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Abstract. Polymer light-emitting devices based on the *in situ* formation of a *p-i-n* junction in a polymer blend have been fabricated by roll lamination. Single-walled carbon nanotubes are used as both the anode and cathode for a fabrication process that does not employ vacuum. The metal-free, thin-film devices exhibit a low turn-on voltage, modestly high efficiency, and brightness. The devices are fairly transparent and exhibit high mechanical flexibility. No failure was observed after repeated bending to a 2.5 mm radius and unbending for 50 cycles. Tandem light-emitting devices were prepared, by stacking a blue device on top of a yellow device, for the convenient generation of various mixed colors. © 2011 Society of Phot-Optical Instrumentation Engineers. [DOI: [10.1117/1.3528271](https://doi.org/10.1117/1.3528271)]

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1 Introduction

Polymer light-emitting diodes (PLEDs) using luminescent conjugated polymers have been envisioned for flexible, low-cost flat-panel display, signage, and lighting applications since their invention two decades ago.^{1,2} Compared to their inorganic counterparts, conjugated polymers can be solution processed and are mechanically compliant. As such, PLEDs can potentially be fabricated into large-area devices using a low-cost process, such as ink-jet printing, screen printing, or roll lamination onto flexible substrates such as polyethylene terephthalate (PET) and metal foils.³ In the past decade, the focus on PLEDs has been on developing flat-panel displays with contrast and efficiency higher than those of liquid-crystal displays. More recently, PLEDs have also become attractive for solid state lighting applications. On the other hand, the envisioned flexibility and low cost have not been fully achieved. This is largely due to the following challenges: (i) The conventional transparent electrode, indium-doped tin oxide (ITO), is brittle. ITO cracks under small tensile strain,^{4,5} and as such, PLEDs fabricated on ITO-coated flexible substrates have limited flexibility. (ii) High-efficiency PLEDs require multilayered device architectures to balance the injection of electrons and holes. The delicate control of the thickness of each layer leads to high fabrication cost. (iii) Reactive metals or salts are commonly used as the electron injection materials in PLEDs. These materials are sensitive to oxygen or moisture and are not very compatible with a solution process. Hermetic encapsulation is generally required for the PLEDs due to the reactivity of these materials and the interfaces. (iv) To obtain a low driving voltage and high power efficiency, the emissive polymer layer is limited to 100-nm thicknesses or thinner. Direct printing of consecutive layers tends to impair the emission layer: The solvents contained in the inks may swell and contaminate the emissive

polymer during printing. Lamination of 100-nm or thinner layers at an elevated pressure and temperature may induce stress concentrations and thickness nonuniformities. Because of these challenges, thermal evaporation in high vacuum has been the preferred method to deposit the cathode.

During the same time, printing and lamination processes have continuously been pursued to drive down the fabrication cost of PLEDs.^{6–10} Huang et al.¹¹ and Wu et al.¹² developed printable electron injection layers using water or alcohol-soluble conjugated polymers. Zeng et al.⁷ fabricated PLEDs by printing three different polymer layers and a silver paste cathode. Lee et al.⁸ reported soft contact lamination using elastomeric polymers to laminate PLEDs in a less intrusive manner.

To improve the flexibility of PLEDs, a variety of transparent conductors have been investigated as compliant anodes to replace the ITO, including solution-processable conducting polymers such as polyaniline and poly(3,4-ethylenedioxythiophene) (PEDOT),^{13–15} metal oxides other than ITO,¹⁶ graphenes,^{17,18} and thin films of carbon nanotubes.^{19,20} On the other hand, the choice for the cathode is limited. Metallic thin films are commonly used; however, they are prone to cracking or delamination when the devices are subjected to repeated bending cycles.²¹ There are few reports on compliant cathodes. Gadisa et al.²² developed a vapor-phase polymerized PEDOT and studied it as the transparent cathode for polymer solar cells. Its relatively high work function makes PEDOT a poor cathode material to inject electrons into the polymer semiconductors.

