

## Electroluminescent Property of Tetrahydrochrysene as a Potential Emitting Layer in Organic Electroluminescent Device

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As a potential electroluminescent material, tetrahydrochrysene (THC) is prepared using the dehydrocyclization following the acyloin condensation of methyl-3-phenyl propionate as key step from *trans*-methyl cinnamate in 3 steps. THC showed emission at 428 and 456 nm after the photo- and electro-excitation, respectively. The luminance of THC doped on PVK was about 25 cd/m<sup>2</sup> at 30 voltage with 70 nm of thickness. The results suggests that a new fluorescent organic dye, THC can be used in organic electroluminescent device.

**key words:** tetrahydrochrysene, light emitting device, electroluminescence

### INTRODUCTION

Electroluminescent devices (ELD) based on organic materials are attractive due to their multi-color emission capability and low operating voltage with possible applications for the large-area flat-panel displays [1,2]. One of the problems remaining in the research and development of ELD is obtaining narrow emission bands at the three primary colors (red, green and blue), with sufficient luminescence efficiency. The EL devices consist of electrodes and hole transfer layer (HTL), emitting layer (EML), and electron transfer layer (ETL) (Figure 1). The holes from HTL and the electrons from ETL are recombined in EML to give thermal deactivation or light emission [3].

The emitting color and refinement are defined mostly by the materials in EMT [4-6]. In an attempt to develop an effective emitting material (EM), we have been interested in the application of the tetrahydrochrysene (THC, 4), a fused stilbene, because of its strong fluorescence and allowance of convenient structural manipulation. Furthermore, the emission wavelengths can be tuned *via* substitution of the electron donating or accepting groups [7]. In this report, the synthesis of tetrahydrochrysene and its spectroscopic properties are described as a potential emitting material for the electroluminescence.

The target molecule, chrysene 4 was easily prepared by the known procedure with minor modification as shown in the scheme [7]. Methyl *trans*-cinnamate was hydrogenated to give the ester 2 under the palladium catalyst in quantitative yield. The acyloin condensation of the methyl ester

2 in the presence of trimethylsilyl chloride gave silyl ether 3 in 36% yield. The PPA (polyphosphoric acid) mediated dehydrocyclization of silyl ether 3 for the chrysene 4 gave lower yield (22%) than the report in the literature. Presumably, the aromatization could be the main side reaction. The electroluminescent characteristics of THC was characterized after being doped on the poly(N-vinylcarbazole) (PVK). THC was doped on the PVK films by spin-coating method with the thickness of THC ranging from 70 to 100 nm. As the elec-

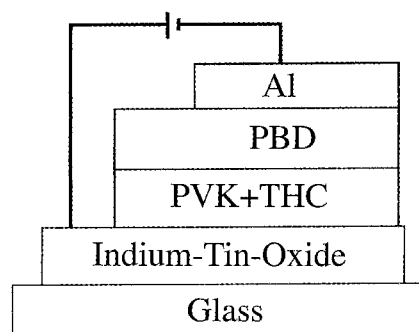
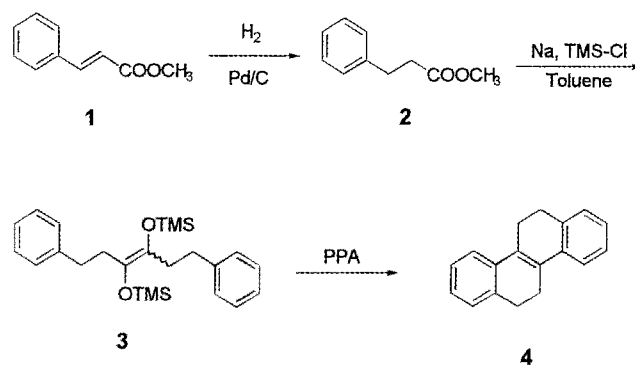


Figure 1. The schematic diagram of the electroluminescence cell.



