# Blue Organic Light-Emitting Diode Based on 1,2,3,4,5-Pentaphenyl-1-(8-Phenyl-1,7-Octadiynyl)Silole

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***Abstract*—A new silole derivative, 1,2,3,4,5-pentaphenyl-1-(8-phenyl-1,7-octadiynyl)silole, is synthesized, characterized, and used as the electron-transport/emission layers in organic light-emitting diodes. Blue emission at 492 nm is observed, with a maximum luminance of 10 460 $\mathrm{cd/m^2}$ at 18 V. The respective maximum current and power efficiencies are 8.47 cd/A and 3.8 lm/W. A triple-layer composite cathode was used, consisting of tris(8-hydroxy-quinolne)aluminum $(\mathrm{Alq_3})$ lithium fluoride and aluminum. The dependence of emission efficiency on the thickness of TPD and $\mathrm{Alq_3}$ is investigated and explained.**

***Index Terms*—Interface charge, organic light-emitting diodes, quantum efficiency, silole.**

## I. INTRODUCTION

As alternatives to liquid crystals for realizing flat-panel displays (FPD) [1]–[3], different organic light-emitting diodes (OLEDs) emitting in all primary colors have been investigated. Those emitting in green are the most mature in terms of development. Both stable fluorescent hosts such as tris(8-hydroxy-quinoline)aluminum $(\mathrm{Alq_3})$ and highly efficient phosphorescent dopants [4] have been reported. Candidates for red and blue emission are less mature. Those reported for blue include dibenzochrysene derivatives [5], dipyrazolopyridine derivatives [6], aluminum chelate [7], and silacyclopentadienes [8].

Silacyclopentadienes, or siloles, belong to a group of silicon-containing conjugated rings with novel molecular structures and unique electronic properties [9]. The labile *cis*-cisoid butadiene structure attached to the silicon atom gives a fluorene-like planar and rigid ring structure. Siloles possess low-lying lowest unoccupied molecular orbital (LUMO) energy levels, associated with the $\sigma^* - \pi^*$ conjugation arising from the interaction between the $\sigma^*$ orbital of the two exocyclic bonds on the ring silicon and the $\pi^*$ orbital of the butadiene moiety [10], [11]. As a result, siloles are efficient electron-transport materials [12], with reported mobility ~100 times that of $\mathrm{Alq_3}$ [13]. The 1,1-substituents on the silicon atom affect the absorption spectra of the siloles in a unique way, such that the electronic structures and properties of the siloles can be readily tuned using molecular engineering of the inductive effects of the 1,1-substituents [9]. 2,3,4,5-tetraphenylsiloles have been reported as efficient blue emission candidates [8].

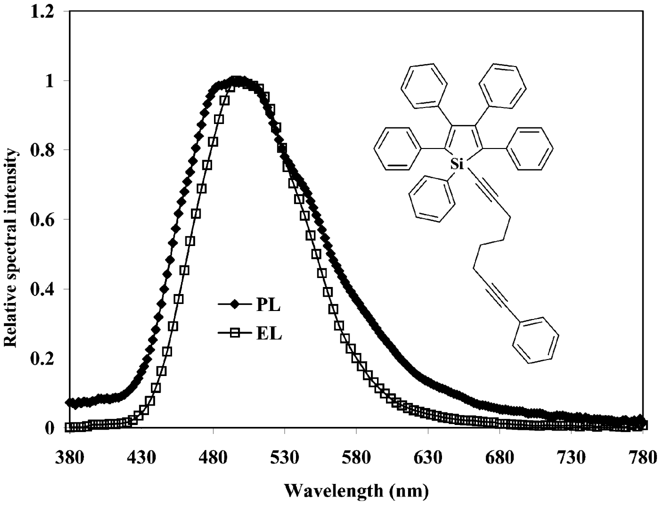

Fig. 1. PL spectrum of 50-nm PPOS thin film and EL spectrum of a Type I OLED. Inset is the chemical structure of PPOS.

In this work, a new blue-emitting silole derivative, 1,2,3,4,5-pentaphenyl-1-(8-phenyl-1,7-octadiynyl)silole (PPOS), was synthesized. The chemical structure of PPOS is shown as an inset in Fig. 1. OLEDs incorporating PPOS were fabricated and characterized. The dependence of emission efficiency on the thickness of $N$, $N'$-diphenyl-$N$, $N'$-bis(3-methylphenyl)-1, $1'$-biphenyl-4, $4'$-diamine (TPD), PPOS, and $\mathrm{Alq_3}$ was investigated and explained. Respective power and effective quantum efficiencies of ~3.8 Im/W and ~4%, comparable to the best results reported for blue emission [14], were obtained.