Single-walled carbon nanotube (SWNT) thin-film electrodes deposited onto various substrates via solution processes have been developed with conductivity and transmittance comparable to those of ITO electrodes.^{23–25} We have shown that ultrathin SWNT electrodes exhibit superior stretchability: The electrodes remain conductive even after >700% elongation.²⁶ These electrodes have been used to drive dielectric elastomers to electrically actuated strains up to 300% in area.²⁷ In addition, actuated strains of 220% have been achieved in carbon nanotube aerogels where they are the sole component.²⁸ The high compliancy of the SWNT electrodes arises from network formation of the high-aspect-ratio nanotubes. The electrodes consist of randomly oriented and highly interwoven carbon nanotubes. SWNT coatings have been studied as the electrodes for various electronic devices, including inorganic light-emitting diodes (LEDs),²⁹ small molecule organic LEDs,^{30,31} PLEDs,³² organic solar cells,^{33,34} and super capacitors.³⁵

Like ITO, SWNT has not been a good choice for the cathode in PLEDs: There is a large energy offset (>2 eV) between the Fermi level of SWNT and the Lowest Unoccupied Molecular Orbital (LUMO) of typical conjugated polymers.^{36–39} To overcome this problem, polymer light-emitting electrochemical cells (PLECs) have been proven to be quite useful.^{40–43} PLECs consisted of a light-emitting polymer, a salt, and an ionic conductor admixed to form the active polymer layer. A *p-i-n* junction is formed *in situ* in the emissive polymer layer when a sufficient bias is applied on the PLECs: The light-emitting semiconductive polymer becomes p-doped along the anode side and n-doped close to the cathode side, with an intrinsic undoped polymer layer in between from which high-efficiency light emission can be induced. In the PLEC devices, the n-doped polymer serves as the interfacial layer to enhance the injection of electrons into the undoped polymers. The cathode therefore can use a metal of high work function. Both the p- and n-doped regions are conductive; the highly resistive i-region of *p-i-n* junction is thinner than the thickness of the polymer layer. Thus, the PLECs can use a relatively thick polymer layer sandwiched between the charge-injection electrodes. The ability to use high work function cathode and thick polymer layer significantly opens up the fabrication of PLECs to various low-cost techniques, such as printing and lamination. For example, PLECs have been fabricated with the light-emitting polymer and an air-stable silver paste cathode both deposited by screen printing.⁴⁴

Thus, the combination of SWNT electrodes and lamination would eventually produce highly flexible, semitransparent, and cost-effective PLECs.⁴⁵ We report the fabrication of PLECs by roll lamination. The devices have a simple sandwich structure of polyester/SWNT/emissive

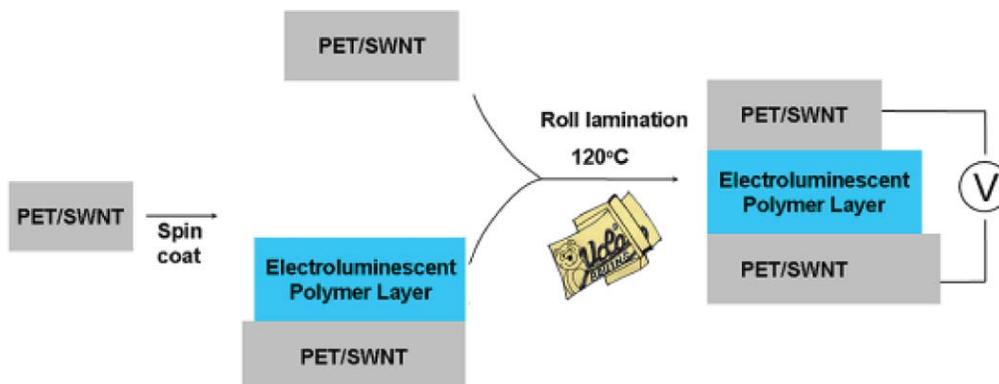


Fig. 1 Schematic illustration of the lamination process to fabricate a PLEC device.

polymer/SWNT/polyester. The emissive polymer layer contains polyfluorene-based copolymers respectively emitting red, yellow, and blue light, as well as an ionic conductor and a salt for junction formation. The devices are semitransparent, with $>70\%$ transmittance outside the wavelength range of the polymer's π - π^* bandgap transition. The devices are also highly flexible. They can be repeatedly bent to a 2.5-mm radius without causing permanent damage to device performance. Various mixed colors were obtained conveniently by stacking the semitransparent devices.

