Ultrahigh-efficiency solution-processed simplified small-molecule organic light-emitting diodes using universal host materials

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Although solution processing of small-molecule organic light-emitting diodes (OLEDs) has been considered as a promising alternative to standard vacuum deposition requiring high material and processing cost, the devices have suffered from low luminous efficiency and difficulty of multilayer solution processing. Therefore, high efficiency should be achieved in simple-structured small-molecule OLEDs fabricated using a solution process. We report very efficient solution-processed simple-structured small-molecule OLEDs that use novel universal electron-transporting host materials based on tetraphenylsilane with pyridine moieties. These materials have wide band gaps, high triplet energy levels, and good solution processabilities; they provide balanced charge transport in a mixed-host emitting layer. Orange-red (~97.5 cd/A, ~35.5% photons per electron), green (~101.5 cd/A, ~29.0% photons per electron), and white (~74.2 cd/A, ~28.5% photons per electron) phosphorescent OLEDs exhibited the highest recorded electro-luminescent efficiencies of solution-processed OLEDs reported to date. We also demonstrate a solution-processed flexible solid-state lighting device as a potential application of our devices.

RESULTS
We synthesized electron-transporting host materials for solution-processed phosphorescent orange-red, green, blue, and white OLEDs; these materials are diphenylbis(3-(pyridine-2-yl)phenyl)silane (2PTPS), diphenylbis(3-(pyridine-3-yl)phenyl)silane (3PTPS), and diphenylbis(3-(pyridine-4-yl)phenyl)silane (4PTPS) with the pyridine moieties in various orientations (Fig. 1). Synthesis of 2PTPS, 3PTPS, and 4PTPS began with lithiation of 1,3-dibromobenzene by using n-butyllithium and subsequent treatment with dichlorodiphenylsilane to give bis(3-bromophenyl)diphenylsilane. 2PTPS was synthesized by Stille cross-coupling reaction between 2-(tributylstannyl)pyridine and bis(3-bromophenyl)diphenylsilane. 3PTPS and 4PTPS were synthesized by Suzuki cross-coupling reaction between pyridineboronic acid and bis(3-bromophenyl)diphenylsilane. The identities of 2PTPS, 3PTPS, and 4PTPS were confirmed using nuclear magnetic resonance (NMR) and mass analysis (Supplementary Materials). 2PTPS, 3PTPS,
and 4PTPS have high thermal stability with high glass transition temperature \(T_g\) (table S1).

To measure the electrochemical properties and energy levels of materials, we performed cyclic voltammetry (CV) in a conventional three-electrode configuration in acetonitrile containing 0.1 M tetrabutylammonium perchlorate (Bu4NClO4) as the supporting electrolyte versus an Ag/AgCl platinum disc as the working electrode and platinum wire as the counter electrode (fig. S1). The estimated highest occupied molecular orbital (HOMO) levels of 2PTPS, 3PTPS, and 4PTPS were \(-6.47, -6.50,\) and \(-6.55\) eV, respectively, and the lowest unoccupied molecular orbital (LUMO) levels were \(-2.41, -2.30,\) and \(-2.27\) eV, respectively (Table 1 and figs. S1 to S4). Lower HOMO and higher LUMO energies of the host than the corresponding energies of the blue phosphorescent dopant molecule \{bis\[2-(4,6-difluorophenyl)pyridinato-N, C2\] (picolinato)iridium (FIrpic), \(-5.90\) and \(-3.00\) eV\} are expected to favor energy transfer from the host to the emitting dopant. Phosphorescence of 2PTPS, 3PTPS, and 4PTPS was measured in a CHCl3 matrix at 77 K (fig. S5). The highest-energy 0-0 phosphorescent emissions located at 2.82, 2.82, and 2.90 eV for 2PTPS, 3PTPS, and 4PTPS, respectively, were used to calculate their triplet energy \(E_T\) (Table 1).

