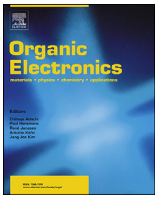
Organic Electronics 28 (2016) 123–134

Contents lists available at ScienceDirect

Organic Electronics

journal homepage: www.elsevier.com/locate/orgel



Heat evolution and dissipation in organic light-emitting diodes on flexible polymer substrates

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ARTICLE INFO

Article history:
Received 23 July 2015
Received in revised form
1 October 2015
Accepted 22 October 2015
Available online 11 November 2015

Keywords:
Flexible organic light-emitting diodes (f-OLEDs)
Polymeric substrates
Heat evolution
Heat dissipation

ABSTRACT

We investigated the effect of the heat generated in organic light-emitting diodes (OLEDs) on their performance with a focus on the thermo-physical properties of the substrates. OLEDs were fabricated on polyethylene terephthalate (PET), polyimide (PI), and glass substrates, and the instantaneous and time-resolved performances of the OLEDs were compared. The device operation temperature (T) was predicted via heat transfer analysis using the finite element method (FEM). The value of T during operation was experimentally measured using an infrared (IR) camera, and the results were compared with the numerically calculated values. The effects of T on the time-resolved electroluminescence (EL) spectra of the OLEDs on the different substrates were also investigated. The experimental results of this study agreed well with the numerical predictions that the T of the OLEDs on the polymers are higher than the T of the OLEDs on glass during operation due to the relatively low thermal diffusivity (α) of the polymer substrates used in this study. The results also showed that the performance and reliability of the flexible OLEDs (f-OLED) are highly dependent on the heat extraction capabilities of the substrates; thus, the current density (J), luminescence (L), voltage (V) characteristics, and efficiencies (η) over time of the OLEDs on PET and PI are inferior to the OLEDs on the glass substrate.

1. Introduction

The demand for flexible devices on compliant substrates has driven the development of transparent conductive electrodes (TCEs) on flexible polymeric substrates compared with traditional rigid glass substrates [1,2]. Improvements in the mechanical flexibility and functional properties of TCEs have produced flexible devices, such as flexible organic light-emitting diodes (f-OLEDs) and flexible organic solar cells (f-OSCs). These devices have demonstrated excellent performance on flexible polymeric substrates, which is similar to their performance on rigid substrates [3–5].

The performance of flexible devices was achieved via a combination of technical improvements in the synthesis of organic semiconductor materials and the fabrication processes. For example, the formation of crystalline nanofibrils in the OSC absorption layer generated improvements in the power conversion efficiency without high-temperature annealing [6]. By inserting various charge transport or buffer layers into the device architecture, the light output and efficiency of OLEDs were also significantly enhanced [7,8].

However, technical barriers to flexible devices must be overcome before they can match the performance of solid-state devices. In particular, the removal of heat from these devices during operation is a critical issue for their long-term reliability. Efficient heat extraction from these devices is vital for reliable performance and reasonable lifetimes of these devices [4,9,10]. Due to the low electrical-to-optical energy conversion efficiency of OLEDs, a considerable portion of the input energy is converted into heat, which chemically and physically degrades the organic materials that comprise the integral parts of OLEDs [11,12].

The low glass transition temperatures (T_g) of the organic semiconductor materials in OLEDs cause the organic layer to undergo morphological transformations at elevated temperatures [9,13]. The stacking of the conjugated polymers, which helps transport charge particles, is disrupted and thus degrades the charge recombination and light emission of OLEDs. The chemical states of the organic materials were also shown to be affected by heat due to the formation of unwanted reactions among the molecules in the different component materials present [10].

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http://dx.doi.org/10.1016/j.orgel.2015.10.023
1566-1199/

Many studies have addressed the thermal management of OLEDs to mitigate heat-induced device degradation [14–16]. Modification of the cathode thickness and composition has resulted in a reduced *T* gradient in OLED devices due to the improved heat dissipation performance of the cathode [14]. Similar heat extraction improvements were observed when thin-film encapsulation was used instead of conventional and epoxy-filled glass encapsulation [15,16]. However, the majority of the thermal analyses performed to date have focused on OLEDs on rigid substrates, such as glass and silicon; thus, additional exploration of the heat evolution and dissipation in f-OLEDs on polymeric substrates is required.

This study is important because the performance of f-OLEDs is approaching the performance of conventional OLEDs on more thermally conductive rigid substrates, even though the polymeric substrates used by f-OLEDs can be generally considered as thermal insulators. Considering the rapid development of the performance of f-OLEDs, in-depth knowledge of the significant factors of the thermal properties of polymer substrates and the performance of f-OLEDs is required.

In this study, we investigated the heat evolution and dissipation in f-OLEDs on polyethylene terephthalate (PET), which has a very low T_g, and PI, which has a relatively high T_g. OLEDs composed of the same materials and structures as the OLEDs on polymer substrates were fabricated on conventional rigid glass substrates and used as reference devices. Their performances were compared with a focus on the effect of the thermo-physical properties of the substrates. The *T* distributions during operation were predicted using FEM analysis and experimentally measured using an infrared IR camera, whereas the variations in *J*, *L*, and *V* characteristics were determined using a source meter and chroma-meter system by varying *V* with a fixed *V* for 5 min. The structure of the OLEDs and the measurement system are schematically described in Fig. 1(a) and (b), respectively. The effects of the thermal stress on the electronic band structure of the organic semiconductor materials of the OLEDs were analyzed using ultraviolet photoelectron spectroscopy (UPS) and verified by measuring the EL spectra of the thermally stressed devices.