2 Experimental

The fabrication process of the polymer light-emitting devices is illustrated in Fig. 1. An ultrathin coating of SWNT was coated on polyester (PET) sheets using a modified procedure described in the literature.⁴⁶ In brief, raw SWNTs were dispersed in water in aid with sodium dodecylsulfonate and high-power ultrasonication. The dispersion was then filtered onto porous alumina filter paper. The SWNT film formed on the filter paper was transferred onto PET substrate (100- μm thickness). Pressure and heating were employed to facilitate the transfer. The sheet resistance was 500 Ohm/sq. The average transmission in the wavelength range between 400 and 1100 nm was 85%. The resulting SWNT/PET substrates were ultrasonically cleaned for 2 min each with detergent water, deionized water, and isopropanol, and then dried in vacuum for 2 h before use.

The emissive polymer layer in the PLECs contained a luminescent conjugated polymer, an ionically conductive component, and a salt. Three luminescent polymers were used, PF-B, PF-G, and PF-R, for blue, yellow, and red light emission, respectively. Their chemical structures are shown in Fig. 2. An ethoxylated trimethylolpropane triacrylate (ETT-15) was used as the ionically conductive component. The salt was lithium trifluoromethane sulfonate (LiTf).

Light-emitting polymers, ETT-15 and LiTf, at the weight ratio of 20:10:1, were codissolved in tetrahydrofuran. The concentration of the polymer in the solutions was kept at 50 mg/mL. The mixture solution was stirred at room temperature for 2 h and then spin-cast at 1500 rpm on SWNT/PET substrates. Thickness of the emissive polymer layers was measured on a Dektak profilometer. The films were vacuumed for 2 h and subsequently roll-laminated with another SWNT/PET sheet to form the top electrode. The lamination temperature was 120°C. All the solution preparation, film casting, and lamination were carried out in a nitrogen-filled dry box with oxygen and moisture level controlled at <0.1 ppm.