## II. EXPERIMENTAL DETAILS

PPOS was synthesized using the reaction of 1-chloro-1,2,3,4,5-pentaphenylsilole and 8-phenyl-1,7-octadiynyl lithium, with the former prepared using a previously reported procedure [15]. The product was subsequently purified using column chromatography.

The flexibility of the long substituent attached to the silicon atom in PPOS significantly influences its physical properties.

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TABLE I
SUMMARY OF OLED CONFIGURATIONS INVESTIGATED IN THIS WORK

| OLED type | CuPc (nm) | TPD (nm) | PPOS (nm) | $Alq_3$ (nm) |
|---|---|---|---|---|
| I | 20 | 50 | 50 | 7 |
| II | 20 | 25 | 50 | 7 |
| III | 20 | 17 | 50 | 7 |
| IV | 20 | 17 | 17 | 7 |
| Va | 20 | 25 | 50 | 7 |
| | 20 | 25 | 67 | 7 |
| Vb | 20 | 17 | 50 | 7 |
| | 20 | 17 | 84 | 7 |

PPOS has a relatively low melting point (62 °C–64 °C) when compared to those of other 2,3,4,5-tetraphenylsilole derivatives [8]. The fact that it evaporates from the liquid phase rather than sublimes from the solid phase makes it easier to control the evaporation process and to adjust the deposition rate.

For OLED fabrication, commercial 20–30 Ω indium-tin oxide (ITO) coated glass was used as the starting substrates. After glass precleaning [16], OLEDs were fabricated by sequential evaporation of the constituent organic layers in a multisource vacuum chamber. Copper (II) phthalocyanine (CuPc) was the anode buffer layer, TPD was the hole-transport layer, and PPOS was the electron-transport/emission layer. A thin layer of $Alq_3$ was also deposited as part of a composite electron-injection cathode [17]. The remaining two layers of the composite cathode, consisting of 1-nm lithium fluoride (LiF) and 150-nm aluminum (Al), were deposited in a separate series of evaporations. The configurations of the various OLEDs investigated are summarized in Table I.

The electroluminescence (EL) of OLEDs and photoluminescence (PL) of 50-nm PPOS film on quartz substrates were measured using a Kollmorgen Instrument PR650 spectrophotometer. The current–voltage characteristics were measured using a Hewlett-Packard HP4145 B semiconductor parameter analyzer.

## III. RESULTS AND DISCUSSION

The PL spectrum of 50-nm PPOS and the EL spectrum of an OLED are shown in Fig. 1. The latter, corresponding to a Type I diode defined in Table I, was constructed using a 7-nm-thick $Alq_3$ layer previously optimized for OLEDs based on 1-methyl-1,2,3,4,5-pentaphenylsilole (MPS) [17]. Both the PL and EL spectra peak at ∼492 nm, corresponding to blue emission from the PPOS layer. The EL spectrum is narrower than the PL spectrum and the relative EL spectral intensity is lower than that of PL for wavelengths longer than 540 nm, due to electrode-induced microcavity effects [18]. The peak EL luminance is 10 460 cd/m$^2$ at 18 V. The peak current efficiency is ∼8.5 cd/A, corresponding to an EL quantum efficiency of ∼4%. The peak power efficiency is ∼3.8 Im/W. These efficiencies are comparable to those for a different silole derivative reported by Murata *et al.* [19] and perylene [14].

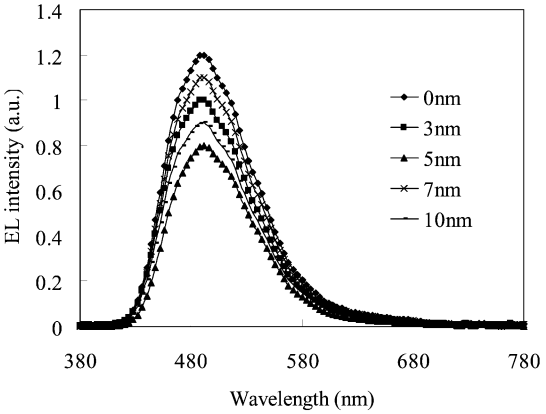


Fig. 2. Spectra of OLEDs with different $Alq_3$ thickness. All peak at 492 nm and have a similar shape. Different scaling factors were used in order to identify the lines.

