

High-current stressing of organic light-emitting diodes with different electron-transport materials

L. Liu^a, S. Li^a, Y.M. Zhou^{a,*}, L.Y. Liu^a, X.A. Cao^{a,b,*}

^a School of Electrical and Electronic Engineering, Hubei Provincial Key Laboratory of Green Materials for Light Industry, Hubei University of Technology, Wuhan 430068, China

^b Department of Computer Science and Electrical Engineering, West Virginia University Morgantown, WV 26506, USA

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ABSTRACT

We conducted accelerated reliability tests of electron-only devices (EODs) and organic light-emitting diodes (OLEDs) differing only in their electron-transport material (ETM). High current stressing of EODs at 50 mA/cm² showed that Bphen ~ Alq₃ > TPBi > TAZ in terms of intrinsic material stability. In addition, the lowest unoccupied molecular orbital (LUMO) level and electron mobility have been identified as two other key material factors affecting the degradation rate of OLEDs. TAZ has a low electron mobility, a LUMO level misaligned with the Fermi level of the cathode, and poor material stability, leading to extremely poor reliability of devices with a TAZ electron-transport layer (ETL). In contrast, the OLED with a Bphen ETL exhibited more stable operation and a 76× longer luminance lifetime. Due to its relatively high electron mobility and good stability as well as perfect energy level alignment with the cathode, Bphen has proven to be the most desirable ETM from the standpoint of OLED reliability.

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1. Introduction

The research and development of organic light-emitting diodes (OLEDs) has attracted considerable interest and led to significant progress over the last two decades [1,2]. The use of OLEDs in flat-panel displays has become a reality. However, OLEDs still suffer from a relatively short lifetime at high brightness, which poses a big challenge for their widespread acceptance for many commercial applications [2]. The reliability issue has been attributed to deterioration of the organic materials and electrodes caused by various extrinsic and intrinsic factors [3–11]. The extrinsic sources of degradation, including residual water, metallic impurities, and other environmental contaminants can be largely suppressed by improving the source material purity and device encapsulation. Multiple intrinsic phenomena have been identified, including ionic migration [5,6], charge accumulation [7], and crystallization or decomposition of organic materials [8–11]. These may cause the generation of charge traps and nonradiative recombination centers, hampering charge transport and decreasing the luminous efficiency [11]. As a result, OLEDs generally exhibit a gradual rise in the voltage and decrease in the luminance without obvious changes in the device

appearance. All these intrinsic processes can be current stimulated and thermally enhanced [12,13]. Therefore, the lifetime of OLEDs is a sensitive function of the driving current density, and strongly affected by the junction and ambient temperatures.

Typical OLEDs have a complex layer-structure including a light-emitting layer (EML) sandwiched between a hole-transport layer (HTL) and an electron-transport layer (ETL). The structure may also comprise charge-injection layers in contact with the cathode and anode. Many previous lifetime studies of OLED devices have been centered on the aging processes in the EML [3,4]. However, reaction of emitter fragments with the neighboring charge transport materials as well as material decomposition or crystallization in the charge transport layers may also represent important mechanisms of the whole device degradation. Furthermore, these changes can be driven by localized charge and heat at the interfaces associated with the charge transport layers. Understanding the impact of the charge transport layers on the OLED reliability would help to choose desirable charge-transport materials to realize reliable operation of OLEDs at high brightness levels. In this work, electron-only devices (EODs) and OLEDs with four different types of common electron-transport materials (ETMs) are fabricated and subjected to accelerated reliability tests via high constant-current stressing. The practical data obtained from side-by-side comparisons of EODs and OLEDs allows us to directly evaluate the stability of commonly-used ETMs and gain insight into the key material factors which have a pronounced influence on the OLED lifetime.

* Corresponding authors at: School of Electrical and Electronic Engineering, Hubei University of Technology, Wuhan 430068, China.

E-mail addresses: zhouym@mail.hbut.edu.cn (Y.M. Zhou), xacao@mail.hbut.edu.cn (X.A. Cao).