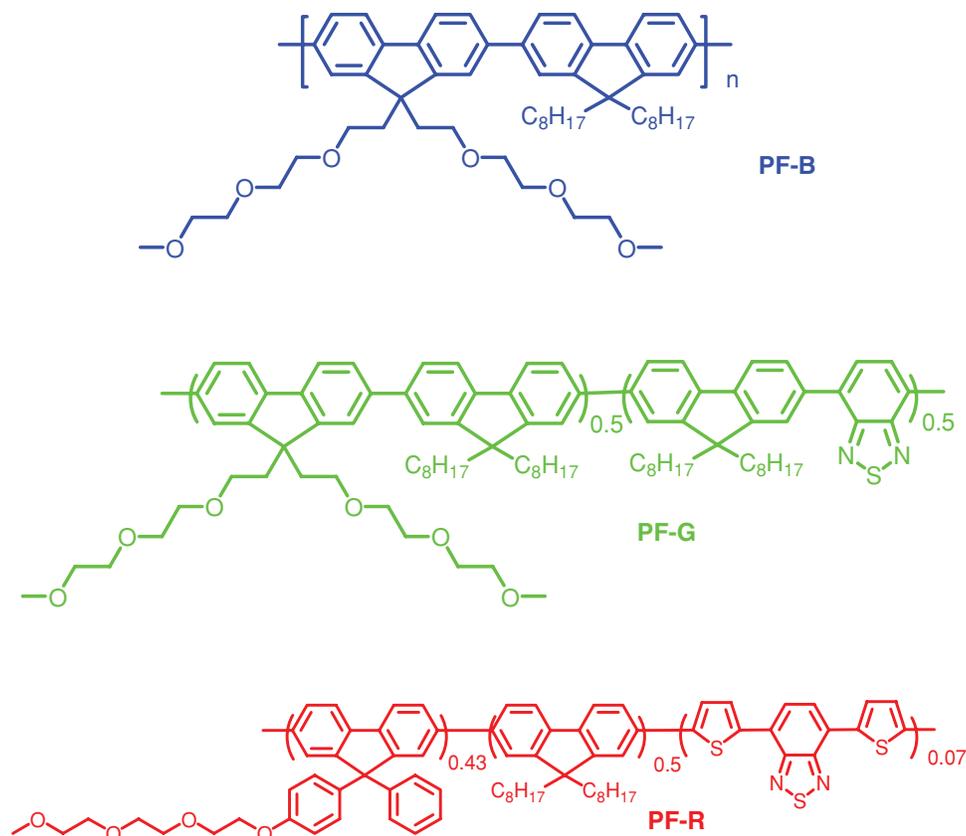


Fig. 2 Chemical structures of the blue (PF-B), yellow (PF-G), and red (PF-R) light-emitting polymers used for laminated polymer light-emitting devices.

3 Thickness of the Active Polymer Layer on Device Fabrication Yield

Ultrathin SWNT electrodes are porous and have been reported with a high surface roughness of >15 nm.^{47,48} In our observation using an atomic force microscope (AFM) to scan a relatively large area ($10 \times 10 \mu\text{m}$) of SWNT on PET (Fig. 3), a number of long tubes erected at least 100 nm above the coating surface were observed that could poke through the emissive polymer layer. This high roughness is detrimental to the performance of conventional PLEDs. During roll lamination of the PLEC devices, high pressure and high temperature (above the glass transition of the emissive polymers) were applied to promote electrical contact and adhesion between the polymer and SWNT electrodes. It was found to be important to employ a relatively thick emissive polymer layer to prevent the potential electrical shorts caused by the poking of long SWNTs deep into the polymer layer. The effect of the thickness of the polymer layer on the fabrication yield of the laminated PLECs is summarized in Table 1. At a polymer layer thickness of ~ 1200 nm, the lamination had a 100% yield. This thickness is about 10X larger than that used in a conventional PLED.^{49,50} The *in situ* formation of a *p-i-n* junction in the polymer layer is necessary to reduce the driving voltage to be comparable to a conventional PLED.

4 Device Transparency

The laminated polymer light-emitting devices with SWNT as both cathode and anode are quite transparent, as shown in Fig. 4. The devices were placed on a sheet printed with “UCLA” patterns

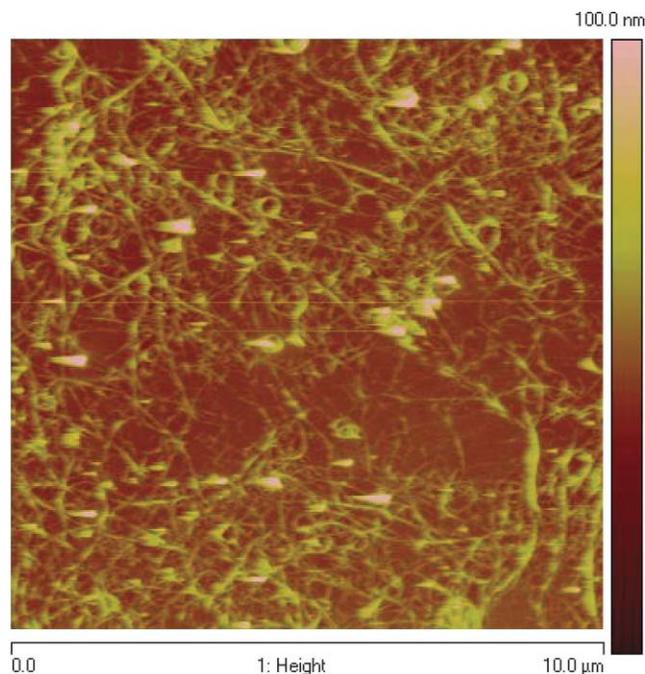


Fig. 3 AFM height image of the SWNT on PET.

to illustrate the transparency. The colors of the yellow and red PLECs are from the absorption of conjugated polymers. The blue PLEC shows the absorption of the SWNT electrodes, and the blue polymer all being negligible in the visible region. The optical transmission spectra of the devices in the wavelength range of 1100–350 nm are shown in Fig. 5. The SWNT/PET bilayer together has a total transmission of 73% averaged from 400 to 1100 nm. The PLECs all exhibit >70% transmission in the wavelength range outside the range where the conjugated polymers absorb. The onsets of abrupt transmission reduction are at 470, 540, and 634 nm for the blue, yellow, and red PLECs, respectively. Figure 6 shows a working blue emission device. The device is in front of a mirror such that the light emitted from back surface can also be seen.