\(E_T\)’s of the new host materials are higher than those of the conventional blue dopant, FIrpic (~2.7 eV) (29), and sufficient for the material to be used as a host material for blue phosphorescent dopant. For a favorable radiative exciton recombination without backward energy transfer from dopant to host, host materials should have higher \(E_T\) than the blue-emitting phosphorescent dopant (29).

To achieve highly efficient simple-structured OLEDs without additional hole-transporting or electron-blocking interfacial layers, we used two approaches in this work: (i) a high work function solution-processed hole injection layer (HIL) that blocks exciton quenching at the HIL/EML interface (30) and (ii) a mixed-host EML that consists of electron- and hole-transporting host materials. Because host materials in the EML have deep HOMO energy levels [for example, \(4,4',4''\)-tris(N-carbazolyl)-triphenylamine (TCTA) \(-5.7\) to \(-5.9\) eV], the large energy barrier at the HIL/EML interface must be overcome to achieve balanced charge injection to the EML without the need for an additional hole-transporting interlayer. Therefore, we used a polymeric HIL, which develops a gradient work function that increases from the anode to the top surface of the HIL by self-organization and that has a very high surface work function (~5.95 eV) (figs. S6 and S7 and table S2). Our gradient HIL (GraHIL) is composed of conventional conducting polymer, poly(3,4-ethylenedioxythiophene) doped with poly(styrenesulfonate) (PEDOT/PSS), and tetrafluoroethylene-perfluoro-3,6-dioxa-4-methyl-7-octenesulfonic acid copolymer [a perfluorinated ionomer (PFI)] in 1:3.6 weight ratio. The GraHIL facilitates hole injection from the anode to an overlying EML by reducing the large energy barrier between the anode and the EML in the simple structure. In addition, enrichment of PFI in the surface layer effectively blocks exciton quenching at the HIL/EML interface (30, 31). The solution-processed EML consists of a 1:1 weight mixture of a hole-transporting host material (TCTA) and an electron-transporting host material, which is selected from among 2PTPS, 3PTPS, 4PTPS, or conventionally used solution-processable small-molecule host material \{2,2,2-(1,3,5-benzenetriyl)tris-[1-phenyl-1H-benzimidazole] (TPBI)\}. The bipolar mixed-host EML consists of electron- and hole-transporting hosts to facilitate charge-carrier injection to the EML and balanced charge transport to a recombination zone in the EML (32, 33). In addition, the bipolar characteristics of this mixed-host system easily distribute excitons in

![Fig. 1. Synthesis routes of 2PTPS, 3PTPS, and 4PTPS.](http://advances.sciencemag.org/)
the EML over a broad recombination zone that can effectively avoid nonradiative exciton quenching, such as triplet-triplet annihilation and triplet-polaron annihilation (32, 33).

To evaluate our phosphorescent host materials, we fabricated simple-structured orange-red, green, and blue phosphorescent OLEDs (Fig. 2A) based on phosphorescent dopants, bis(2-phenyl benzothiazolato-N,C2′)iridium(acetylacetonate) [Bt2Ir(acac)], tris[2-phenylpyridinato-C2,N]iridium(III) [Ir(ppy)3], and Flrpic, respectively. Compared to devices with a conventional electron-transporting host (TPBI), all Bt2Ir(acac)-based orange-red phosphorescent OLEDs with our novel tetraphenylsilane host exhibited superior electrical and luminescent characteristics (Fig. 2, B and C). Solution-processed orange-red phosphorescent OLEDs with our host materials achieved much higher luminous current efficiencies (CEs) and external quantum efficiencies (EQEs) (device with 2PTPS ~95.7 cd/A, 35.4%; 3PTPS ~97.5 cd/A, 35.5%; 4PTPS ~76.4 cd/A, 28.0%) than did those with TPBI (~60.6 cd/A, 21.2%) (Fig. 2C, figs. S8 to S10, and Table 2). These high electroluminescent efficiencies of orange-red OLEDs can originate from the chemical structure of phosphorescent emitter molecule [Bt2Ir(acac)], in which the orientation of the transition dipole moment is preferentially horizontal (34). OLEDs that use an EML in which emitter molecules are aligned horizontally have higher optical outcoupling efficiency than those that use an EML in which emitter molecules are distributed isotropically (34–36). A possible additional source of increased electroluminescent efficiency is a weak microcavity effect caused by the combination of the low refractive index of the GraHIL (n ~1.4) and the high refractive index of indium tin oxide (ITO) (n ~1.9) in our device architecture (37). Devices with 2PTPS and

