

Higher efficiency conjugated polymer-based LEDs by control of polymer film morphology and interchain interactions

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Abstract

Aggregation of conjugated polymer chains in optoelectronic devices promotes good carrier transport but quenches luminescence, leading to a fundamental trade-off in optimizing film morphology for applications. Changing the solution from which a conjugated polymer film is cast can control the degree of aggregation in the film. In this paper, we show that the efficiency of light-emitting diodes (LEDs) based on poly(2-methoxy-5-(2'-ethylhexyloxy)-1,4-phenylenevinylene) (MEH-PPV) can be improved by constructing tri-layer devices, which use a central MEH-PPV layer with reduced interchain interactions between layers of more highly aggregated MEH-PPV near the electrodes.

Keywords: Conjugated polymer; Light emitting diode; Aggregates/Interchain interactions

Introduction

Conjugated polymers have been studied extensively due to their potential in photonic devices such as light-emitting diodes, photodiodes, and photovoltaics. LEDs have been improved greatly in recent years with lifetimes, efficiencies, and luminances comparable to commercial devices based on inorganics. Most of this improvement in polymer LED performance has resulted from changes in device architecture, such as adding electron and/or hole transporting layers, encapsulation, etc. In this work, we show that polymer device performance can be improved by controlling the film morphology without changing the device architecture. We can control the polymer film morphology quite simply by changing the solvent and polymer concentration of the solution from which the films are cast or by annealing the polymer films [1,2].

In previous work, we found that aggregation of MEH-PPV chains is promoted in solvents such as chlorobenzene (CB), where the polymer chain has a more open conformation, and is restricted in solvents such as tetrahydrofuran (THF), where the chains tend to form a tighter coil [3]. Evidence from a variety of experiments suggested that the degree of aggregation in solution survives the casting process and carries into the films [1]. As a result, MEH-PPV LEDs based on films cast from THF, which have a lower degree of interchain interactions, have lower injection current, but higher quantum efficiency than corresponding devices based on films with a larger number of aggregates cast from CB [4]. This illustrates a basic trade-off when trying to optimize the performance of polymer-based devices: the same interchain interactions

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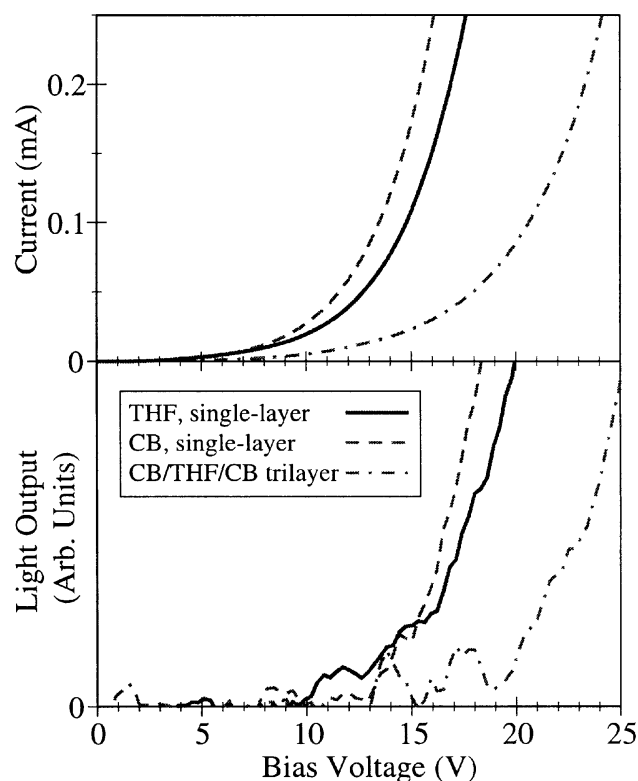


Fig. 1. Current-voltage (a) and brightness-voltage (b) curves for MEH-PPV devices with identical overall thickness.

that promote charge transport are detrimental to luminescence efficiency [3].