2. Experimental procedures and model descriptions

2.1. Experimental procedures

Slide glasses (Marienfeld, Germany) with a thickness of 1 mm were used as glass substrates. The thickness of the PET substrate was 188 μm, and the PET contained a hard backside coating. The thickness of the colorless PI (Mitsubishi Gas Chemical Company, Japan) was 200 μm. The glass, PET and PI substrates were cleaned by sonication in isopropyl alcohol and deionized water baths prior to the deposition of indium tin oxide [9]. ITO was coated on the PET, PI, and glass materials as the transparent anodes for the OLEDs.

Crystalline ITO (*c*-ITO) films with a thickness of 100 nm were deposited onto the PET, PI, and glass substrates via direct current (DC) magnetron sputtering. The substrate temperature was maintained at 75 °C during deposition. The ITO target, which had a 90:10 weight ratio of In_2O_3 to SnO_2, was obtained from the Advanced Nano Products Company in Korea. During sputtering, Ar and O_2 gas flows were maintained at 70 sccm and 0.1 sccm, respectively. The sputtering power was fixed at 50 W.

Using a four-point probe, the sheet resistance (R_s) of the *c*-ITO films was measured. The as-deposited *c*-ITO films on the glass were annealed for 30 min inside a nitrogen glove box at 150 and 200 °C to reduce the R_s of the *c*-ITO films on the glass to below 140 Ω/sq. The *c*-ITO films and OLED names, which are summarized in Table 1, were selected based on the R_s of the *c*-ITO anodes. The variation in the degree of crystallinity of the *c*-ITO films was analyzed using a high-resolution X-ray diffractometer (HR-XRD) at a 1.2° incident angle.

Poly(2,3-dihydrothieno-1,4-dioxin)-poly(styrenesulfonate) (PEDOT:PSS, Clevios P, Heraeus Clevios GmbH, Germany), red polymer (livilux™ SPR-001 from Merck KGaA, Germany), and cesium carbonate (Cs_2CO_3 from Sigma–Aldrich) were used as the hole transport layer (HTL), emissive layer (EML) and electron transport layer of the OLED, respectively (Fig. 1(a)). The PEDOT:PSS solution was spin coated at 6000 rpm for 60 s and annealed at 90 °C for 30 min. Inside a nitrogen gas glove box, a 1.2-wt% red polymer solution in toluene was spin coated at 1000 rpm for 60 s and annealed at 90 °C for 30 min. The ETL, which was composed of 0.5-wt% Cs_2CO_3 in 2-ethoxyethanol, was spin coated at 6000 rpm for 60 s and annealed for 20 min. The samples were transferred to a separate evaporation chamber to deposit 100-nm Al films as the cathode.

The *J*–*V* and *L*–*V* characteristics of the OLEDs were measured using a Keithley 2400 source-meter and a Konica-Minolta CS-200 chroma-meter, respectively. The *T* profiles and variations during the *L*–*V* testing were measured using an IR imaging camera (NEC Avio InfRec R300). The chroma-meter was setup perpendicular to the surface of the OLEDs. The IR camera was positioned 20° from the normal of the OLED surface. The *T* measurement varied by less than 5 °C from this setup when the OLED surface was coated with a thin carbon coating, and the IR camera was oriented perpendicular to the surface of the devices. The instrumental setup to determine the *L*–*V* characteristics and *T* is schematically shown in Fig. 1(b). The variations in *J*, *L*, and *T* were measured both by varying the applied *V* and at a fixed *V* for a certain period of time. Data from at least two specimens for each sample were analyzed.

To determine the onset of morphological changes with respect to the temperature of the red polymer, the T_g of the red polymer was analyzed using a differential scanning calorimeter (DSC). The variation of the electronic band structure of thermally stressed organic polymers was analyzed using UPS. The thermal stress was applied to the PEDOT:PSS and red polymer by heating the organic polymer films on an ITO-coated Si wafer at 140 °C for 5 min inside a nitrogen-gas-filled glove box prior to UPS analysis. The OLEDs on the polymer and glass substrates were also heat-treated under the same conditions. The time-resolved EL spectra variations of the thermally stressed OLEDs were measured using a Konica CS 2000 spectroradiometer. To minimize the effects of the atmospheric water vapor and oxygen on the device performance during EL analysis of the thermally stressed OLEDs, the OLEDs were encapsulated with glass, which was attached to the devices using UV-curable epoxy, inside a nitrogen-gas-filled glove box before the EL analysis began.

2.2. Numerical analysis

FEM analysis (Abaqus Student Edition package, Providence, Rhode Island, USA) was used to determine the *T* distribution in the OLEDs during operation. To simplify the calculation to a two-dimensional problem, the specimen was modeled as a cylindrical and axisymmetric specimen, as shown in Fig. 1(c); steady-state and transient heat transfers were simulated via this two-dimensional model. The thickness of the model was assumed to be the thickness of the substrates (Table 2) and the OLED component layers (Table 3) combined but excluding the electron transport layer, which was less than 10 nm. The lengths of the OLED and substrates were considered to be 1.5 and 4.5 mm, respectively, which were half of the real device dimensions.

At the axis of symmetry, a thermally insulated boundary was defined. Natural convection was defined with an environmental temperature of 22 °C on the surfaces of the OLED and the substrate.