Organic Electronics 15 (2014) 1815–1821

Contents lists available at ScienceDirect

Organic Electronics

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

Analysis of alternating current driven electroluminescence in organic light emitting diodes: A comparative study

Le Zhang [a,b], Hajime Nakanotani [a], Kou Yoshida [a], Chihaya Adachi [a,b,*]

[a] *Center for Organic Photonics and Electronics Research (OPERA), Kyushu University, 744 Motooka, Nishi, Fukuoka 819-0395, Japan*
[b] *International Institute for Carbon Neutral Energy Research (WPI-I2CNER), Kyushu University, 744 Motooka, Nishi, Fukuoka 819-0395, Japan*

ARTICLE INFO

Article history:
Received 13 February 2014
Received in revised form 16 April 2014
Accepted 9 May 2014
Available online 28 May 2014

Keywords:
AC-driven double-insulated OLED
Space charges
Phase shift
EL saturation

ABSTRACT

The alternating current (AC) responses of double-injection and double-insulated organic light-emitting diodes (OLEDs) were investigated and compared. To reveal the electroluminescent (EL) processes in these devices, the AC voltage and frequency dependence of the EL intensity and capacitive current were studied in the time domain with a focus on phase difference analysis. It was found that the voltage-dependent transit time and frequency-dependent carrier distribution were important for the AC-driven performance of the double-injection OLEDs. In contrast, although the double-insulated OLEDs shared some similarities with the double-injection OLEDs, they had some unique characteristics, which were the absence of resistive current and phase shift of EL profiles. It was revealed that the EL in the double-insulated OLEDs was driven by the displacement current generated by the ionization of the doped layers, which, however, formed space charge regions and undermined the EL emission. The space charge redistributed the electric field across the devices after the initiation of EL, making the EL maintain for a limited time interval. This effect was significant under low frequency and high AC voltage. Comparing the phase difference between both devices, it was indicated that the space charge effect was responsible for the observed EL phase shift and the asymmetric EL profiles at low frequency and high AC voltage in the double-insulated OLEDs. The proposed model was also of help to understand the EL saturation phenomena with AC frequency and voltage in those devices.

1. Introduction

Organic light emitting diodes (OLEDs) have advanced rapidly during the last several decades from academic studies to real applications [1–3]. To further explore the ability of OLEDs, new operating mechanisms are expected. Currently, alternating current (AC)- or field-driven organic electro-luminescent (EL) devices are drawing much attention because of their unique characteristics such as less dependence on metal electrodes than traditional OLEDs, easy encapsulation, and possible operation under AC power lines [4–9].

In fact, the idea of AC-driven OLEDs was firstly proposed to make use of the reverse bias region [4,10]. The devices used were common double-injection type OLEDs. Although traditional OLEDs were initially believed to emit light only under forward bias conditions, after introducing two polyaniline layers, light emission was observed under both forward and reverse bias conditions [11–13]. Meanwhile, to avoid the problems associated with low-work-function metal electrodes, organic thin-film electroluminescent devices with a double-insulated structure were fabricated [5,14–16]. Although these devices worked under AC-driven

* Corresponding author at: Center for Organic Photonics and Electronics Research (OPERA), Kyushu University, 744 Motooka, Nishi, Fukuoka 819-0395, Japan. Tel.: +81 92 802 6920; fax: +81 92 802 6921.
E-mail address: adachi@cstf.kyushu-u.ac.jp (C. Adachi).

http://dx.doi.org/10.1016/j.orgel.2014.05.009
1566-1199/

conditions, the driving voltages were rather high. Recently, another structure for AC-driven OLEDs including one injection electrode and one blocking electrode, i.e., a single-insulated structure, has been reported [7,17,18] However, this structure requires careful considerations of the injection electrode [19]. Because of their unique properties, we focused on OLEDs with double-insulated structures in this study.

As to double-insulated OLEDs, there are two important issues. One is the charge carrier generation process, and the other is the space charge effect. Several methods have been proposed for the charge carrier generation process, such as by introducing nanoparticles (NPs), colloidal quantum dots, or doping layers (p–i–n structure) [5,8,9]. In each case, there is a corresponding operating mechanism. For the p–i–n structure, it is suggested that charge carriers (holes and electrons) are generated in the charge generation layers (p-doped and n-doped regions) when the forward AC voltage exceeds the turn-on voltage. Then, the generated carriers are driven by the AC voltage, leading to bipolar carrier transport and radiative recombination. The ionized dopants can be neutralized under the reverse bias condition through a proposed tunneling mechanism [9]. In this way, the electroluminescence (EL) is repeatedly emitted under the forward bias condition. For the p–i–n structure, the carrier concentration can be tuned by the doping concentration [9]. In addition, the operating voltage can be greatly reduced by using insulators with a high dielectric constant [8,20].

