Stable Bending Performance of Flexible Organic Light-Emitting Diodes Using IZO Anodes

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We report luminescent characteristics and mechanical stability of a flexible organic light-emitting diode (FOLED) using an amorphous ZnO-doped In$_2$O$_3$ (a-IZO) anode with a low sheet resistance of 18.9 $\Omega/\square$ and a high optical transparency of 86%. The FOLED consisting of a-IZO/poly(3,4-ethylenedioxythiophene) poly(styrenesulfonate) (PEDOT:PSS)/poly[(9,9-di-n-octylfluorenyl-2,7-diyl)-alt-(benzo[2,1,3]thiadiazol-4,8-diyl)] (F8BT)/LiF/Al exhibits the efficient luminescent characteristics, which are nearly identical with the photoluminescence spectrum of the organic emitting material in our FOLED. This observation clearly indicates that the luminescent characteristics of the FOLED are solely ascribed to molecular exciton formation within the F8BT layer, since exciplex and charge transfer exciton formation are strictly suppressed by both tunneling/thermionic injection of holes at the PEDOT:PSS/F8BT interface and enhanced hole transport. Furthermore, the use of the considerably flexible a-IZO anode and PEDOT:PSS acting as a strain-relief buffering material enables good retention of the efficient luminescent characteristics of the FOLED even after continuous bending of up to 1000 times.

In recent years, flexible organic light-emitting diodes (FOLEDs) have emerged as one of the basic components in next-generation display systems owing to the evolution of portable electronic devices that require lightweight, bendable and unbreakable display devices$^{1-3}$. To achieve the FOLEDs utilized in flexible display systems, it is essential to satisfy efficient luminescent characteristics and stable performance issues in the FOLED regardless of its repeated deformation. Over the past two decades, many studies have aimed at enhancing performance in OLEDs by employing multi-layer structures comprising organic semiconductors and a blended polymer as an organic emission layer (EML)$^{4-7}$. However, OLEDs with multi-layer structures usually exhibit exciplex and/or charge transfer (CT) exciton formations at the interface resulting in inefficient luminescent properties, which originate from the offsets of the highest occupied molecular orbitals (HOMOs) and the lowest unoccupied molecular orbitals (LUMOs) between two organic semiconductors$^{8,9}$. In order to design efficient FOLED with desirable and efficient luminescent properties, structural optimization should be considered together with the choice of appropriate materials in the FOLED. Herein, we propose a simple FOLED structure consisting of a EML made of poly[(9,9-di-n-octylfluorenyl-2,7-diyl)-alt-(benzo[2,1,3]thiadiazol-4,8-diyl)] (F8BT) and a hole transport layer (HTL) inserted between the flexible transparent anode and the EML. Poly(3,4-ethylenedioxythiophene) poly(styrenesulfonate) (PEDOT:PSS) is utilized here as a hole transporting material upon consideration of the work function of the anode and the HOMO/LUMO energy levels of F8BT$^{10,11}$. In addition, the PEDOT:PSS layer effectively improves the hole injection toward the F8BT layer which is well-known as an n-type organic semiconductor and is a trapping for holes$^{12}$. An amorphous ZnO-doped In$_2$O$_3$ (a-IZO) is chosen for our FOLED as the most appropriate anode material, because it has been strongly regarded as offering a very promising flexible transparent anode due to its remarkable mechanical flexibility as well as its excellent optical and electrical properties$^{13,14}$. The choice of the flexible transparent anode in the FOLEDs is one of the crucial issues related to a stable performance retaining luminescent characteristics regardless of its repeated deformation. In this study, in order to realize the highly efficient and stable performance, we design a FOLED with a simple structure of a-IZO/PEDOT:PSS/F8BT/LiF/Al on the flexible plastic and investigate its luminescent and electrical characteristics, and mechanical stability. Figure 1 shows molecular structures of PEDOT:PSS (left) and F8BT (right) (Fig. 1(a)), and a schematic diagram of the FOLED constructed on the flexible plastic (Fig. 1(b)).
IZO/PEDOT:PSS/F8BT/LiF/Al constructed on a flexible plastic substrate.

thickness of the resistance (material for good performance in FOLEDs.

