

Alkyl free design of anthracene based host material for solution processed blue fluorescent organic light-emitting diodes



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ABSTRACT

An alkyl free anthracene based host material, 9-([1,1':2',1''-terphenyl]-2-yl)-10-phenylanthracene (AnTP), was developed as a soluble host material for solution processed blue fluorescent organic light-emitting diodes by increasing the solubility of anthracene type host material using a twisted aromatic substituent. The good solubility of AnTP even without any alkyl substituent allowed solution coating of AnTP based emitting layer and fabrication of solution processed blue fluorescent organic light-emitting diodes with a current efficiency of 3.7 cd/A.

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1. Introduction

Solution processed organic light-emitting diodes (OLEDs) are attractive as next generation OLEDs surpassing current vacuum processed OLEDs in terms of material usage, production cost, and large size pixel patterning. In particular, they can be a disrupting technology in the large size OLED panel because the fabrication of large size panel by vacuum process is very challenging. However, the technology for solution processed OLEDs is immature for practical application and several core technologies are under development.

One of core technologies for the soluble OLEDs is the development of solution processed fluorescent blue OLEDs [1,2]. Although phosphorescent OLEDs can provide high quantum efficiency (QE), instability of the phosphorescent OLEDs makes them impractical in real applications [3–7]. Therefore, more effort should be directed to develop high performance blue fluorescent OLEDs rather than blue phosphorescent OLEDs for solution process application. However, it was very challenging to develop soluble blue fluorescent OLEDs due to intrinsic poor solubility of common blue host material built on an anthracene core structure. The anthracene is the most famous building unit of the blue fluorescent host material [8–11], but planar and aromatic nature of the anthracene moiety was a hurdle to solubilize the anthracene type

host material. Although alkyl modification approach could solubilize the anthracene type host material [8,12–14], it should be excluded in the molecular structure due to intrinsic weak chemical bonding of the alkyl moiety to the anthracene backbone structure. However, only the alkyl modification method could assist increasing the solubility of the host material and coating of the host material by solution process. Therefore, a different approach solubilizing the anthracene type host material is desired.

In this work, an anthracene derived host material, 9-([1,1':2',1''-terphenyl]-2-yl)-10-phenylanthracene (AnTP), was developed as a soluble host material for blue fluorescent OLEDs. An *ortho*-linked terphenyl was a solubilizing unit of AnTP for better solubility in aromatic solvent and the AnTP host material realized good solubility and smooth film morphology. The solution processed AnTP host material allowed current efficiency of 3.7 cd/A by doping 2,5,8,11-tetra-*tert*-butylperylene (TBPe) as a fluorescent dopant.

2. Experimental

2.1. General information

2,2'-Dibromo-1,1'-biphenyl, phenylboronic acid, *n*-butyl-lithium solution (2.5 M in *n*-hexane) and triethyl borate from Sigma Aldrich Co. were used without purification. 9-Bromo-10-phenylanthracene and tetrakis(triphenylphosphine) palladium(0) were received from P&H Tech Co. Potassium carbonate, magnesium sulfate anhydrous, hydrochloric acid, methylene chloride, *n*-hexane and tetrahydrofuran were purchased by Duksan Sci. Co.

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2.2. Synthesis

2.2.1. 2-Bromo-1,1':2',1''-terphenyl

2,2'-Dibromo-1,1'-biphenyl (1.17 g, 9.60 mmol) and phenylboronic acid (15.0 g, 48.08 mmol) were dissolved in tetrahydrofuran (40 mL) in three neck round-bottomed flask. After bubbling N₂ gas for 15 min, a solution of potassium carbonate (5.53 g, 40.01 mmol) in distilled water (20 mL) was added to the solution and the flask was filled with N₂ gas. After 20 min, tetrakis(triphenylphosphine) palladium(0) was added to the flask followed by refluxing for 10 h and cooling to room temperature. The reaction mixture was extracted with methylene chloride and distilled water. The organic layer was dehydrated with magnesium sulfate anhydrous and the solvent was removed using rotary evaporator after filtering the magnesium sulfate. A crude product was purified by column chromatography (methylene chloride: *n*-hexane = 1:6) to obtain a transparent liquid [7].

2.2.2. [1,1':2',1''-terphenyl]-2-ylboronic acid

2-Bromo-1,1':2',1''-terphenyl (0.62 g, 2.01 mmol) was dehydrated under a vacuum condition for 24 h and was dissolved in dried tetrahydrofuran (40 mL). The solution was cooled to -78 °C under a N₂ environment. *n*-Butyllithium (0.94 mL, 2.34 mmol) was added to the solution by dropping and the solution was stirred at -78 °C for 1 h. Triethyl borate (0.41 mL, 2.40 mmol) was dropped in the reaction mixture and it was warmed to room temperature overnight. After that, the solution was quenched with 2 M HCl. Solvent was removed by evaporation and *n*-hexane was added. A white powder was precipitated and it was filtered followed by drying without further purification (0.60 g). MS (*m/z*): 274.2 ([M+H]⁺).