2. Experimental details

EODs and green OLEDs were fabricated on glass substrates with pre-patterned indium-tin-oxide (ITO) whose sheet resistance was $\sim 15 \Omega/\square$. The substrates were first cleaned with acetone, methanol and deionized water, and treated with oxygen plasma for 5 min. They were then transferred to a thermal evaporation system with a base pressure of 1×10^{-7} τ , where the organic and cathode materials were deposited on unheated substrates at a pre-calibrated rate ~ 0.1 nm/s. The EODs were based on a single layer of one of the following four types of ETMs: 3-(Biphenyl-4-yl)-5-(4-*tert*-butylphenyl)-4-phenyl-4H-1,2,4-triazole (TAZ), Tris-(8-hydro-xyquinoline)aluminum (Alq_3), 1,3,5-tris(2-*N*-phenylbenzimidazolyl) benzene (TPBi), and Bathophenanthroline (Bphen). To block hole injection from the ITO anode, 1 nm LiF layer was inserted between the ITO/ETL. The OLED structure consisted of a 50 nm *N,N*-bis-(1-naphthyl)-*N,N'*-diphenyl-1,1'-biphenyl-4,4'-diamine (NPB) HTL, a 30 nm Alq_3 EML, and a 30 nm ETL based on one of the above four ETMs. Finally, a 0.5 nm LiF/100 nm Al cathode was deposited through a shadow mask, which defined the active area of all these devices to be 0.1 cm^2 . Four identical devices were fabricated on each substrate. They were encapsulated with a glass lid in a N_2 -filled glovebox and characterized at room temperature. Fig. 1 illustrates the energy diagram of the OLED structures, in which the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) energy levels of the organic materials as well as the Fermi levels of the electrodes are indicated.

The current density-voltage (J-V) characteristics of the devices were tested using a Keithley 2634B system sourcemeter, and the electroluminescence (EL) spectra of the OLEDs were recorded using an Ocean Optics fiber-optic spectrometer. To measure the luminance of the OLEDs, the devices were placed directly onto the surface of a calibrated silicon photodetector (1 cm in diameter) and all emitted photons from the glass side were captured. To evaluate the device reliability, as-fabricated EODs and OLEDs were stressed at 50 mA/cm^2 , a current density much higher than the industrial standard. The voltage and luminance data were collected periodically during the constant-current stressing process.

3. Results and discussion

Fig. 2 shows the J-V characteristics of the EODs based on different ETMs. The TAZ-based EOD has the highest turn-on voltage ~ 4.7 V, whereas the Bphen-based EOD has the lowest turn-on at 1.8 V. At 20 mA/cm^2 , the EODs based on TAZ, Alq_3 , TPBi and Bphen have an operational voltage of 7.4, 6.3, 5.5, and 2.7 V, respectively. The J-V characteristics reflect the behaviors of electron injection from the cathode and

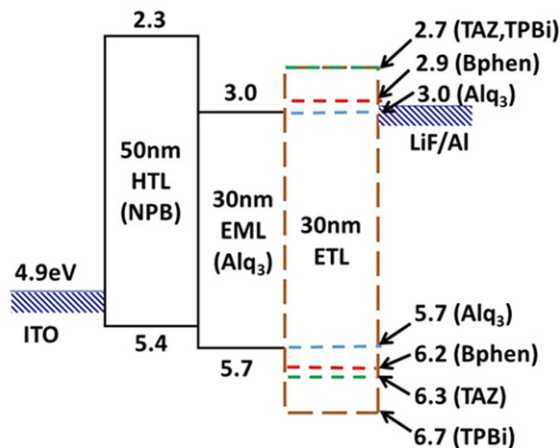


Fig. 1. Energy level diagram of green fluorescent OLEDs with a TAZ, Alq_3 , TPBi, or Bphen ETL.

