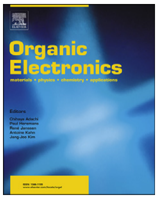
Organic Electronics 39 (2016) 348–353

Contents lists available at ScienceDirect

Organic Electronics

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



Transient analysis on stored charges in organic light-emitting diodes and their application in alternating current driven electroluminescence

Chengwen Zhang [a, b], Bo Qiao [a, b], Suling Zhao [a, b, *], Zheng Xu [a, b], Peng Wang [a, b], Yuening Chen [c], Feifei Yin [a, b, c], Gilbert Teyssedre [d], Christian Laurent [d], Xurong Xu [a, b]

[a] *Key Laboratory of Luminescence and Optical Information, Ministry of Education, Beijing Jiaotong University, Beijing 100044, China*
[b] *Institute of Optoelectronics Technology, Beijing Jiaotong University, Beijing 100044, China*
[c] *Physics Department, Liaoning University, Shenyang 110036, China*
[d] *Université de Toulouse, UPS, INPT, CNRS, LAPLACE (Laboratoire Plasma et Conversion d'Energie), 118 route de Narbonne, F-31062 Toulouse Cedex 9, France*

ARTICLE INFO

Article history:
Received 9 August 2016
Received in revised form
22 September 2016
Accepted 10 October 2016

Keywords:
OLEDs
Stored charges
Transient EL measurement
AC driven EL

ABSTRACT

AC-driven organic light-emitting diodes (OLEDs) can overcome some reliability-related drawbacks to traditional DC-OLEDs. They imply the use of insulating layers in the device. In this work OLEDs containing an insulating layer of poly (methyl methacrylate) (PMMA) in storing charges with the thickness of 2, 6, 8, 10 nm have been prepared and investigated. The emission mechanisms of the device are analyzed considering transient and AC electroluminescence (EL) measurements. We show that charges are stored in the PMMA layer as surface charges and bulk charges. The former contribute to the occurrence of EL spike after the driving pulse with a decay tail for about 80 μs to 10nm PMMA device, and the latter can be released to emit light under reversed voltage more than 2 V because they are immobile unless under the stronger reversed field. Stored charges commonly are harmful for the performance of OLEDs devices due to quenching, nonradiative transition and the thermal energy accumulation even degradation. Whereas when operating under alternating current (AC) stress, we not only obtain the EL peak from injection charges under forward voltage, but also obtain another peak below the built-in voltage 4.3 V, whose peak point lies in −0.2 V. It turns to originate from stored charges so they become beneficial instead of weakening EL for reusing to produce light. All the results pave the way to realize AC driven OLEDs devices using these stored charges, to uncover the AC EL mechanisms and to improve their EL performance.

1. Introduction

Organic light-emitting diodes (OLEDs) have been a very promising option in flat-panel displays and solid state lighting applications since the first breakthrough by Tang et al., in 1987 [1], and great efforts have been undertaken to achieve the dramatic improvement of their EL performance [2–8]. Until now, the external quantum efficiency (EQE) of OLEDs with thermally activated delayed fluorescence (TADF) material can reach up to 29.6% [9]. Though great progress has taken place, there are still some problems that cannot be ignored, e.g. excess charges. These charges can always exist in OLEDs during operation in various energy levels, so as to form stored charges [10–12], which does harm the performance of OLEDs. First, excitons can be quenched by extra polarons [13]. Second, stored charges can lead to field-induced quenching because of the additional electric field created by these charges [14]. Third, the trap-assisted carriers result in the non-radiative recombination of charges to compete with the radiative emitting, and eventually to decrease the EQE [12,15]. Finally, the storage of charges in the interface leads to local overheating and even degradation [16]. Taking into account that these charges go against the performance of devices, it is urgent to address the problem.

Recently, unlike the traditional OLEDs driven by direct current (DC), a novel device structure, alternating current (AC) driving OLEDs (AC-OLEDs), have drawn increasing attention [17–26]. To realize operation under AC voltage, a dielectric layer can be

---

* Corresponding author. Key Laboratory of Luminescence and Optical Information, Ministry of Education, Beijing Jiaotong University, Beijing 100044, China.
E-mail address: slzhao@bjtu.edu.cn (S. Zhao).

http://dx.doi.org/10.1016/j.orgel.2016.10.019
1566-1199/

introduced into the device in order to form light emitting capacitor. Currently, there are two basic architectures in fabricating AC-OLEDs device, i.e., symmetric and asymmetric structures. The former is using two insulator layers with organic layer between them, while the latter is just using one insulator adjacent to organic layer, which refers to different electron dynamic mechanisms. For symmetric structure, as no charges can be injected owing to insulator layers laid on both electrodes, the electroluminescence (EL) can take place in two ways that are far away from the common mechanism in OLEDs. The first is based on solid-state cathodoluminescence (SSCL) [17–19], and the second arises from carriers self-generated by inserting conducting-doping layers [21,26]. For asymmetric structure, as placing only one insulator layer, the EL can arise from the recombination of holes and electrons injected from the electrode without insulator layer [20,23–25]. Additionally, by inserting charge generation layer into electron only device, it is also possible to realize the AC induced EL [22], whose mechanisms are the recombination of injected and generated charges from different junctions under AC voltage. In general, AC driving EL (AC-EL) has its own noticeable advantages, e.g., when inserting dielectric layer, it can prevent possible electro-chemical reaction between organic layer and electrode and protect the active layer from the degradation due to moisture and oxygen in the atmosphere [20]. When operating under the frequent reversal voltage, the accumulation of charges can be avoided [21]. Hence it can pave a way to make use of the stored charges in OLEDs, to uncover the AC-EL mechanisms and to improve their EL performance [27,28].