2.2.3. 9-([1,1':2',1''-terphenyl]-2-yl)-10-phenylanthracene (AnTP)

9-([1,1':2',1''-terphenyl]-2-yl)-10-phenylanthracene (AnTP) was synthesized according to the synthetic method of 2-bromo-1,1':2',1''-terphenyl except the reactants. The reactants were [1,1':2',1''-terphenyl]-2-ylboronic acid (0.60 g, 2.19 mmol) and 9-bromo-10-phenylanthracene (0.88 g, 2.64 mmol). The product after purification was a yellow powder (0.83 g, yield 65.4%). ¹H NMR (300 MHz, DMSO-*d*₆): δ 7.63–7.50 (m, 6H), 7.41 (d, 2H, *J* = 9.3 Hz), 7.35 (d, 3H, *J* = 6.3 Hz), 7.30–7.21 (m, 5H), 7.10–6.89 (m, 7H), 6.80 (d, 1H, *J* = 7.5 Hz), 6.70 (d, 2H, *J* = 6.9 Hz). ¹³C NMR (125 MHz, chloroform): δ 142.98, 141.91, 140.81, 139.51, 138.00, 137.04, 135.83, 133.01, 132.81, 132.08, 131.58, 130.39, 129.89, 129.66,

129.43, 128.47, 127.71, 127.55, 127.51, 127.11, 126.67, 126.63, 126.27, 124.57. MS (*m/z*): 483.3 ([M+H]⁺).

2.3. Device fabrication and measurements

Device structure was described below.

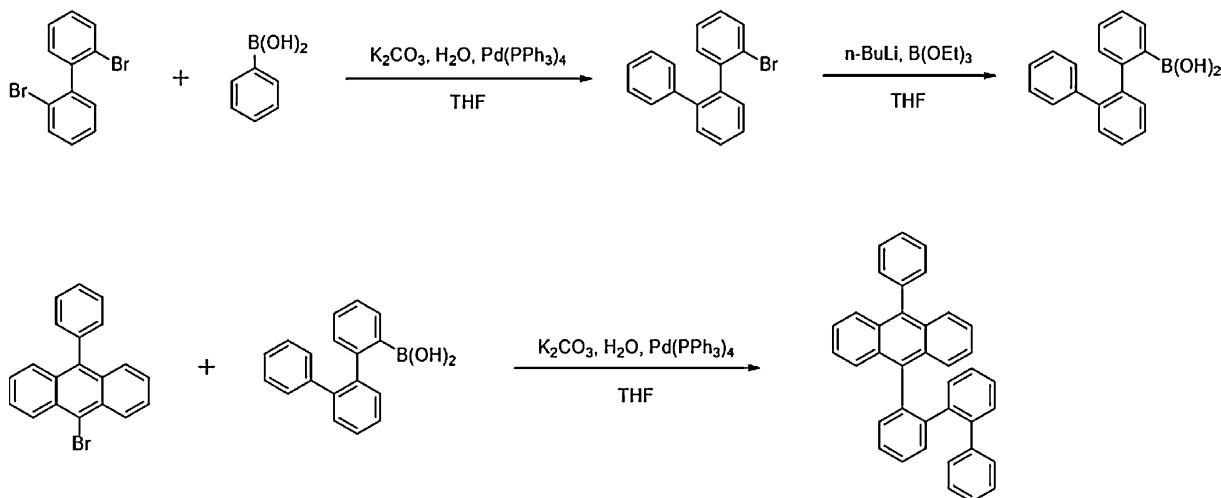
ITO (120 nm)/PEDOT:PSS (60 nm)/PVK (15 nm)/AnTP:TBPe (25 nm)/TSPO1 (5 nm)/TPBi (30 nm)/LiF (1 nm)/Al (200 nm).

Poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) was diluted with 2-propanol and stacked on the indium tin oxide (ITO) substrate by spin coating at a rate of 2000 rpm. After that, the PEDOT:PSS coated sample was annealed for 10 min on a hot plate at 140 °C. Poly(9-vinylcarbazole) (PVK) was coated at the same spin speed and the PVK film was annealed for 20 min at 120 °C under a N₂ gas. Molecular weight of PVK was 1,000,000 to avoid intermixing between PVK and spin coated emitting layer. After coating of PVK, 1.0 wt% toluene solution of AnTP and TBPe was spin coated on the PVK layer. The AnTP:TBPe emitting layer was annealed for 20 min at 80 °C under a N₂ gas. Diphenyl(4-(triphenylsilyl)phenyl)phosphine oxide (TSPO1), 1,3,5-tris(N-phenylbenzimidazole-2-yl)benzene (TPBi), LiF and Al were layered by evaporation.

3. Results and discussion

The host materials of solution processed blue fluorescent OLEDs need to be highly soluble in aromatic solvent and molecular design approach adopted was to attach *ortho*-linked terphenyl as a side group of anthracene. As the solubility of host material is improved by penetration of solvent molecules between host molecules, the *ortho*-linked terphenyl was introduced as a bulky side group separating host molecules. Although the *ortho*-terphenyl has only aromatic units, the *ortho*-linkage between aromatic units distorts the *ortho*-terphenyl moiety, which increases free volume between molecules. Therefore, the solubility of the AnTP host material can be increased compared to that of other anthracene type host materials such as a well-known MADN or ADN. In fact, 2-methyl-9,10-di(naphthalene-2-yl)anthracene (MADN) or 9,10-di(naphthalene-2-yl)anthracene (AND) host material was not soluble in aromatic solvent like toluene, but the AnTP host was dissolved in toluene solvent up to 23.3 wt%.

Synthesis of AnTP was simply carried out by Suzuki coupling of a functionalized anthracene with a functionalized *ortho*-terphenyl as clearly presented in Scheme 1. The simple coupling reaction



Scheme 1. Synthetic scheme of AnTP.

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