Figure 1 | (a) The molecular structures of PEDOT:PSS (left) and F8BT (right), and (b) a schematic diagram of the FOLED with the structure of a-IZO/PEDOT:PSS/F8BT/LiF/Al constructed on a flexible plastic substrate.

Results

The optical transmittance spectra show that the optical transparency of the a-IZO film on a poly-ether-sulfone (PES) substrate is as high as 86% at a wavelength of 550 nm and its average optical transparency in the visible range (400–700 nm) is 83% as plotted in Fig. 2. This excellent optical transparency is comparable to those of other flexible transparent anodes such as single-walled carbon nanotube (SWCNT) (~90%) and Ag NWs network (~86%) deposited on polycarbonate or polyethylene terephthalate substrates. The sheet resistance of the a-IZO film measured in this work is as low as 18.9 Ω/□, and such a low sheet resistance allows for efficient hole injection from anode to organic semiconductor layers. From the optical transparency (T) and sheet resistance (Rsh) of the a-IZO film on the PES substrate, a figure of merits (ΦTC) of the transparent anode can be calculated using Equation (1), which is suggested by Haacke.

\[ \Phi_{TC} = \frac{T_{10}}{R_{sh}} \] (1)

The ΦTC value of the a-IZO film in this study is estimated to be 12.3 × 10^2 Ω^-1, which is higher than those of other flexible transparent anodes, indicating that the a-IZO film is a promising anode material for good performance in FOLEDs.

We examine a ratio of sheet resistance change-to-initial sheet resistance (ΔR/R0), as a function of bending radius. From the experimental results of this study, which are essential requirements for the construction of the FOLEDs. AuOx has a much lower sheet resistance of ~6.0 Ω/□, but it has a poor optical transparency (~60%), so that it is unsuited to be used as flexible anode for FOLEDs. In contrast, SWCNT and PEDOT:PSS/SWCNT nanocomposite show the good mechanical flexibility as well as the high optical transparency, but they have inferior electrical properties which lead to inefficient injection of holes from anode to organic semiconductor layers due to their high sheet resistance.

Figure 2 | Optical transmittance curves of the bare PES substrate and the a-IZO film on the PES substrate.

Discussion

Table 1 lists several key parameters of flexible transparent films including the experimental results of this study, which are essential requirements for the construction of the FOLEDs. AuOx has a much lower sheet resistance of ~6.0 Ω/□, but it has a poor optical transparency (~60%), so that it is unsuited to be used as flexible transparent anode for FOLEDs. In contrast, SWCNT and PEDOT:PSS/SWCNT nanocomposite show the good mechanical flexibility as well as the high optical transparency, but they have inferior electrical properties which lead to inefficient injection of holes from anode to organic semiconductor layers due to their high sheet resistance.

In case of PEDOT:PSS and conducting polymer, they are not appropriate to be utilized as the flexible transparent anodes, because of their high sheet resistance and high ratio of sheet resistance change under the continuous bending with a low strain. In spite of the excellent optical and electrical properties, furthermore, the ratio of the sheet resistance change of the Ag NW network increases to about 3 for the 250 bending times. This observation indicates the poor mechanical flexibility of the Ag NW network under the continuous
mechanical bending test, revealing its unsuitability as a flexible transparent anode for the FOLEDs. Compared with those flexible transparent anode films, the α-IZO film shows relatively superior properties in terms of sheet resistance, optical transparency, and mechanical flexibility. Hence, among flexible transparent anodes, the α-IZO anode sputtered in this work is the most appropriate flexible transparent anode for realizing efficient and stable performance in the FOLED.

In both of a normalized photoluminescence (PL) spectrum taken from the spin-coated F8BT layer (black) and a normalized electroluminescence (EL) spectrum of the fabricated FOLED at a biased voltage of 7.5 V (olive), the green colored emission is dominant as shown in Fig. 4(a); a photograph of the green colored emission from the FOLED in the dark is presented in the inset. The green colored emission is typically associated with keto-emission via keto-defects in F8BT23. The peak position (535 nm) and the full width

![Figure 3](image-url) | Ratio of sheet resistance change (ΔR/𝑅_0) of the α-IZO film sputtered on a flexible plastic substrate (a) as a function of bending radius and (b) as a function of the number of bending times (#). And the optical profiler images of the α-IZO films (c) before and (d) after 1000 times cyclic bending test with a bending radius of 10.5 mm, respectively. The inset shows a photograph when the bending test for mechanical flexibility of the α-IZO film is performed with a home-made bending machine.

