

Bucky Light Bulbs: White Light Electroluminescence from a Fluorescent C₆₀ Adduct—Single Layer Organic LED

K. Hutchison,[‡] J. Gao,[‡] G. Schick,[†] Y. Rubin,^{*†} and F. Wudl^{*†}

Exotic Materials Institute
Department of Chemistry and Biochemistry
University of California
Los Angeles, California 90095-1569
Institute for Polymers and Organic Solids
Departments of Physics and Materials
University of California, Santa Barbara, California 93106

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Recently it was reported that a C₆₀ adduct, T_h-hexapyrrolidine (THP, Figure 1a), has a substantial yellow-green fluorescence.¹ The photophysical properties of THP are atypical of C₆₀ and its derivatives.² The fluorescence of C₆₀ and its monoadducts is very weak, with reported quantum yields on the order of 10⁻⁴.^{3–6} The origin of the fluorescence properties of THP may be due to a reduction of conjugation within the fullerene core leading to higher excitation energies and larger single-triplet gaps. The result is a displacement of the spectrum to the blue, a small reduction of the excited-state S₁ → T₁ intersystem crossing, an increase in the rate of singlet state radiative decay (fluorescence rate), and a decrease in the rate of triplet decay to the ground state (T₁ → S₀) by thermal deactivation.¹

Due to the photophysical properties of THP, we have utilized it as a chromophore in the fabrication of a white light organic LED. Though lightly *n*-doped as well as pristine fullerene C₆₀ was shown to exhibit electroluminescence with apparent decomposition,⁷ to the best of our knowledge, this is the *first time electroluminescence has been observed from a C₆₀ derivative*, an unprecedented result in light of the fact that C₆₀ is universally an efficient luminescence quencher.⁸

Traditionally, organic electroluminescent devices are multi-layered, with the electron-transport, hole-transport and emissive materials segregated.⁹ In contrast, we fabricated the organic LEDs by blending THP (emissive material) with a hole transport material, poly(9-vinylcarbazole) (PVK), and an electron-transport material, 2,5-bis-(4-naphthyl)-1,3,4-oxadiazole (BND), to form a single-layer device (Figure 1). This molecularly-doped polymer device configuration has been used by others.^{10–14}

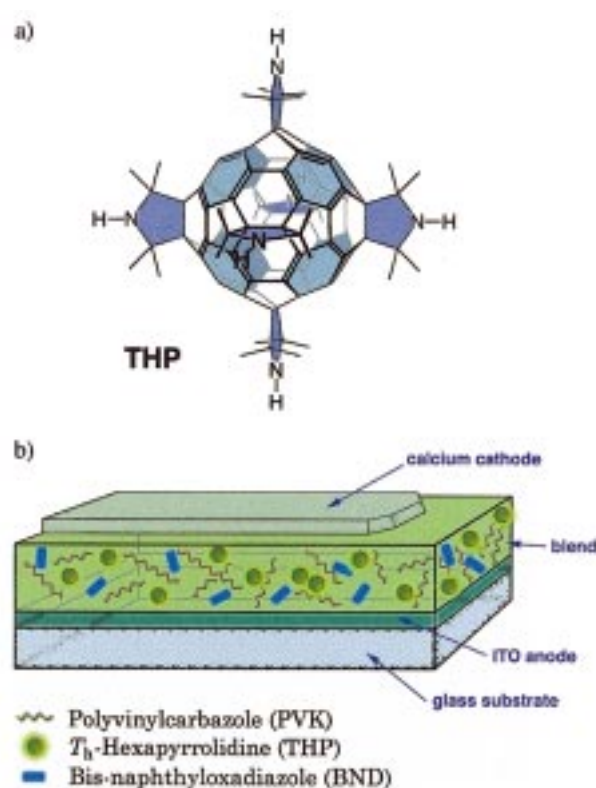


Figure 1. (a) T_h-hexapyrrolidine (THP). (b) Device configuration.

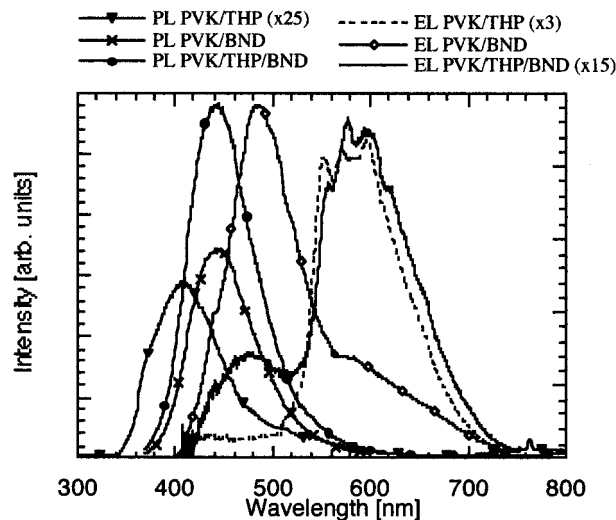


Figure 2. Photoluminescence and electroluminescence of the blends utilized in the LEDs.

