



The effects of compressive stress on the performance of organic light-emitting diodes



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ABSTRACT

We investigated the effects of compressive stress on the performance of polymer light-emitting diodes (PLEDs). Vertical contraction of the PLEDs under compressive stress was induced by loading the PLEDs during operation. The current density–luminescence–voltage (J – L – V) characteristics of the PLEDs under stress were measured and compared with those of unloaded PLEDs. To explain the changes in the PLED performance due to the stress, the chemistry and electronic energy band structures of each stressed layer constituting the PLEDs were analyzed via X-ray photoelectron spectroscopy (XPS) and ultraviolet photoelectron spectroscopy (UPS), respectively. According to the analysis, an irreversible deterioration in the fluorescent characteristics of the active layer and an increased electron energy barrier between the layers of the PLEDs were induced by the applied stress, which resulted in the significant degradation of the J – L – V characteristics.

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1. Introduction

In the past few decades, the applications of organic light-emitting diodes (OLEDs) have been extended from general lighting to light sources for displays and sensors [1–5] because of the superior characteristics of OLEDs, such as their wide viewing angle, vivid colors, and low power consumption, compared with other solid-state light sources [6]. As fabrication processes and the materials synthesis technologies rapidly have progressed, the lifetime and reliability of OLEDs have been dramatically improved [7–9].

Because wearable and flexible electronics have been a focus of research for various applications, flexible OLEDs (f-OLEDs) have also gained considerable attention [10–12]. Successful fabrication of stretchable and bendable OLEDs on polymer substrates has been demonstrated by the present authors and others [1,13,14]. In previous work, f-OLEDs have been demonstrated to operate well under tensile and compressive stresses; however, the luminescence (L) and efficiency (η) were permanently degraded compared with f-OLEDs operated in flat configurations [14].

Compared with the rapid progress in device fabrication technology based on f-OLEDs, little is known about the mechanism for the performance degradation of f-OLEDs under applied stress. Numerous works have investigated the effects of mechanical stress on the emissive layer of small molecular OLEDs (SMOLEDs)

[15–19]. In these studies, it was revealed that there are significant changes in the photoluminescent and electroluminescent properties of the emissive layers due to the enhancement of π – π interactions and the decrease in the band gap induced by the stresses [15–19]. However, most of these results were obtained for the luminescent materials of SMOLEDs and, most importantly, without integration of the materials into SMOLEDs.

In this work, we investigated the effect of stress on the performance of polymer light-emitting diodes (PLEDs) and identified possible mechanisms for performance degradation in OLEDs. When operated under inward bending, compressive stress is induced in the PLEDs. To measure the current density–luminescence–voltage (J – L – V) characteristics of PLEDs under compression, loading was imposed on the PLEDs when they were turned on, and the stress was varied from 0 to 100 kPa. When there was an irreversible change in the J – L – V characteristics caused by the stress, the induced changes in the chemical and electronic properties of the materials and interfaces that constituted the PLEDs were analyzed using X-ray photoelectron spectroscopy (XPS) and ultraviolet photoelectron spectroscopy (UPS), respectively. Based on the results of the analysis, the critical factors for performance degradation were determined.

2. Experimental procedures

Indium tin oxide (ITO) ($\text{In}_2\text{O}_3\text{:Sn}_2\text{O}_3 = 90\text{:}10$ in wt%, Advanced Nano Product Corp., Daejeon, Korea), the anode material, was deposited by direct-current (DC) magnetron sputtering onto a

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polyimide (PI) and a glass substrate ($2.5 \times 2.5 \text{ cm}^2$) covered with a shadow mask at substrate temperatures (T_s) of 100 and 75 °C, respectively. The working pressure was $5.7 \times 10^{-3} \text{ Pa}$ at a DC power of 50 W. The degree of crystallinity of ITO was analyzed via X-ray diffraction (XRD). PLEDs were fabricated on both the ITO-coated PI and glass substrates. The ITO was O_2 plasma-treated at 140 W for 90 s to increase the surface energy and for cleaning purpose.

Poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS) (Heraeus, Germany) was selected as the hole transfer layer (HTL) and spin-coated onto the ITO. Red polymer (Livlux[®], Merck, USA), the emission layer (EL) material, was dissolved in toluene and spin-coated onto the PEDOT:PSS in a nitrogen-filled glove box. Cs_2CO_3 (Sigma–Aldrich, USA) solution (0.5 wt% in 2-ethoxyethanol) was successively spin-coated at 3000 rpm to serve as the EL as the electron transfer layer (ETL). A 100 nm-thick layer of Al was deposited by thermal evaporation to serve as the cathode. The thickness (h) of each layer was measured using a surface profiler (Alpha Step 500, KLA-Tencor Co., USA). The detailed PLED fabrication parameters are summarized in Table 1.

The structure of the PLEDs and the configuration of the compression test are schematically described in Fig. 1a and b, respectively. The abbreviations shown in parentheses in Fig. 1a correspond to the initial letter of each material. The dimension of a PLED cell was $5 \times 5 \text{ mm}^2$. By increasing the load (P in Fig. 1b), the compressive stress (σ_{zz} in Fig. 1b) was varied from 0 to 10, 30 50, and 100 kPa. During the compression of the PLED cells, the J - V characteristics were measured using a Keithley 2400 Sourcemeater, and the L - V relationships were evaluated using a Konica Minolta CS-200 luminance meter.