Because double-insulated OLEDs are not injection-type, the ionized charge generation layers form space charge regions during the forward bias condition (charge generation process), which redistribute the electric field across the device [8,21]. Besides the ionized charge generation regions, the trapped charges also serve as an important source of the space charge [22–24]. Before the initiation of EL, the ionized dopants are electronically screened by the mobile carriers. The applied AC voltage is mainly to drive the mobile carriers to counter electrodes. However, once the EL occurs, the amount of mobile carriers decreases, leading to an additional electric field generated by the unscreened space charges. The direction of the generated electric field is opposite to the applied AC field, resulting in a decrease of the voltage across the emitting layer and sharp electroluminescent profiles in the time domain [8,9,20,21]. In other words, under the forward bias condition, there are four processes, i.e., ionization of the doped layers, double-carrier transport, recombination and space charge field establishment. The space charge effect will be discussed in the following sections. Although there are increasing studies on the double-insulated OLEDs, the above two issues have still not been completely addressed [8,9,20,25,26]. For a clear understanding of the operating mechanism of double-insulated OLEDs, a comparison of double-injection and double-insulated OLEDs is expected.

In the present study, the characteristics of double-injection and double-insulated OLEDs under AC-driven conditions were investigated in a wide frequency region and compared with a focus on their AC frequency and amplitude dependence. Experimental results suggested that although both devices shared some behavior, the double-insulated structure possesses several unique characteristics that are attributed to the limited charge generation ability and space charge effect. In addition, it was indicated that the phase difference analysis, which was overlooked in the previous studies, was of help for a clear understanding of the operating mechanism.

2. Experiments

The devices used here have a homo-junction triple-layer p–i–n diode structure. The double-injection device (Device A) was fabricated as follows. A patterned indium tin oxide (ITO) substrate was used as a transparent anode after cleaning and UV/ozone treatment. Then, a hole injection layer of MoO_3 (20 wt.%) doped 4,4′-bis[(N-carbazole) styryl]biphenyl (BSB-Cz) with a thickness of 70 nm was evaporated on the ITO surface [24,27]. Next, a neat BSB-Cz layer with a thickness of 50 nm was deposited. Subsequently, a 50 nm thick layer of Cs (20 wt.%) doped in BSB-Cz was deposited as an electron injection layer. Finally, an Ag electrode with a thickness of 100 nm was deposited as the cathode. During the evaporation processes, the vacuum pressure was kept to about 10^{-4} Pa. The film thickness was monitored using a quartz crystal microbalance and the evaporation rate was about 1 Å/s. For the double-insulated device (Device B), the electrodes and organic layers were exactly the same as those of Device A. The carrier injection layers in Device A served as carrier generation layers in Device B. Hafnium oxide (HfO_2) insulating layers were prepared by rf-sputtering from a HfO_2 target with a diameter of 5 cm under argon atmosphere with a flow of 50 sccm. The sputtering power was set to about 50 W, resulting in an evaporation rate of about 2.1 nm/min. The thickness of the HfO_2 insulating layers was about 100 nm. All devices were encapsulated in dry nitrogen atmosphere shortly after fabrication. The structures of both devices are shown as the insets in Fig. 1.

For the AC characterization, a wave function generator (NF, W1974) was used to produce sinusoidal wave with frequency from 1 Hz to 1 MHz, which was amplified by a bipolar high-speed amplifier (NF, HSA4101) from 0 to 75 V. In the study, the device characteristics were measured with respect to the AC voltage amplitude, i.e., 0.5 V_{pp}, where V_{pp} is the peak-to-peak voltage. The intensity of electroluminescence (EL, V_{EL}) was measured using a photo-multiplier tube (Hamamatsu, R925). During experiments, the EL intensity was monitored with respect to AC amplitude and frequency. In addition, the current flowing through the devices ($I_{R_s} = \frac{V_{R_s}}{R_s}$) was measured using a 50-ohm resistor (R_s) connected in series with the devices. All signals were displayed in an oscilloscope (Agilent, DSO5034A).

3. Results and discussion

3.1. Double-injection OLEDs (Device A)

Fig. 1(a) shows the dependence of EL intensity on the applied AC voltage amplitude (V_{ac} = 0.5 V_{pp}) at an AC frequency of 1 kHz for Device A. For comparison, its current