![Diagram](http://advances.sciencemag.org/)
3PTPS showed better electrical characteristics and higher efficiencies than did those with 4PTPS. Therefore, we also fabricated Ir(ppy)$_3$-based green phosphorescent OLEDs with 2PTPS and 3PTPS. Green-emitting devices that used our host materials also exhibited very high device efficiencies (device with 2PTPS ~101.5 cd/A, 29.0%, 3PTPS ~88.5 cd/A, 23.7%), which were also higher than those with TPBI (~79.5 cd/A, 19.9%) (Fig. 2D). These very high CEs and EQEs of orange-red and green phosphorescent OLEDs are the highest recorded in solution-processed small-molecule OLEDs in a simplified structure.

3PTPS showed better electrical characteristics and higher efficiencies than did those with 4PTPS. Therefore, we also fabricated Ir(ppy)$_3$-based green phosphorescent OLEDs with 2PTPS and 3PTPS. Green-emitting devices that used our host materials also exhibited very high device efficiencies (device with 2PTPS ~101.5 cd/A, 29.0%; 3PTPS ~32.6 cd/A, 19.6%; 4PTPS ~28.8 cd/A, 14.3%) and those with TPBI (~18.6 cd/A, 10.0%) than did those of other color-emitting devices (Fig. 2E, fig. S10, and Table 2). The larger difference between blue OLEDs with our host materials and those with TPBI than the differences between green and orange-red OLEDs with our host materials and those with TPBI can be attributed to an additional origin: the higher $E_T$ of our host materials, which can serve effective triplet exciton confinement on a phosphorescent dopant by blocking backward energy transfer to host materials (Flrpic, $E_T$ ~2.7 eV) (32, 33). The peaks in the EL spectra of OLEDs with 2PTPS were at ~563 nm in the B$_2$Ir(acac)-based orange-red OLED, at ~513 nm in the Ir(ppy)$_3$-based green OLED, and at ~472 nm in the Flrpic-based blue-emitting OLED (Fig. 2F).

The conventional TCTA/TPBI mixed-host forms exciplexes by intermolecular interaction, and its major recombination mechanism is energy transfer from the exciplexes to phosphorescent dopants (figs. S11 to S13) (38–40). In contrast, the dominant exciton generation mechanism of exciplex-free TCTA/2PTPS is direct recombination in phosphorescent dopants by charge-carrier trapping on dopant molecules (figs. S11 to S13). Direct charge injection and trapping on phosphorescent dopants in TCTA/2PTPS facilitate balanced charge transport and efficient direct recombination in TCTA/2PTPS mixed-host EML (fig. S13) (39–42). Relatively low electroluminescent efficiencies of blue phosphorescent OLEDs that use Flrpic occur because it has a deeper HOMO energy level (~5.9 eV) than B$_2$Ir(acac) (~5.6 eV) and Ir(ppy)$_3$ (~5.2 eV) (43–45). The deepest HOMO energy level of Flrpic reduces hole injection from HIL, whereas Ir(ppy)$_3$ and B$_2$Ir(acac) have shallower HOMO energy levels than the host materials and therefore facilitate hole injection by direct injection of holes to the dopants (figs. S14 to S16). The reduction in hole injection can cause a charge imbalance in EML and thereby reduce device efficiencies of OLEDs.