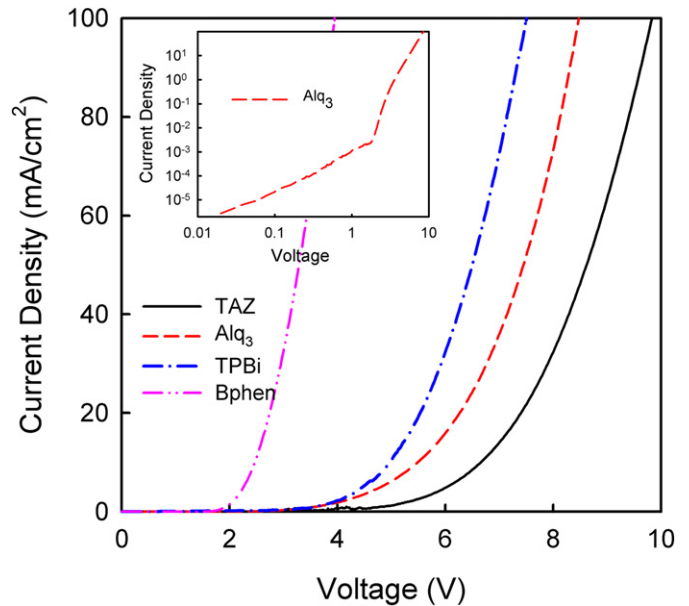


Fig. 2. Forward J-V characteristics of EODs based on four different ETMs: TAZ, Alq_3 , TPBi and Bphen. The inset shows the data of the Alq_3 EOD on a log-log scale.

electron transport inside the ETL. The former is strongly influenced by the energy level alignment at the ETL/cathode interface, and the latter is mainly determined by the electron mobility of the ETM. All these ETMs are energetically misaligned with the Al cathode. However, inserting a thin layer of polar LiF creates an interfacial dipole, which effectively reduces the surface potential of the cathode [14]. As seen in Fig. 1, the LUMO levels of Alq_3 and Bphen align perfectly with the effective Fermi level of LiF/Al, so electron injection in those two EODs is very efficient, whereas in the EODs based on TAZ or TPBi, electrons must overcome a small energy barrier of ~ 0.2 eV at the ETL/cathode interface. Meanwhile, the electron transport behaviors in these ETLs are quite different. The electron mobilities of TAZ, Alq_3 , TPBi and Bphen have been measured using the space-charge limited current method to be 1.57×10^{-6} , 8.05×10^{-6} , 6.53×10^{-5} , and $3.25 \times 10^{-4} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, respectively [15]. As seen, the larger the mobility of the ETM is, the smaller is the EOD voltage. This correlation indicates that the electrical characteristics of the EODs are largely determined by the electron transport process whereas the difference caused by electron injection is small. The EOD based on Bphen has the lowest turn-on and operational voltages because Bphen has the highest electron mobility and is energetically aligned with the cathode. All four EODs have similar mechanisms of electron transport which can be seen more clearly from the same data plotted on the log-log scale. As seen from the representative J-V curve plotted in the inset of Fig. 2, an ohmic region ($J \propto V$) exists at low voltages. As the bias increases, the current becomes trap-charge limited ($J \propto V^n$, $n > 2$), and then moves toward a space-charge limited-current region ($J \propto V^2$) [16].

Fig. 3 illustrates the voltage evolution of the EODs under 50 mA/cm^2 current stressing. The EODs based on Alq_3 or Bphen exhibit a graduate increase in the voltage. After 20 h stressing, their respective voltage increases are 1.5 V and 0.5 V. Over the same time period, the voltage of the TPBi EOD increases by 4.7 V. In a sharp contrast, the voltage of the TAZ EOD rises sharply and reaches the 15 V compliance voltage within 1 h. The voltage increase is mainly due to the formation of charge traps and build-up of space charge in the ETMs, slowing down electron transport. Charge accumulation may also occur at the ETL/cathode interface, leading to localized joule heating and accelerated material degradation. However, this adverse effect should be greatly mitigated by the LiF dipole layer and only play a minor role in the device degradation process [17]. The data in Fig. 3 thus suggest that Alq_3 and Bphen have very good stability, TPBi is less stable, whereas TAZ is the most unstable one. Note

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