In this paper, the OLEDs with thin insulator layer PMMA were prepared. With the transient measurement, the stored charges were found and the electron dynamics mechanisms were elucidated. To efficiently make use of these charges, AC voltage was chosen to drive the devices. It was found that the AC-EL not only included EL driven by injection but also included EL of stored charges, which is distinguished from the common AC-EL processes. By transient analysis, we succeeded to demonstrate the electron dynamics mechanisms of stored charges induced AC-EL, which provides a way to take advantages of these harmful stored charges.

2. Experimental details

The devices were fabricated on patterned indium tin oxide (ITO) with a sheet resistance 20 Ω/□. The structure was carried out as ITO/PEDOT: PSS/poly (methyl methacrylate) (PMMA) (2 nm, 6 nm, 8 nm, 10 nm)/Tris-(8-hydroxyquinoline) aluminum (Alq$_3$) (60 nm)/LiF (0.6 nm)/Al (80 nm). The structure and energy level diagram of the OLEDs are shown in Fig. 1. The ITO substrate was cleaned with detergent water, ethanol and de-ionized water in sequence by an ultrasonic cleaner and dried with nitrogen, then followed by a UV-ozone treatment for 10 min. PEDOT: PSS was prepared by spin-coating at 4000 round per minute (rpm) for 40 s and annealed at 150 °C for 10 min. PMMA was dissolved in chloroform and spin-coated on ITO with different speeds to form various thickness. Alq$_3$ was thermally evaporated at pressure of 5×10^{-4} Pa. Then LiF and Al were sequentially deposited under a high vacuum condition of 2×10^{-4} Pa through a shadow mask. Their thicknesses were monitored and controlled with quartz crystal monitors. The active area of the device is 0.09 cm^2.

The electroluminescence (EL) spectra were detected by a charge-coupled device (CCD) spectrometer. The transient EL was performed under a series of forward and reversed pulses. The forward pulse and AC voltage were both generated by RIGOL DG1022 Function/Arbitrary Waveform Generator, and the reversed pulse was generated by Agilent 8114A High Power Pulse Generator. Both the generators were controlled by DG535 Digital Delay/Pulse generator. The characteristics of transient EL were detected by a Zolix Instruments Model PMTH-S1C1-CR131 Photomultiplier Tube and recorded with a Tektronix Model DPO 4104 digital phosphor oscilloscope. All measurements were carried out at room temperature under ambient atmosphere.

3. Results and discussion

3.1. EL characteristics and transient EL measurements

To investigate the influence of PMMA layer on the OLED devices, their spectra were firstly measured under DC condition, which is

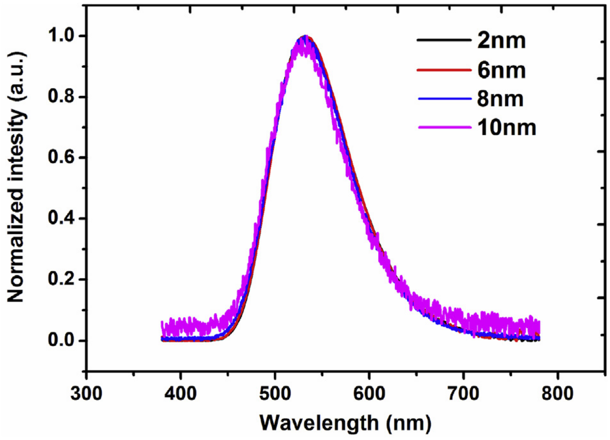


Fig. 2. The EL spectra of OLEDs device with different PMMA thickness of 2 nm, 6 nm, 8 nm and 10 nm under DC condition.

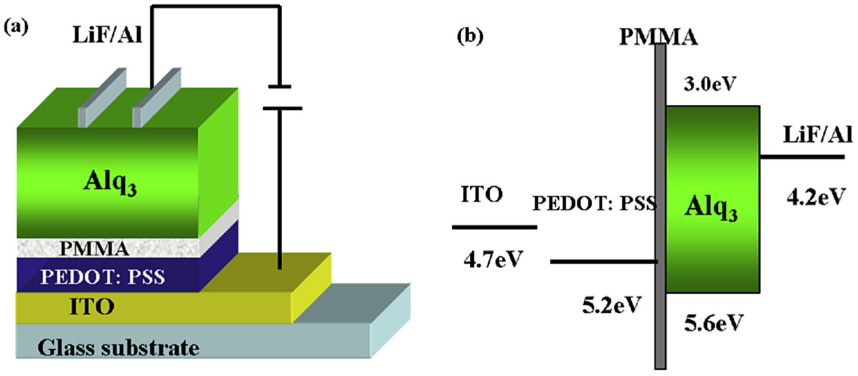


Fig. 1. (a) Device structure of the OLEDs (b) Energy level diagram of the OLEDs.