To investigate electrical properties of devices with various electron-transporting hosts, we performed several kinds of electrical analyses. The capacitance-voltage (C–V) characterization of OLEDs provides information about injection and accumulation of charge carriers under varying applied voltages. A sharp increase of capacitance at low voltage suggests accumulation of major charge carriers that have been injected and transported from the electrode to the EML (45). The accumulated major charge carriers start recombining with injected minor carriers at high voltage, so the capacitance between electrodes starts decreasing (46, 47). Solution-processed phosphorescent orange-red OLED with 2PTPS exhibited peak capacitance at the lowest voltage (Fig. 3A); this result implies that OLEDs with 2PTPS/TCTA EML have the fastest charge transport and best-balanced electron-hole recombination, so they had both the lowest operating voltages and the highest device efficiencies of OLEDs (Fig. 2, B and C). The voltage of the peak capacitance ($V_{peak}$) of the devices with 2PTPS, 3PTPS, 4PTPS, and TPBI increased in the order 2PTPS < 3PTPS < 4PTPS < TPBI (Fig. 3A).

To prove that the use of electron-transporting host materials affected both the electrical and luminescent properties of solution-processed OLEDs, we also performed transient EL characterization (Fig. 3B). Transient increase of EL signal directly represents the charge transport and recombination characteristics while an applied voltage pulse causes OLEDs to emit light. EL increased much faster with our host materials than it did with the TPBI; this means that they have better electron-transporting capability than the TPBI. Because delayed electron transport shifts the recombination zone toward the cathode, the traveling distance of the holes is extended in OLEDs; this increase in traveling distance of hole carriers delays the increase in EL during transient EL characterization. Therefore, the results of transient EL also imply that TCTA/2PTPS, TCTA/3PTPS, or TCTA/4PTPS mixed-host EMLs have better electron-transporting capability than TCTA/TPBI EML in solution-processed OLEDs.

### Table 2. Device efficiencies of solution-processed OLEDs.

|        | $C_{E_{\text{max}}}$ (cd/A) | $E_{Q_{\text{max}}}$ (%) |
|--------|----------------------------|--------------------------|
| **2PTPS** |
| Orange | 95.7                       | 35.4                     |
| Green  | 101.5                      | 29.0                     |
| Blue   | 38.7                       | 20.9                     |
| White  | 61.5                       | 23.1                     |
| **3PTPS** |
| Orange | 97.5                       | 35.5                     |
| Green  | 88.5                       | 23.7                     |
| Blue   | 32.6                       | 19.6                     |
| White  | 74.2                       | 28.5                     |
| **4PTPS** |
| Orange | 76.4                       | 28.0                     |
| Green  | 28.8                       | 14.3                     |
| Blue   | —                          | —                        |
| White  | —                          | —                        |
| **TPBI** |
| Orange | 60.6                       | 21.2                     |
| Green  | 79.5                       | 19.9                     |
| Blue   | 18.6                       | 10.0                     |
| White  | 40.5                       | 15.4                     |
We also fabricated an electron-only device (EOD) [ITO/branched polyethylenimine (PEI) (10 nm)/solution-processed electron-transporting host layer (100 nm)/TPBI (50 nm)/LiF (1 nm)/Al (100 nm)] (Fig. 3C) to investigate the electron-transporting ability of our electron-transporting host materials in the absence of a hole-transporting host (TCTA). A spin-cast thin PEI layer is an effective electron injection layer in OLEDs and an efficient extraction layer in organic photovoltaics (3, 48). Because a thin PEI interfacial layer induces a shift in the vacuum level by surface dipoles, the effective work function of an underlying electrode is reduced, so electrons are easily injected into overlying layers (3, 48). However, we adopted a thin PEI interfacial layer to block the hole injection from the anode into the electron-transporting host layer by reducing the work function of the anode (ITO). Because the deep HOMO energy level (≤6 eV) of electron-transporting hosts and the reduced work function of the ITO anode can effectively inhibit hole injection, we can demonstrate only electron-transporting properties with solution-processed EOD architecture. Results of electron current densities in EODs have identical order with current densities of OLEDs: the OLEDs with 2PTPS exhibited the highest electron current density, and TPBI showed the lowest electron current density (that is, 2PTPS > 3PTPS > 4PTPS > TPBI) (Fig. 3D). The higher electron current density with our host materials means that our host materials have higher electron mobility ($\mu_e$) in the solution-processed film than TPBI. To prove higher electron mobility of our electron-transporting host materials, we additionally performed impedance spectroscopy measurement to determine $\mu_e$ of electron-transporting materials (figs. S17 and S18). Calculated $\mu_e$'s of TPBI were $\sim 10^{-5}$ cm$^2$/V·s, which concurs with those measured by using time of flight in a previous report (fig. S18) (49). $\mu_e$ of 2PTPS according to electric field range was on the order of $\sim 10^{-4}$ cm$^2$/V·s, which is an order higher than that of TPBI. $\mu_e$ also increased in the order 2PTPS > 3PTPS > 4PTPS > TPBI (fig. S18); this order is identical with that of current density in solution-processed EODs and OLEDs. The higher $\mu_e$ of our electron-transporting host materials increases electron transport in the EML that includes a hole-transporting host (TCTA) [$\mu_h$ of TCTA, $\sim 3 \times 10^{-4}$ cm$^2$/V·s] (50), thereby leveling the charge-carrier balance and broadening the electron-hole recombination zone in the EML. In contrast, the use of a conventional TCTA/TPBI EML provides relatively poor electron-hole balance because TPBI has lower $\mu_e$ ($\sim 10^{-5}$ cm$^2$/V·s) than $\mu_h$ of TCTA (49).