The temperature (T) changes of the PLEDs during operation were also measured using an infrared (IR) imaging camera. A thin layer of graphite was coated on the bottom of the substrate to reduce the reflection of IR light from the surface of the substrates. The photoluminescence (PL) of specimens that exhibited irreversible degradation in J - L - V under the applied stress was measured using a Macro Raman PL measurement system (Dongwoo Optron Co., Korea) with a 532-nm diode-pumped solid-state (DPSS) laser, and the result was compared with that for the load-free specimen.

The changes in the electronic energy structures induced by stress were investigated via UPS analysis (AXIS Ultra DLD, Kratos Inc., UK) using a helium (He-I) gas discharge lamp with an excitation energy of 21.2 eV and energy resolution of 200 meV over a sampling area with a diameter of 110 μm . A sample bias of -15 V was used to reduce static charge accumulation. The measurement procedures are schematically illustrated in Fig. 1c. The layers analyzed were deposited sequentially on the substrate and growing stack of layers. Then, the specimen was compressed to the load at which degradation occurred. The surfaces of the compressed layers were analyzed using UPS, and the results were compared with a load-free stack.

The changes in the interfacial chemistry between layers of the PLEDs were analyzed using XPS (K-alpha, Thermo Scientific[™],

USA) with a monochromatic Al-K α radiation source (1486.6 eV). The sampling area diameter and pressure inside the chamber during the measurements were 200 μm and $4.92 \times 10^{-9} \text{ mbar}$, respectively. The XPS analysis procedures are similar to those of the UPS analysis, as shown in Fig. 1c. To examine the morphological changes in the materials that constituted the PLEDs, the surface of each layer was analyzed using atomic force microscopy (AFM) (XE-Bio, Park Systems, Korea) and field effect scanning electron microscopy (FE-SEM) (JEOL-6701, JEOL Ltd., Japan).

3. Results and discussion

The XRD analysis results for ITO are shown in Fig. 2. Similar degrees of crystallinity were attained in the ITO grown on PI and that grown on glass. The major growth orientation was (222), which is known to have superior electrical properties and crystalline qualities compared with other orientations [20].

The thickness of each layer of the PLEDs was very thin compared with the width (Fig. 1a). Hence, the PLED cells can be regarded as a laminate composite with effective mechanical properties based on a rule of mixtures [21]. Assuming that the PLED cells were elastically deformed by σ_{zz} and the friction on them was negligible, the in-plane strains, ϵ_{xx} and ϵ_{yy} (transverse extension) in Fig. 1b, are related to the out-of-plane strain (axial contraction), ϵ_{zz} , as follows [21]:

$$\sigma_{zz} = E_z^{\text{eff}} \epsilon_{zz} \quad (1)$$

$$\epsilon_{xx} = -\nu_{zx}^{\text{eff}} \epsilon_{zz}; \quad \epsilon_{yy} = -\nu_{zy}^{\text{eff}} \epsilon_{zz} \quad (2)$$

$$\epsilon_{xx} = \epsilon_{yy} \quad (3)$$

In Eq. (1), E_z^{eff} is the effective elastic modulus of the composite (PLED) in the z -direction. ϵ_{xx} , ϵ_{yy} , ν_{zx}^{eff} , and ν_{zy}^{eff} in Eq. (2) are the strains in the x - and y -directions (Fig. 1b) and the effective Poisson's ratio of the composite relating strains in the x -, y -direction to that in the z -direction, respectively [21]. Based on the rule of mixtures [21], E_z^{eff} is calculated according to Eqs. (3) and (4), respectively [21,22]:

$$\frac{1}{E_z^{\text{eff}}} = \frac{\nu_1}{E_1} + \frac{\nu_P}{E_P} + \frac{\nu_R}{E_R} + \frac{\nu_C}{E_C} + \frac{\nu_A}{E_A} \quad (4)$$

where ν_1 , ν_P , ν_R , ν_C , ν_A , E_1 , E_P , E_R , E_C , and E_A are the volumetric ratios and elastic moduli of each layer of the composite. The subscripts on ν and E are defined in the previous section.

When considering the thicknesses and magnitudes of the elastic moduli of the constituent materials, E_z^{eff} in Eq. (4) is determined by that of PEDOT:PSS [23], which is approximately 5 GPa. Based on the calculation result and Eq. (1), ϵ_{zz} is in the range of 0–0.002% when σ_{zz} is increased from 0 to 100 kPa. These values are less than the elastic deformation limit (<0.2%) [21], which verifies our assumption. Because Poisson's ratio is less than 1, the magnitude of transverse extension due to the axial contraction described in Fig. 1b is less than 0.002%. However, high interfacial shear stresses are expected because of the small thicknesses of each layer.

Table 1
PLED fabrication parameters.

Sample types	Substrate	PEDOT:PSS		Red polymer		Cs_2CO_3		Al
		Type	Spin coating speed, time (rpm, s)	Conc. (wt%)	Spin coating speed, time (rpm, s)	Conc. (wt%)	Spin coating speed, time (rpm, s)	
P	PI	Clevios [™] PH 500	1500, 12	1.5	2000, 120	0.5	3000, 60	100
G	Glass	Clevios [™] P	3000, 60	1.2	1000, 60	0.5	3000, 60	100

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