### A. *Effects of Different Thickness of* $Alq_3$*, TPD, and PPOS*

The effects of varying the thickness of $Alq_3$ were studied by keeping the same thickness for all other layers but for $Alq_3$ in a Type I diode. The corresponding EL spectra are shown in Fig. 2. The similarity of the EL spectra, consistently peaking at ∼492 nm, is an indication that holes are injected to and electrons are effectively blocked on the PPOS sides of the diodes. Consequently, the recombination zone is located inside the PPOS layer. The thin $Alq_3$ contributes negligibly to the emission and, combining with LiF and Al, acts primarily as a promoter for electron injection. Such enhanced electron injection (or reduction in turn-on voltage as shown in the inset of Fig. 3) has been attributed to either Li doping of $Alq_3$ by LiF dissociation [20] or interface dipole formation [21].

The dependence of the current efficiency on $Alq_3$ thickness is shown in Fig. 3. The current efficiency is quite low without the $Alq_3$ layer. This is attributed to the low level of electron injection at the cathode/silole interface. The imbalance created by conduction dominated by holes is responsible for the low efficiency. With a thin layer of $Alq_3$ inserted between the LiF and PPOS, the imbalance is reduced and efficiency is improved. At a thickness of 7 nm, a peak efficiency of 8.5 cd/A was obtained. This optimal thickness is the same as that used in the previously reported MPS-based OLEDs [17]. With further increase of the thickness of $Alq_3$, an unexpected reduction rather than saturation of the efficiency was observed.

With the $Alq_3$ thickness fixed at 7 nm, the CuPc thickness at 20 nm, and the PPOS thickness at 50 nm, a series of OLEDs were constructed with varying TPD thickness (Type I: 50 nm, Type II: 25 nm, and Type III: 17 nm). The current efficiency (Fig. 4 and Table II), measured at a current density of 100 A/m$^2$, decreases monotonically from 7.7 cd/A for a Type I diode to 1.4 cd/A for a Type III diode, as the TPD thickness is decreased. The similarity of the EL spectra in Fig. 5 again indicates that the recombination zone is located within the PPOS layer, for all three diode types.

With the $Alq_3$ fixed at the optimum 7 nm, a different set of OLEDs was fabricated. The TPD thickness of one group (Type Va) was fixed at 25 nm, while the PPOS thickness was either 50 or 67 nm. The TPD thickness of the second group (Type Vb) was fixed at 17 nm, while the PPOS thickness was either 50

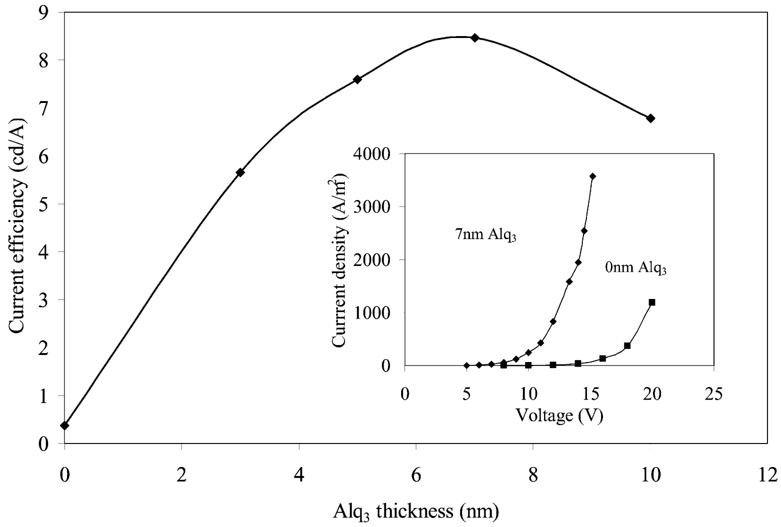


Fig. 3. Dependence on current efficiency on $\mathrm{Alq_3}$ thickness. Inset is the current–voltage characterization of OLEDs with and without $\mathrm{Alq_3}$.

TABLE II
SUMMARY OF OLED PERFORMANCE

| OLED Type | Maximum current efficiency (cd/A) | Maximum quantum efficiency (%) | Maximum power efficiency (lm/W) | Turn-on voltage (V) |
|---|---|---|---|---|
| I | 8.47 | 3.9 | 3.8 | 4.5 |
| II | 5.93 | 2.33 | 2.0 | 3.8 |
| III | 1.48 | 0.46 | 0.6 | 4.1 |
| IV | 0.32 | 0.18 | 0.14 | 3.3 |

* Defined to be the voltage required to drive a current density of 100A/m$^2$.