| Anode        | Sheet resistance (Ω/□) | Transmittance@550 nm (%) | Work function (eV) | Figure of Merits (10⁻³ V² m⁻¹) | Mechanical flexibility | Ref. |
|--------------|------------------------|--------------------------|--------------------|--------------------------------|------------------------|------|
| α-IZO        | 18.9                   | 86                       | 5.2                | 12.3                           | -0.6                   | this study |
| AuOx         | 6.0                    | 60                       | 5.7                | 1                              | N/A                    | 15   |
| SWCNT        | 300                    | 90                       | 4.7–5.2            | 1.2                            | N/A                    | 15   |
| PEDOT:PSS    | 296                    | 82                       | 5.0                | 0.4                            | 0.9                    | 1600 |
| PEDOT:PSS/SWCNT nanocomposite | 40–60                  | 85                       | 4.9                | 0.1                            | 0.3                    | 1500 |
| Conducting polymer | 2000                 | 80–85                    | 4.5                | 3.4                            | 7.7                    | 250  |
| Ag NW network | 30                    | 86                       | 4                  | 1500                           | 20                     | 16   |

Table 1 | Several key parameters of flexible transparent films

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at half maximum (FWHM) (66 nm) of the EL spectrum of the FOLED are surprisingly identical to those of the PL spectrum obtained from the spin-coated F8BT layer, corresponding to 541 and 66 nm, respectively. This observation clearly reveals that the luminescent characteristics of the FOLED are solely ascribed to molecular exciton formation within the F8BT layer, implying a highly efficient performance in the FOLED. The phenomenon has rarely been observed in the field of the OLED, which implies a well-optimized structure of our FOLED without exciplex emission.

Considering the energy diagram of our FOLED as illustrated in Fig. 4(b), it is possible that our FOLED shows exciplex emission ascribed to interfacial exciplex states formed at the PEDOT:PSS/F8BT interface because the HOMO offset of ~0.7 eV between PEDOT:PSS and F8BT leads to accumulation of holes at the PEDOT:PSS/F8BT interfacial potential barrier. Actually, G. Bernando et al. reported the presence of an emissive exciplex between F8BT and PEDOT:PSS through the photophysical characterization using PL, PL excitation spectra and fluorescence decay measurement. And, in the EL spectrum of their previous study, two peaks were observed at 550 and 574 nm and the emission broadening was apparent from the difference in the FWHM of EL (~87 nm) and PL (~72 nm) spectra. However, against our expectation, no evidence for the exciplex emission at the PEDOT:PSS/F8BT interface is observed in the EL spectrum of the FOLED (Fig. 4(a)). The EL spectrum without exciplex emission can be understood as follows. When an electric field higher than the tunneling threshold voltage is applied to the FOLED, holes easily transport via tunneling and/or thermionic injection to the F8BT layer, so that molecular exciton formation is enhanced within the F8BT layer as depicted in Fig. 4(c). The higher turn-on voltage of ~4 V than the tunneling threshold voltage of ~3.8 V in this study demonstrates that tunneling and/or thermionic injection of holes at the PEDOT:PSS/F8BT interface are associated with the efficient luminescent characteristics of the FOLED. In addition, the absence of any CT emission in the EL spectrum (Fig. 4(a)) can be explained by the filling of sub-bands localized in the F8BT layer.

In comparison with the luminescent properties of OLEDs consisting of F8BT as the EML, the structure of a-IZO/PEDOT:PSS/F8BT/LiF/Al in this study shows the most efficient luminescent characteristics as demonstrated in Table 2. There are three valid reasons for the better performance of our FOLED compared with other F8BT based OLEDs: i) the PEDOT:PSS layer, ii) the a-IZO anode, and iii) the LiF/Al cathode. As mentioned above, PEDOT:PSS used as the HTL allows more holes to be trapped in the F8BT layer, and
Table 2 | Luminescent characteristics of OLEDs consisting of F8BT as the EML