5 Current Change with Bending

The laminated PLECs were tested in a nitrogen-filled drybox with oxygen and moisture level of <0.1 ppm. The current–voltage and light intensity–voltage responses were measured with a Keithley 2400 source meter and a calibrated silicon photodetector by sweeping the applied voltage from 0 to 10 V at 100-mV increments. Figure 7 shows the current responses of the device. The device was tested under different bending curvatures. A slight current decrease was observed, as shown in Fig. 7, when the device was bent. The current at 10 V dropped by 5 and 10% when the device was flexed into a 6- and 2.5-mm radius, respectively. The current drop may be caused by the resistance increase of the SWNT electrodes after bending. A 2.5-mm bending radius corresponds to a strain of 2% in the SWNT films, which could cause an 8% increase of sheet resistance of the SWNT electrodes.⁴⁸

Table 1 Fabrication yield of the laminated PLECs as a function of the thickness of the emissive polymer layer.

Thickness (nm)	500	800	1000	1200
Yield (%)	0	20	80	100



Fig. 4 Photographs of (from left to right) blue, green, and red PLECs, and two layers of SWNT/PET substrates laminated without any emissive polymer layer.

6 Emission Characteristic of the Flat and Bent Devices

For a flat blue emission device, the turn-on voltage was 3.8 V (1 cd/m² light-emission intensity). The emission intensity reached 1400 cd/m² at 10 V. The maximum efficiency was 2.2 cd/A at 480 cd/m². This efficiency value is on par with those reported for green-emission small molecular LEDs (Ref. 30) or PLEDs (Ref. 32) using a SWNT anode and evaporated metallic cathode. The flexibility of this device was demonstrated in Fig. 8, in which the device was deformed to conform onto a 2.5-mm-radius glass tube. No significant damage was observed during bending. The bent device remained uniform and exhibited similar brightness as the flat device at the same driving voltage. Accurate measurement of the emission intensity of the bent device could not be performed because the setup was not calibrated for curved light sources.

Yellow and red PLECs were also fabricated with laminated SWNT cathode and anode. These devices had a similar sandwich structure as the blue emission device except that PF-G and PF-R were employed as the light-emitting polymers. These devices had similar flexibility and could also be bent to a radius of curvature of 2.5 mm without failure. No significant changes in light-emission intensity and uniformity were observed after the devices were bent to a 2.5-mm radius (Fig. 9). The bent devices were wrapped with a double-sided tape to keep them from bending back to a flat shape.

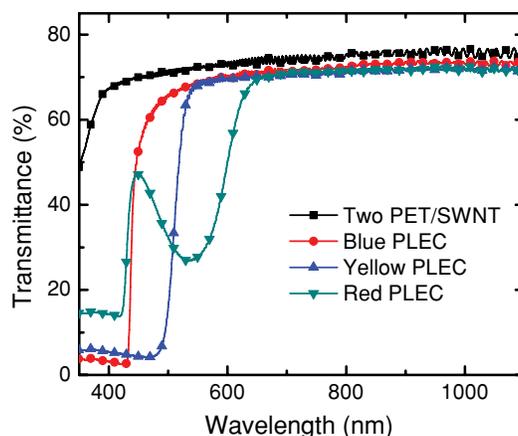


Fig. 5 Optical transmittance spectra for blue, green, and red PLECs, and two layers of SWNT/PET substrates laminated without any emissive polymer layer.

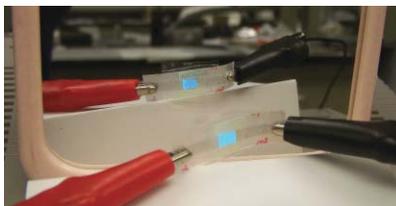


Fig. 6 Photograph of a working blue PLEC in front of a mirror showing emission from both surfaces.