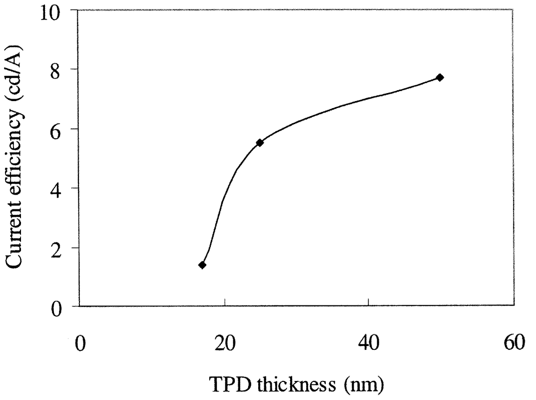


Fig. 4. Dependence of current efficiency on TPD thickness. Current density was fixed at 100 A/m$^2$.

or 84 nm. The results shown in Fig. 6 indicate that the current efficiency was always improved with thicker PPOS.

### *B. Charge Carrier Distribution and Effects of Organic Layer Thickness*

With the much smaller diffusion current neglected [22], the respective hole $(J_h)$ and electron $(J_e)$ currents in the hole-transport layer (HTL) and electron-transport layer (ETL) are

$$J_h = pq\mu_h E_h \text{ and } J_e = nq\mu_e E_e \tag{1}$$

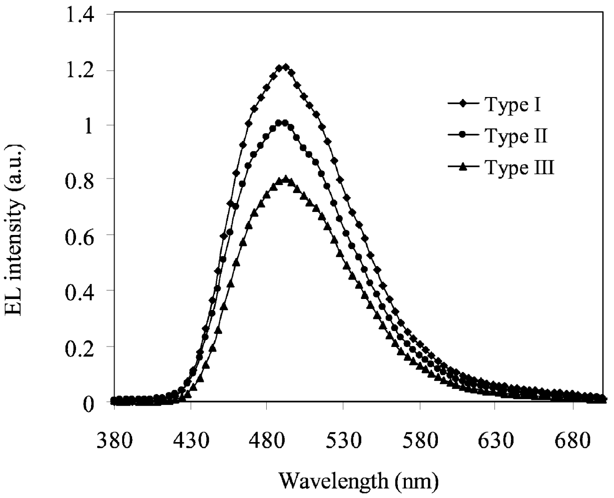
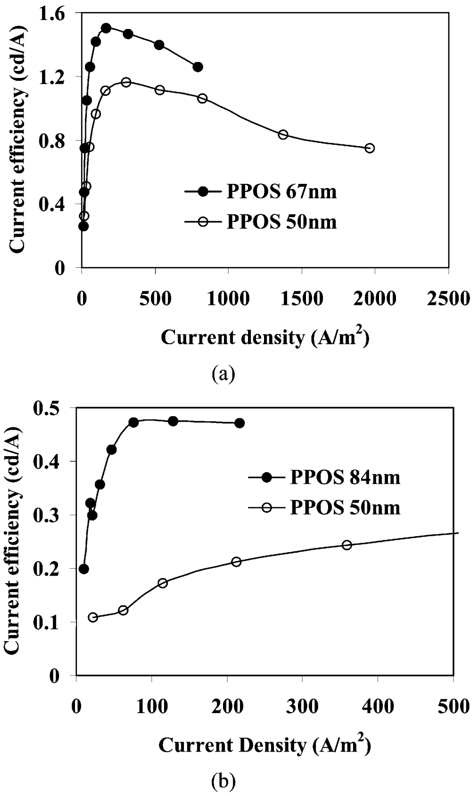


Fig. 5. Measured EL spectra of Types I, II, and III OLEDs. All peak at 492 nm and have a similar shape. Different scaling factors were used in order to identify the lines.

where $\mu_h$ and $\mu_e$ are the respective hole and electron mobility, $p$ and $n$ are the respective hole and electron concentrations within HTL and ETL, and $E_h$ and $E_e$ are the corresponding electric



(a)

(b)

Fig. 6. Current efficiencies of OLEDs with different PPOS thickness. Thickness of TPD was fixed: (a) 25 and (b) 17 nm.

field strength. The electric field strength $(E)$ is governed by Gauss' law

$$\frac{dE}{dx} = \frac{\rho}{\varepsilon} \tag{2}$$

where $\rho$ is the volume charge density and $\varepsilon$ is the permittivity. Combining (1) and (2), the spatial dependence of $E_h$ and $E_e$ can be obtained

$$E_h = \sqrt{\frac{2J_h}{\mu_h \varepsilon} x_c + c_1} \quad \text{and} \quad E_e = \sqrt{\frac{2J_e}{\mu_e \varepsilon} x_a + c_2} \tag{3}$$

where $C_1$ and $C_2$ are integration constants determined, respectively, by the boundary conditions at the cathode/organic and anode/organic interfaces. The same $\epsilon$ is assumed for both the HTL and the ETL. $x_c$ and $x_a$ are the distance from the corresponding interfaces. It should be emphasized that constant $\mu_h$ and $\mu_e$, independent of $E_h$ and $E_e$, are assumed in this simple approximate treatment.

