much more. Further studies of EL property are underway. We summarized EL properties in Table 1.

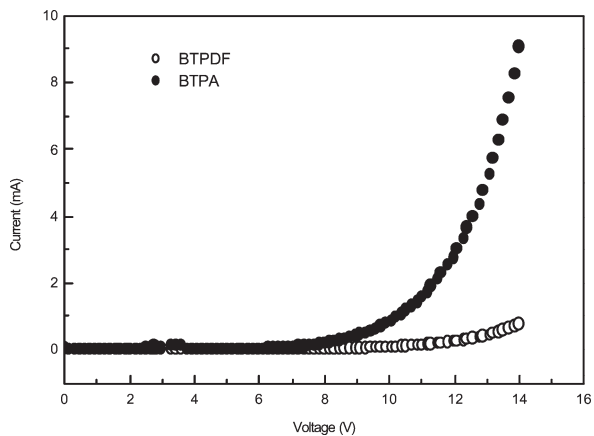


FIGURE 3 Current-voltage(I-V) characteristics of BTPDF and BTPA devices.

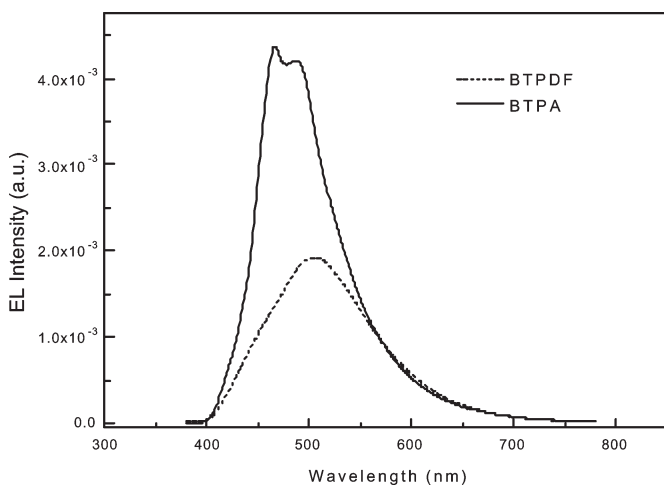


FIGURE 4 EL spectra of devices using BTPDF and BTPA as an emissive layer.

TABLE 1 Luminescent Properties of the Device (at 10 mA/cm²) using BTPDF(**1**) and BTPA(**2**) as an Emitting Layer

Emitting material	EL peak [nm]	Efficiency [lm/W] @ 120 cd/m ²	Efficiency [cd/A]	Voltage [V] @ 120 cd/m ²	CIE coordinate	
					<i>x</i>	<i>y</i>
BTPDF	504	0.18	0.82	13.7	0.248	0.384
BTPA	466	0.37	1.10	9.30	0.190	0.283

BTPDF and BTPA devices showed 0.82 cd/A and 1.10 cd/A with the CIE chromaticity coordinates of (0.248, 0.384) and (0.190, 0.283), respectively.

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