White OLEDs (WOLEDs) were fabricated using the same methods as those used to fabricate single-color phosphorescent OLEDs except that the EML was composed of two phosphorescent dopants of complementary colors: blue-emitting FIrpic and orange-red-emitting Bt2Ir(acac) (Fig. 4A). The WOLEDs with 2PTPS and 3PTPS also exhibited superior current density–voltage characteristics (Fig. 4B) and higher CEs and EQEs (device with 2PTPS ~61.5 cd/A, 23.1%; 3PTPS ~74.2 cd/A, 28.5%), than did those with TPBI (~40.5 cd/A, 15.4%) (Fig. 4C, fig. S10, and Table 2); these are the highest recorded device efficiencies among solution-processed WOLEDs without light outcoupling structure reported to date. All WOLEDs fabricated in this work showed good white spectra (Fig. 4D) and Commission Internationale de l’Eclairage chromaticity coordinates of (0.37, 0.46) (color-rendering index, 53) at a normal incidence. Finally, we fabricated highly efficient solution-processed flexible solid-state lighting devices on 5 cm × 5 cm polyethylene terephthalate (PET) substrate (Fig. 4E), which demonstrates a strong potential application of solution-processed OLEDs for solid-state lightings.
DISCUSSION

We report efficient simple-structured phosphorescent orange-red, green, blue, and white OLEDs fabricated by a solution process using universal electron-transporting host materials with tetraphenylsilane and pyridine moieties. The wide energy band gaps (4.06 to 4.28 eV) and high $E_T$’s (2.82 to 2.90 eV) of our electron-transporting host materials allow efficient exciton confinement in the EML; the good electron-transporting ability of the materials can provide balanced charge-carrier transport in the mixed-host EML. In C-V characterization, the low $V_{\text{peak}}$’s of OLEDs with our materials proved their superior electron-transporting abilities, and faster EL rising in transient EL characterization also demonstrated enhanced electron transport of the mixed-host EML with our host materials compared to that with conventional material (TPBI). Finally, the improved electron current densities of EODs and impedance spectroscopy confirmed that they have higher electron mobilities than TPBI. In addition, the use of the polymeric HIL (GraHIL) that serves gradient and high surface work function reduces the hole injection energy barrier, efficiently blocks exciton quenching at the interface between the HIL and EML, and thereby enables a simple device structure without additional hole-transporting or electron-blocking interlayers. As a result, solution-processed orange-red (~97.5 cd/A, 35.5%), green (~101.5 cd/A, 29.0%), and white OLEDs (~74.2 cd/A, 28.5%) with our electron-transporting hosts exhibited very high electroluminescent efficiencies. We also fabricated a flexible solution-processed solid-state lighting device with our solution-processed phosphorescent WOLEDs. The wide use of our electron-transporting host materials in various-color OLEDs with high device efficiencies demonstrates that our materials and
approaches have potential applications in low-cost, printable OLEDs for solid-state lighting sources and flexible full-color displays.