| Structure                     | EL spectrum          | PL spectrum          | Comparison       |
|-------------------------------|----------------------|----------------------|------------------|
|                               | Peak position (nm)   | FWHM (nm)            | Peak position (nm) | FWHM (nm) | ΔPeak position (nm) | ΔFWHM (nm) | Ref.     |
| α-IZO/PEDOT:PSS/F8BT/LiF/Al   | 535                  | 66                   | 541              | 66        | 6                   | 0          | this study |
| cITO/PEDOT:PSS/F8BT/Ca/Al    | −541                 | −62                  | −562             | −65       | −21                 | −3          | 26       |
| cITO/PEDOT:PSS/F8BT/Mg       | −550                 | −87                  | −550             | −72       | −0                  | −25         | 24       |
| cITO/F8BT/Ca/Al              | −574                 | −88                  | −530             | −70       | −24                 | −18         | 23       |
| cITO/F8BT/Al                 | −560                 | −67                  | −533             | −100      | −27                 | −33         | 27       |

consequently the balanced electron and hole current in the F8BT layer contributes to efficient molecular exciton formation within the F8BT layer28. Furthermore, we consider our FOLED utilizing an α-IZO anode and LiF/Al cathode to be superior to others using a crystalline ITO (c-ITO) anode and Ca/Al cathode, since the amorphous structure of our α-IZO anode reduces the strain applied to the organic semiconductor layers relatively more than the c-ITO anode and LiF/Al cathode, which helps in minimizing non-radiative emission and/or maximizing out-coupling in the FOLED29,30. The FOLED structure proposed in this study, α-IZO/PEDOT:PSS/F8BT/LiF/Al, is ideal for demonstrating the luminescent property of the F8BT used as the organic emitting material in our FOLED.

A current density versus voltage (J-V) curve of our FOLED is plotted in Fig. 5 in log J-log V scale and reveals the existence of three regions associated with different types of charge carrier transport mechanisms. At lower voltage than an turn-on voltage of about 4 V, the current of charge carriers in region (i) follows Ohm’s law (J ∝ V) with a low leakage current density (<1 μA/cm²). This low leakage current density is associated with the absence of the PEDOT:PSS between the F8BT layer and the α-IZO anode. The PEDOT:PSS layer spin-coated on the α-IZO anode plays the role of a buffer layer relaxing the roughness of the surface before the deposition of the F8BT layer. As a result, the low leakage current density reduces the electrical short between the F8BT layer and the α-IZO anode31. In region (ii), where the biased voltage range from about −4 to −6 V, the relation of J ∝ V² indicates the mechanism of a trap-filled limited current (TFLC) which means that sub-bands in the F8BT layer are filled and the injected charge carriers are free to move32. This TFLC is basically concerned with luminescent properties of the FOLED that are nearly identical to the PL spectrum of the PEDOT:PSS layer. In order to evaluate the mechanical flexibility of our FOLED, the ratio of intensity change-to-initial intensity ([I − Io]/Io = ΔI/Io) and the peak position change of the EL spectrum at a biased voltage of 7.5 V were examined through a continuous bending test with a strain of 1% up to 1000 times. The intensity and the peak position of the EL spectrum obtained from the FOLED were measured at 50, 100, 250, 500, 750, and 1000 bending times, respectively. The intensity and the peak position of the EL spectrum are changed a lot for the first 50 bending cycles, and then remain stable for the 1000 bending cycles as shown in Fig. 6(a). This clearly reveals excellent stable performance of the FOLED attributable to the good mechanical flexibility of the α-IZO anode and the strain-buffering effect of the PEDOT:PSS layer used as the HTL. Polymers such as the PEDOT:PSS layer have usually been employed in the field of flexible optoelectronics in order to relieve the strain applied to devices33,34. The strain-buffering effect of PEDOT:PSS is confirmed from comparison with EL spectra of the FOLED in flat and bent states with a strain of 1% (as shown in Fig. 6(b)). The luminescent properties of the FOLED in flat and bent states with a bending radius of 10.5 mm (a bending strain of 1%) are nearly identical. The peak position of the EL spectrum of the FOLED in the bent state is completely identical to that in the flat state, which clearly demonstrates that the electronic structure of the F8BT layer is not changed even under a strain of 1%, by virtue of the strain-buffering effect of the PEDOT:PSS layer. The FOLED designed in this study has excellent flexibility properties, resulting from the good mechanical flexibility of the α-IZO anode and the efficient buffering role of the PEDOT:PSS layer.