Three different organic LEDs were made from blends consisting of PVK/THP/BND:100/4/28 (component ratio by weight), PVK/BND:100/28, PVK/THP:100/2. The devices made with PVK/THP/BND blends exhibited white light with turn-on voltages of ca. 13 V (Figures 2 and 3). The maximum brightness of 90 cd/m² was achieved at 28 V, with a current density of 228 mA/cm² and external electroluminescent efficiency of 0.0013%.

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[†] University of California, Los Angeles.

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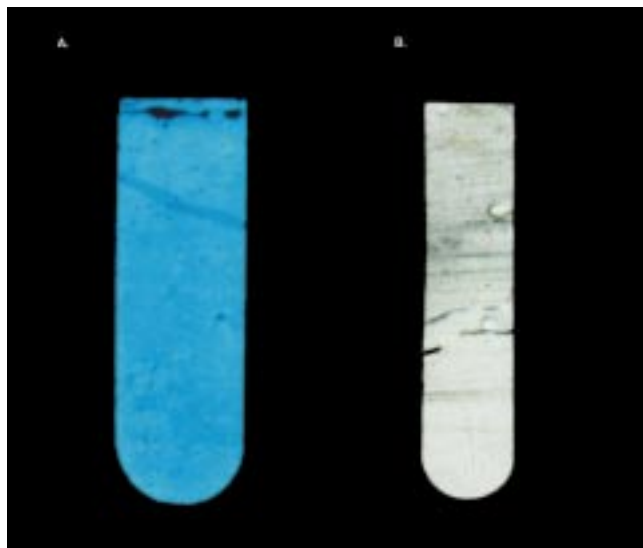


Figure 3. Electroluminescence of PVK/BND (A, blue) and PVK/THP/BND (B, white).

As a control, a device was made with PVK/BND to prove that the white light was due to THP. This device emitted blue light, which is characteristic of PVK emission¹⁰ (Figures 2 and 3). These devices were marginally less bright than the PVK/THP/BND-based devices and had external EL efficiencies of 0.008% and a maximum current density of 125 mA/cm².

A final set of devices were made by blending only PVK and THP. These devices emitted pale yellow light and exhibited brightness, efficiency (9×10^{-4} %), and current densities (38 mA/cm²) lower than the other blends (Figure 2). Hence, the electron-transport material, BND, is a necessary component for device performance. The presence of BND provides a balance of carriers and bipolar transport.¹⁰

A comparison of the photoluminescence and electroluminescence of the blends demonstrates the disparity between them

(Figure 2). All three blends emit blue light when they are optically excited. The photoluminescence from the PVK/THP blend was weaker than the other two blends. The absorption spectrum of THP (λ_{max} 300 and 360 nm)¹ and emission spectrum of PVK barely overlap, which is an indication of inefficient energy transfer from the PVK to THP and may explain the absence of photoluminescence from THP in the blends.

In contrast, only the electroluminescence from the PVK/BND blend was greenish-blue. The slight red shift of the EL of the PVK/BND blend compared to its PL is due to the formation of an exciplex between the dopants.^{12,13} The devices made with blends of PVK and THP exhibit electroluminescence approximately twice as broad as the fluorescence spectrum of THP in solution.¹ When all three components are included in the blend, the EL spans the visible spectrum from 400 to 750 nm. We propose a "carrier trapping mechanism," where the holes (carried by PVK) and electrons (carried by BND) are trapped in the HOMO and LUMO of THP, respectively, their recombination induces THP to emit light.^{10,11}

In summary we have fabricated a single layer, white light, organic LED by blending the fluorescent *T_h*-hexapyrrolidine C₆₀ adduct (THP) with PVK and BND. This is the first known example of electroluminescence from a C₆₀ derivative. The white light is a result of fortuitous overlap of emission from PVK/BND (λ_{max} 480 nm) and recombination of electrons and holes on the fullerene derivative (λ_{max} 600 nm). Further development of luminescent fullerene derivatives with higher quantum yields is being carried out to obtain higher efficiency LEDs.¹⁵

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(15) To improve on the film-forming properties of THP and compatibility with PVK, we are modifying the alkyl groups of the pyrrolidines on their two possible modifiable sites; at the ring (currently methyl groups) and nitrogen (currently hydrogen). These should prevent phase segregation and self-quenching of the luminescence.