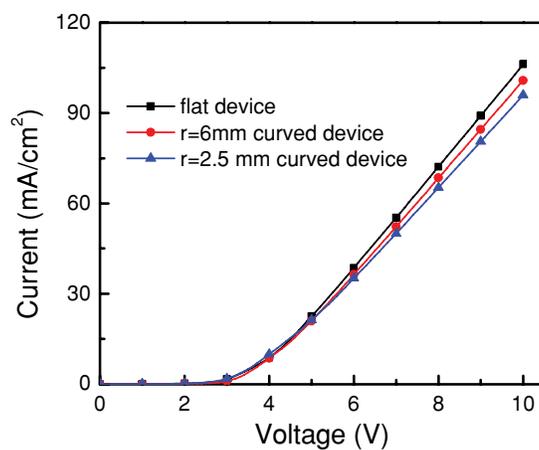


Fig. 7 $I-V$ characteristic of a blue emissive device under different curvatures.

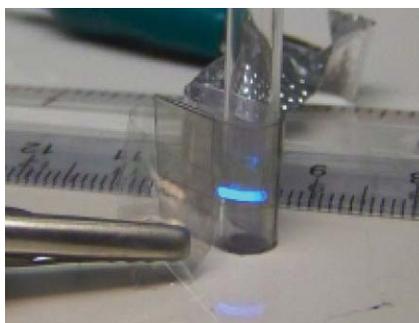


Fig. 8 A blue PLEC bent (conformed) around a 2.5-mm-radius glass tube.

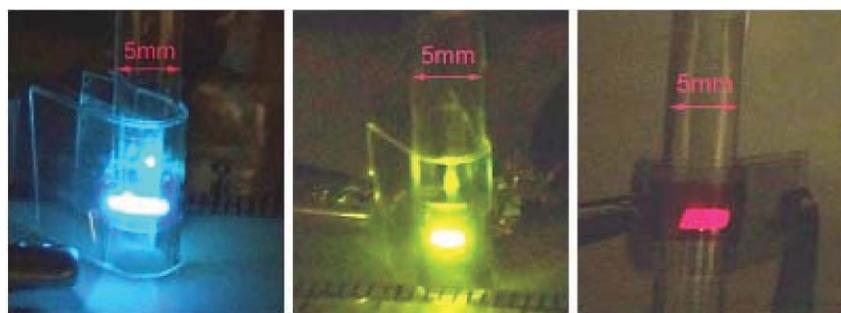


Fig. 9 Photographs of bent blue, yellow, and red PLECs operated at 10 V. The devices are bent around a 2.5-mm-radius glass tube.