Scheme

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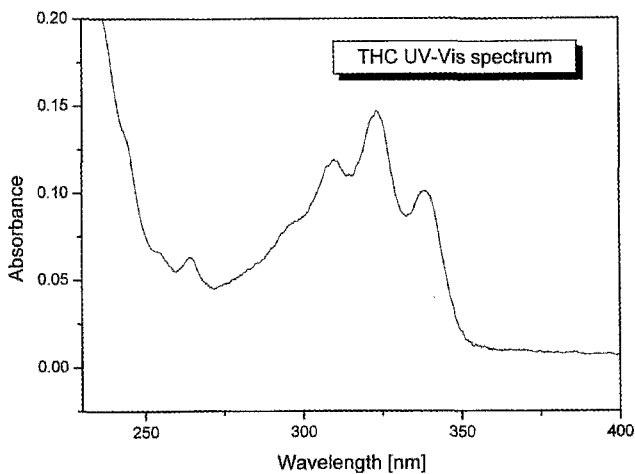
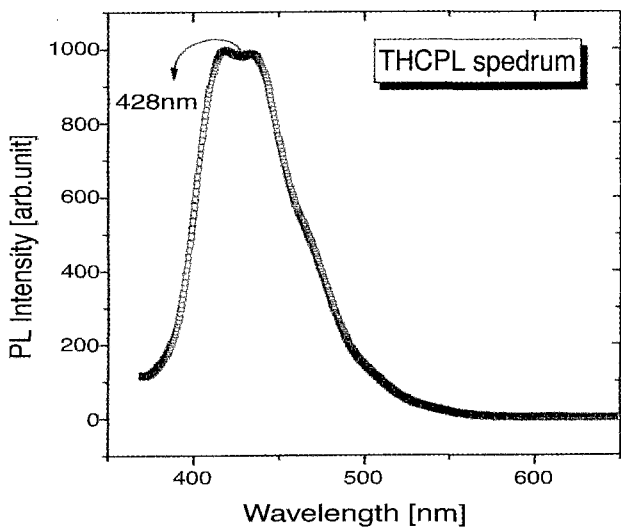
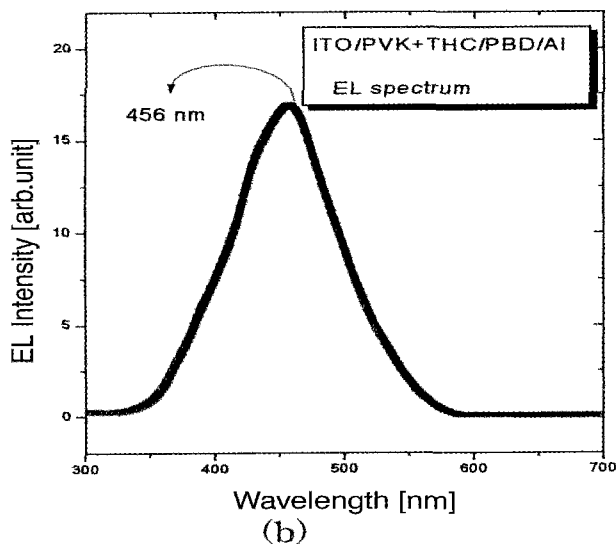


Figure 2. The UV-visible spectrum of the THC solution in MeOH.



(a)



(b)

Figure 3. (a) The PL and (b) the EL spectra of the THC films.

tron transporting layer PBD (2-(4-biphenyl)-5-(4-*t*-butylphenyl)-1,3,4-oxadiazole) also emits at 380 nm, it was vacuum-deposited and the thickness of layer was fixed to 300 nm. The current density-voltage ( $J$ - $V$ ) characteristics and luminance of organic ELD were measured with Keithley 238 electrometer and Minolta chromameter CS100, respectively. All processing steps and characterization measurement were carried out in the air at room temperature.

The organic EL cell used in this study consisted of ITO/(PVK doped THC)/PBD/Al as shown in the Figure 1. The UV-visible absorption (Figure 2) was measured in methanol solution, which imply the band gap of 3.5 eV (353 nm). The photoluminescence (PL) and the electroluminescence (EL) spectra (Figure 3) of THC were obtained from the films on the quartz plate and the device of ITO/(PVK doped with THC)/Al, respectively. The PL emission peak of THC has been observed at the wavelength of 428 nm, in the blue region. However, the peak in the EL spectrum was redshifted to 456 nm (Figure 3) which can be explained by the generation of a exciplex between the emitting material and the hole transfer material [8].

The dependence of the injection current on applied voltage under the forward bias condition was observed as in the Figure 4. When the thickness of PVK film doped with THC decreases, the turn-on voltage of the device decreased and the current density ( $J$ ) increased. In the Figure 5, the dependence of luminance on the applied voltage of the device is shown. The luminance was proportional to the injection cur-