Here, we show that by understanding the polymer film morphology, we can fabricate LEDs with improved performance. We will investigate the performance of

"heterostructure" LEDs composed of a layer of MEH-PPV cast from THF sandwiched between layers cast from CB ("tri-layer" device). The idea is that highly aggregated CB-cast chains near the electrodes will allow good current transport while the low-mobility THF-cast central layer will trap carriers and provide for recombination with a higher luminescence quantum yield. We find that the trilayer devices have higher efficiency than comparable single-layer devices cast from either CB or THF solution.

Experimental

For single-layer devices, MEH-PPV in either CB or THF is spin-cast onto indium-tin-oxide (ITO)-coated glass substrates. For tri-layer devices, MEH-PPV in CB was spin-cast onto the ITO substrate, the resultant film was allowed to dry, then another solution of MEH-PPV in THF was spin-cast over the CB-cast layer. Finally, a third layer of MEH-PPV in CB was spin-cast on top of other two layers. The device fabrication details are described elsewhere [1,4]; spin speeds were chosen to ensure that all the devices in this study had the same total thickness of MEH-PPV.

Discussion

The importance of polymer film morphology in device performance is demonstrated in Figure 1, which shows the current-voltage (upper panel) and light output-voltage (lower panel) curves for single-layer device cast from CB or THF and for CB/THF/CB trilayer devices. Single-layer devices fabricated from CB-cast MEH-PPV films show higher current and lower EL efficiency than devices based on THF-cast films. This is because the poorer interchain contact in the THF-cast films leads to lowered carrier mobility [1]. Even though there are highly aggregated MEH-PPV chains cast from CB near the electrodes, the tri-layer devices have a poorer working current than either of the 2 single-layer devices because of poor charge transport across the internal interfaces as well as the generally poor transport through the central THF layer.

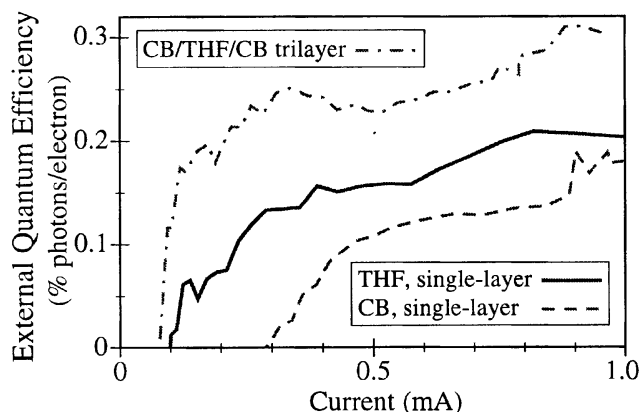


Fig. 2. External EL quantum efficiency for MEH-PPV devices with the same overall total thickness of MEH-PPV.

Despite the much lower current, the EL efficiency of the tri-layer devices is significantly higher than the single-layer devices, as shown in Fig. 2. The higher efficiency of the tri-layer device results from a combination of factors. First, the trilayer devices have higher EL efficiency than the single-layer devices because charge injection in the tri-layer devices is more balanced due to the larger number of aggregates present in the CB-cast layers near the electrodes. The second factor is that recombination of charges in the tri-layer devices is largely confined to the central THF layer where the lower degree of aggregation leads to a higher overall luminescence quantum yield. Finally, because carriers tend to recombine in the low-mobility THF layer in the center of the tri-layer device, quenching interaction with the electrodes are reduced relative to either the CB- or THF-cast single-layer devices. Therefore, the efficiency of the tri-layer device is higher than for either type of single-layer devices at the same overall thickness of the MEH-PPV active layer.

Conclusions

We have shown that controlling morphology by varying the solvent from which the polymer films are cast and combining several layers of MEH-PPV in different solvents can improve the device performance without changing the device architecture. Clearly, this leaves room to improve all polymer-based devices, independent of architecture, by optimizing the chain morphology for the desired application. We are presently working to extend these ideas to a new class of phenylene vinylene polymers with amino side-groups [5]. By protonating or deprotonating the amino groups, we can change this material from a neutral polymer to a polyelectrolyte, allowing exquisite control over the solution conformation [6]. If this conformational control carries into the film, we will then have a new way to improve the efficiency of conjugated polymer-based devices. Overall, by combining morphology control of the active polymer layer with changes in the device architecture, we should be able to further optimize the performance of polymer-based LEDs for use in commercial products in the near future.

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