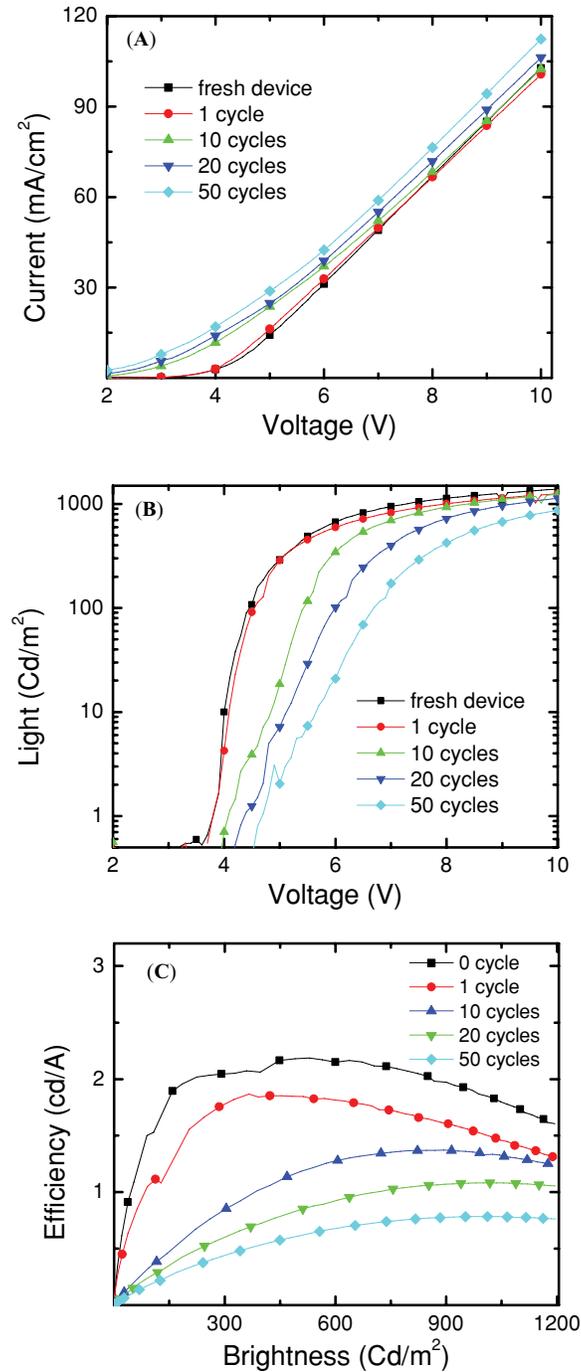


Fig. 10 Responses of (a) current–voltage, (b) emission intensity–voltage, and (c) efficiency brightness of a blue PLEC in a flat shape after specified cycles of bending to 2.5-mm radius and unbending.

7 Bending Cycle Test

Bending cycle lifetime of the laminated devices was investigated using the blue PLECs. The devices were bent back and forth repeatedly between a flat shape and a 2.5-mm-radius curvature for up to 50 cycles. Figure 10 shows the current and light emission responses of a flat device after specified number of bending-unbending cycles. The turn-on voltage changes very

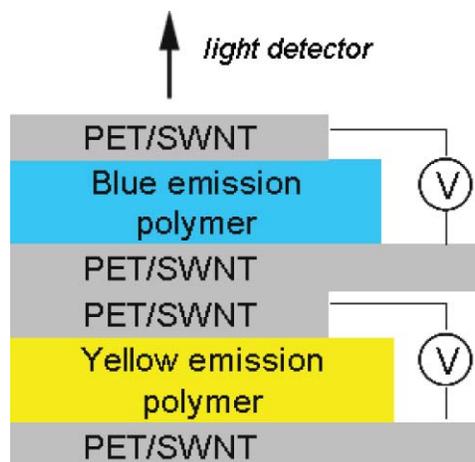


Fig. 11 Schematic of a stacking configuration consisting of one blue emission device on top and a yellow emission device on the bottom; the light emission was detected from the blue device side.

little after one bending cycle. It gradually increases with number of cycles, from 3.8 to 4.0 V in 10 cycles, 4.4 V in 20 cycles, and to 4.7 V in 50 cycles. The light intensity at 10 V was 1400 cd/m² for the fresh device and decreased to 1260 cd/m² after one cycle of bending, 1240 cd/m² in 10 cycles, 1140 cd/m² in 20 cycles, and 870 cd/m² in 50 cycles.

The devices after 50 cycles of bending remained uniform, with blue light emitted uniformly over the entire emissive area. The current density shows a trend of rising with the number of bending cycles [Fig. 10(A)]. The increase is more significant at lower bias voltage.

The luminous efficiency decreases with bending cycles as shown in Fig. 10(C). The maximum efficiency in the fresh blue-emission device is 2.2 cd/A, and decreases to 1.9 cd/A after one bending cycle, 1.4 cd/A after 10 cycles, 1.1 cd/A after 20 cycles, and 0.8 cd/A after 50 cycles.