At the interface of the HTL and ETL, discontinuity in $E_h$ and $E_e$ is accounted for by an interface charge $(\sigma)$

$$\varepsilon(E_e - E_h) = \sigma. \tag{4}$$

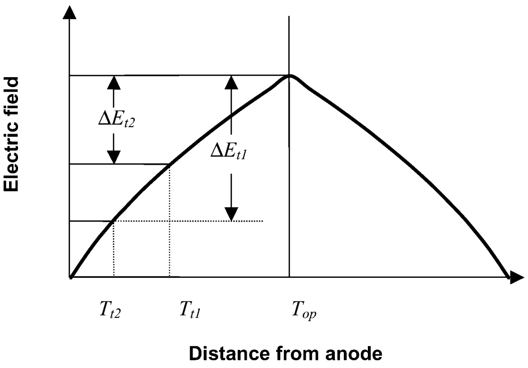


Fig. 7. Schematic drawing of electric field distribution with different HTL thickness.

For a given current density, a "balanced" injection and transport $(J_h \approx J_e)$ results in a $\sigma$ of zero. If $\mu_h \approx \mu_e$, as for TPD and PPOS, balance is achieved when the thickness of the HTL and ETL are comparable. The OLED is considered optimal and the electric field distribution is schematically illustrated in Fig. 7. $E$ is continuous across the HTL/ETL interface and $\sigma$ is zero. When the HTL thickness is decreased to $T_{t1}$ from the "optimal" thickness of $T_{\rm op}$, the electric field, lower on the HTL side $(E_{t1})$ and higher on the ETL side, is no longer continuous at the interface, leading to a finite accumulation of holes (hence, a positive $\sigma$) at the interface. The magnitude of $\sigma$ increases with further reduction in the thickness of the HTL to $T_{t2}$. If 50-nm PPOS is not thick enough for complete bulk recombination of the excess holes, the resulting "leakage" of holes to the cathode [23] leads to an obvious reduction in quantum efficiency (Fig. 4).

The reduction in the quantum efficiency of an OLED with $\mathrm{Alq}_3$ thicker than the optimum 7 nm can be similarly explained. Any additional voltage drop across the "excess" $\mathrm{Alq}_3$ is equivalent to that across an effective PPOS layer with a thickness contributing to an equivalent resistance. Because of ~100 times lower mobility of $\mathrm{Alq}_3$ compared to that of PPOS, the equivalent PPOS thickness can be very large. This results in a large effective difference between the thickness of TPD and PPOS, hence, a large and positive $\sigma$. The balance formerly achieved with the optimum $\mathrm{Alq}_3$ thickness is now greatly disturbed, leading to a reduction in current efficiency (Fig. 3).

For Types Va and Vb OLEDs, the increase in current efficiency with increasing PPOS thickness (hence, larger deviation from balance) can be explained as follows. If the PPOS is sufficiently thick, it is still possible for excess holes to largely recombine within the ETL before reaching and being collected by the cathode. This differs from OLEDs with excess $\mathrm{Alq}_3$, for which the increase in PPOS thickness is apparent rather than real. For Types Va and Vb OLEDs, the 50-nm PPOS is not thick enough for complete recombination of excess holes; therefore, the probability of recombination is improved when the thickness of PPOS is increased (Fig. 6).

Designed to reduce the excess $\sigma$ expected at the HTL/ETL interface of a Type III OLED, a Type IV OLED was constructed with a thinner PPOS comparable in thickness to TPD. But, the efficiency of Type IV diode is quite low, as shown in Table II. The reduction in the efficiency can be attributed to radiation quenching due to proximity of the recombination zone to the cathode.

## IV. SUMMARY

A new silole derivative PPOS was synthesized and found to emit *blue* light at ~ 492 nm. OLEDs incorporating this compound as the electron-transport/emission layers were fabricated and characterized. With suitable choices of layer thicknesses, a maximum luminance of 10 460 cd/m$^2$ was measured at 18 V. A peak current efficiency of ∼8.5 cd/A (corresponding to an effective quantum efficiency of ∼4%) and a peak power efficiency of ∼3.8 Im/W were obtained. The dependence of emission efficiency on the thickness of TPD, PPOS, and $\mathrm{Alq_3}$ was investigated and explained.

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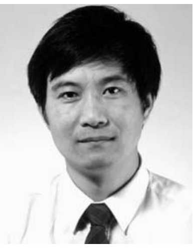

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