**MATERIALS AND METHODS**

**Materials**

1,3-Dibromobenzene, 2-bromopyridine, tributyltin chloride, 2.5 M n-butyllithium, 1.6 M n-butyllithium, dichlorodiphenylsilane, 3-pyridineboronic acid, 4-pyridineboronic acid, and tetrakis(triphenylphosphine)palladium were purchased from Aldrich Co. and used as received. The other chemical reagents were common commercial grade and used as received.

**Material characterization**

Infrared spectra were recorded using a Genesis II FT-IR spectrometer. ¹H NMR was recorded using Avance 300-MHz NMR Bruker spectrometers, and chemical shifts were reported in parts per million with tetramethylsilane as an internal standard. Thermogravimetric analysis was performed under N₂ on a TA Instrument 2050 thermogravimetric analyzer. The sample was heated at 10°C/min from 50°C to 700°C. Differential scanning calorimetry was conducted under N₂ in a TA Instrument 2100 differential scanning calorimeter. The sample was heated at 10°C/min from 30°C to 350°C. Mass spectra were measured using a JEOL JMS-700 mass spectrometer. Ultraviolet-visible (UV-vis) absorption spectra were measured using a PerkinElmer LAMBDA-900 UV/VIS/NIR spectrophotometer and an LS-50B luminescence spectrophotometer. The cyclic voltammogram of the material was recorded on an epoxy E3 at room temperature in a 0.1 M solution of tetrabutylammonium perchlorate (Bu₄NClO₄) in acetonitrile under N₂ at a scan rate of 50 mV/s. A Pt wire was used as the counter electrode and an Ag/AgNO₃ electrode as the reference electrode.

**Device fabrication**

Patterned ITO/glass substrates were cleaned by sequential sonication in acetone and isopropanol, each for 30 min. GraHIL (blend of PEDOT/PSS and PFI in a weight ratio of 1:3.6) was coated on the cleaned ITO and baked at 150°C for 30 min. A 40-nm-thick EML was spin-coated from tetrahydrofuran (THF; SAMCHUN chemical Co.) solution in a nitrogen glove box. The EML was composed of a hole-transporting host (TCTA), an electron-transporting host, and phosphorescent dopants. Phosphorescent dopants were Bt2Ir(acac) (orange-red), Ir(ppy)₃ (green), and Flrpic (blue). After deposition of EML, a 50-nm-thick TPBI was thermally deposited under high vacuum (<5 × 10⁻⁷ torr) as an electron-transporting layer. Finally, LiF (1 nm) and aluminum (100 nm) were sequentially deposited on the TPBI layer. The devices were encapsulated using UV-curable epoxy resin.

**Device characterization**

The current-voltage-luminescence characteristics were measured using a Keithley 236 source measurement unit and a Minolta CS-2000 spectrophotometer. C-V characterizations of orange-red phosphorescent OLEDs were conducted using electrochemical impedance spectroscopy (Bio-Logic SP-300). All the devices were biased from 0 to 10 V at a constant frequency of 1000 Hz in the dark. For transient EL characterization of orange-red phosphorescent OLEDs, a constant electrical pulse (200-ms width, 100-Hz frequency) was applied to devices by using a pulse generator (HP 8116A). The emitted light was detected using a photon-counting spectrofluorometer (ISS PC1), and the EL rising output signal was monitored and recorded using an oscilloscope (Agilent Infinium 54832B DSO).

**SUPPLEMENTARY MATERIALS**

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/2/10/e1601428/DC1

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