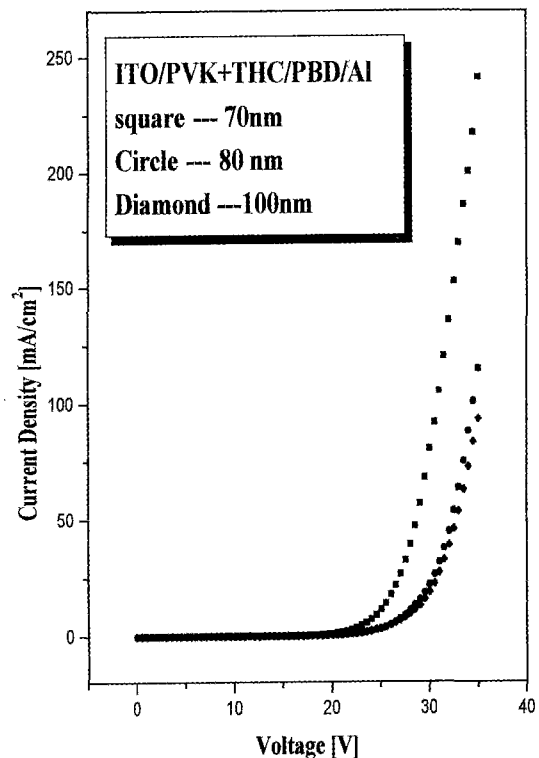


Figure 4. Current density-voltage ( $J$ - $V$ ) characteristics of the device.

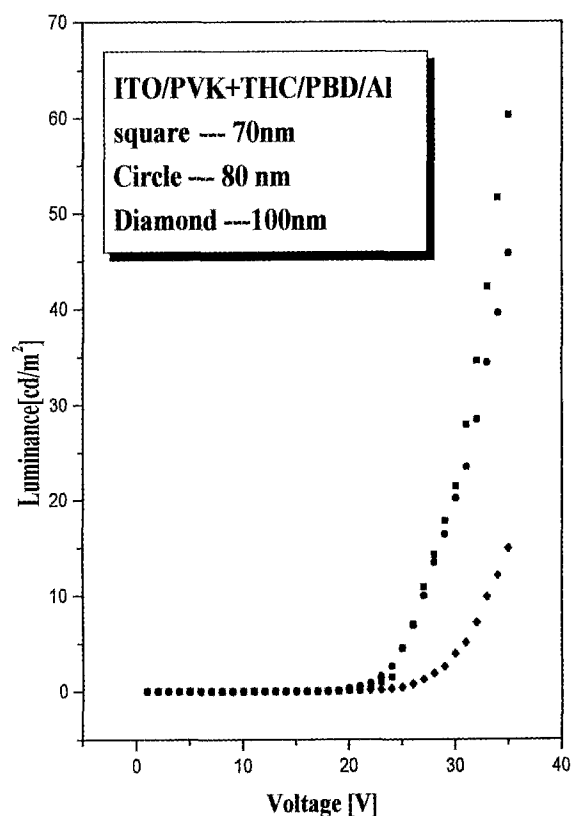


Figure 5. The relationship of luminance versus applied voltage in the THC doped device.

rent. As the thickness of THC doped on PVK film decreases, the luminance of the device increases.

In conclusion, the photoluminescent THC also represented electroluminescence. The electroluminescence of THC doped on PVK was observed at 456 nm in blue region. The luminance of THC was about 25  $\text{cd}/\text{m}^2$  at 30 voltage with 70 nm of thickness. Even though the device is not much bright, it is meaningful that a new fluorescent organic dye is applied in organic ELD (OELD). The results demonstrate possibility that THC and its derivatives can be used in OELD. The next step will be a preparation of EL device with THC substituted with electron donor-acceptor at 2,8-positions.

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9. **Methyl-3-phenyl propionate (2)**: Yield 95%,  $^1\text{H-NMR}$  (200MHz,  $\text{CDCl}_3$ )  $\delta$ 2.66 (t, 2H,  $\text{CH}_2\text{COO}$ ) 2.94 (t, 2H, benzylic H) 3.69 (s, 3H,  $\text{OCH}_3$ ) 7.20~7.31 (m, 5H, ArH) · **3,4-Bis(trimethylsilyloxy)-1,6-bis(phenyl)-3-hexene (3)**: Yield 36%,  $^1\text{H-NMR}$  (200MHz,  $\text{CDCl}_3$ )  $\delta$ 0.23~0.34 (m, 18H,  $\text{SiCH}_3$ ) 2.33 (t, 4H,  $\text{CH}_2\text{CO}$ ) 2.81 (t, 4H, benzylic H) 7.24~7.37 (m, 10H, ArH)  $^{13}\text{C-NMR}$  (200MHz,  $\text{CDCl}_3$ ) 140.6, 132.5, 127.1, 126.9, 91.6, 32.5, 23.0 · **5,6,11,12-Tetrahydrochrysene (4)**: Yield 22%; mp 98-101°C;  $^1\text{H-NMR}$  (200MHz,  $\text{CDCl}_3$ )  $\delta$ 2.69 (t, 4H, allylic protons) 2.91 (t, 4H,  $\text{ArCH}_2$ ) 7.16~7.35 (m, 8H, ArH)  $^{13}\text{C-NMR}$  (200MHz,  $\text{CDCl}_3$ ) 125.6, 125.1, 125.0, 26.8, 23.1.