In conclusion, we demonstrate and examine the luminescent characteristics and mechanical stability of a FOLED consisting of solution-processed PEDOT:PSS and F8BT layers on the α-IZO anode with a high optical transparency of 86%. The low sheet resistance of 18.9 Ω/□ of the α-IZO film is nearly constant under a cyclic bending test with a strain of 1%, which reveals its suitability as a flexible transparent anode to realize stable performance in the FOLED regardless of its repeated deformation. The EL spectrum of the FOLED is nearly identical to the PL spectrum of the spin-coated F8BT layer. These efficient luminescent characteristics of the FOLED originate from molecular exciton formation within the F8BT layer, since the FOLED is clearly optimized with the structure of α-IZO/PEDOT:PSS/F8BT/LiF/Al. Moreover, this efficient performance in the FOLED is retained well even after the cyclic bending test performed up to 1000 times, which is attributable to the good mechanical flexibility of the sputtered α-IZO anode and the strain-buffering effect of the PEDOT:PSS layer. Our results reveal that FOLEDs with efficient and stable performance are made with FOLED structures suitable for organic emitting materials. Both structural optimization and the choice of materials in the FOLEDs are determining factors in order to realize efficient and stable performance FOLED for the development of future flexible display systems.
Figure 6 | (a) Ratio of intensity change (ΔI/I₀) (left) and peak position (right) of the EL spectrum obtained from the FOLED measured as a function of the number of bending times (N) up to 1000 times and (b) the normalized EL spectra of the FOLED in flat and bent states, with a bending radius of 10.5 mm (a bending strain of 1%); inset shows a photograph of the green colored emission from the FOLED when it is in the bent state.

Methods

Fabrication of the FOLEDs. The PES substrate with a thickness of 200 μm (i-components Co., Ltd.) was used as a flexible substrate (25 × 25 mm²) due to its excellent optical transparency and mechanical flexibility⁴. PEDOT:PSS and F8BT were purchased from HC Stark (Clevios AI 4083) and Sigma-Aldrich, respectively. The FOLED consisting of PEDOT:PSS and F8BT was constructed on the flexible plastic according to the following procedures. At first, a-IZO film was patterned on the flexible plastic by photolithography process using a photo mask, and sputtered from an IZO target (10 wt.% In₂O₃-doped In₂O₃) at room temperature under pure ambient Ar. The a-IZO film was then treated with UV-ozone for 20 minutes to improve the hydrophilic property and to remove organic contaminants such as photosensitive materials⁵—⁶. Moreover, the UV-ozone treatment enhances the injection efficiency of holes for a high work function anode³⁸. After the UV-ozone treatment, the PEDOT:PSS layer was formed on the a-IZO anode by spin-coating method, and then dried on a hot plate at a temperature of 90°C for 30 minutes, in order to remove solvent. F8BT dissolved in p-xylene (10 mg/ml) was spin-coated on the PEDOT:PSS layer, and dried at 70°C for 30 minutes. Finally, LiF (1 nm) and Al (100 nm) as the cathode of the FOLED were deposited by thermal evaporation. The emission layer area of the fabricated FOLED was 3 × 3 mm².

Measurements. The optical transparency and sheet resistance of the a-IZO film sputtered on the PES substrate were measured by a spectrophotometer (Shimadzu UV-3101PC) and a four-point probe system (Advanced Instrument Technology CMT-series). A real-time characterization of the bending test was obtained from the non-contacting optical profiler (Vecco NT-1100). The electrical characteristics of the fabricated FOLED were examined with a semiconductor parameter analyzer (Agilent 4155C). The PL spectrum of the spin-coated F8BT layer and the EL spectrum of the FOLED were taken with a spectrofluorophotometer (Ocean Optics USB2000plus), respectively. Mechanical bending tests of the a-IZO film and the fabricated FOLED were performed with a home-made bending machine⁴⁴. All measurements were carried out in air at room temperature.

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**Author contributions**

S.K. analyzed the experimental results and wrote the manuscript. K.C. contributed to preparation of the organic materials as well as transparent flexible films, and prepared manuscript. K.K. fabricated all samples and conducted the experiments, and analyzed the experimental data. All authors analyzed and discussed the experimental results.

**Additional information**

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