The flexibility of the laminated light-emitting devices is much higher than those achieved in PLEDs using ITO or a conducting polymer as one or both of the opposite electrodes. The maximum bending curvature was limited to a 7-mm radius when ITO was used as the electrodes.^{51,52} Conducting polymers are more compliant than ITO. With a conducting polymer employed as the anode and a metallic layer as the cathode, cracks were found to initiate along the cathode side when the devices were bent to a 3-mm radius.⁵³ Ultrathin SWNT coatings not only offer higher mechanical compliancy but also are more conductive than the conducting polymers studied thus far for PLED electrodes. The widely used transparent conducting polymers, poly-3,4-ethylenedioxythiophene-polystyrenesulfonate (PEDOT:PSS), has a conductivity <100 S/cm. Treatment with high boiling point additives can raise the conductivity up to 500 S/cm (Ref. 54), which is still 3X lower than that of SWNT electrodes. SWNT electrodes have also shown excellent compliancy in repeated bending. The sheet resistance of SWNT electrodes was found to change little during bending between a flat shape and a 12.5-mm-radius curvature for over 10,000 cycles.⁴⁷

8 Tandem PLECs

Transparent light-emitting devices are not only useful for see-through displays, but also for full color displays by vertically stacking red, green, and blue devices into tandem configurations.^{55–57} The semitransparent blue and yellow PLECs were stacked, and the light-emission blue device side was detected as illustrated in Fig. 11. Varying the relative intensity of the two stacked devices could generate various mixed colors as shown in Fig. 12. The emission

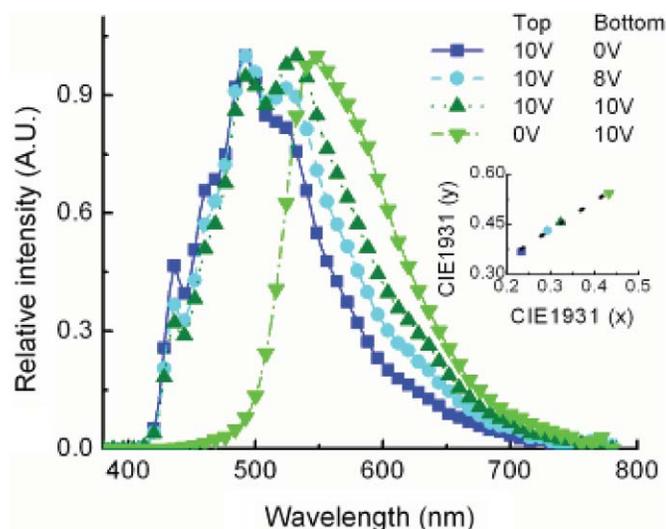


Fig. 12 Emission spectra of the tandem device shown in Fig. 11, with the two PLECs independently operated at the specified voltages.

color is sky blue with CIE coordinates (0.22, 0.37) when only the blue device was operated at 10 V. The emission color is yellow with CIE coordinates (0.43, 0.54) when only the yellow device was operated at 10 V. The color is tunable between the two colors by adjusting the applied voltage on each device. For example, when 10 V was applied on both devices, the emission color is greenish blue with CIE coordinates (0.32, 0.45).

9 Conclusion

SWNT electrodes have been successfully used as both the cathode and anode for polymer light-emitting devices. The devices had a simple layered structure and were fabricated by a roll-lamination process. The *in situ* formation of a light-emitting *p-i-n* junction allows the use a relatively thick polymer layer that remains intact by the high surface roughness of the SWNT electrodes and the heating and pressing during the lamination. The blue devices showed good electroluminescent performance: low turn-on voltage (3.8 V), high efficiency (2.2 cd/A at 480 cd/m²), and high brightness (1400 cd/m² at 10 V). Yellow and red light-emitting devices have also been fabricated. All these devices are highly flexible and can be bent to a 2.5-mm radius, repeatedly, without causing failure. After 50 cycles of bending to a 2.5-mm radius and unbending, the blue devices had a turn-on voltage of 4.7 V, brightness of 870 cd/m² at 10V, and maximum efficiency of 0.8 cd/A at 980 cd/m². All devices were highly transparent with transmittance of >70% in the wavelength range where the emissive polymers do not absorb. Stacking of the semitransparent devices was employed to conveniently generate various mixed emission colors.

Acknowledgment

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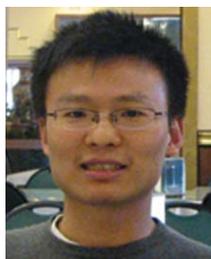
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