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Stressing organic light-emitting diode under constant-brightness driving mode

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ABSTRACT

The degradation of the organic light-emitting diodes (OLEDs) was studied under the constant-brightness driving mode. The time-dependent current exhibits a long period of linear increase followed by an exponential increase before the eventually catastrophic failure featured by a vertical increase. A new lifetime T_{th} is defined as the time for the device to reach the end of the linear increase stage. Similar to the well-known relation between the lifetime and the brightness in the constant-current driving mode, the lifetime and the brightness in the constant-brightness driving mode also fit the formula $L^n \times T_{th}$ = Const., where L is the brightness and n is the acceleration exponent. By examining the current density–voltage–luminance characteristics and the photoluminescence intensity of the devices before and after the stress, it is found that both the reduction of the charge injection efficiency, and the loss of the emissive centers, contribute to the OLEDs' degradation. The extra power supplied to the device to keep the brightness constant, raises the junction temperature, and eventually leads to the catastrophic failure of the devices.

1. Introduction

Since the discovery of the organic light-emitting diode (OLEDs) in 1980s [1], the research and development by both academics and industries has pushed the technology into the applications of the displays and the solid-state lighting [2–4]. As highly efficient materials have been synthesized, and novel device architectures have been proposed and realized, the internal quantum efficiency of OLEDs now can reach 100%, for example, in phosphorescence system [5–7], and in thermally activated delayed fluorescence (TADF) system [8–11]. Though the efficiency of the OLEDs has reach a level which is enough for commercial products, the lifetime is one of the most critical challenges facing OLED's commercialization.

The degradation of the OLEDs can be attributed to two mechanisms: interfacial degradation [12,13], and the degradation of the bulk active layer [14]. Interfacial degradation reduces the injection of the charges from the anode/cathode, which is responsible for the black spot formation [15,16]. At the interface between the hole transporting layer (HTL)/electron transporting layer (ETL), the accumulation of immobile positive charges is found to be linearly correlated with the decreasing luminance efficiency [17]. Further study reveals that the interaction between the hole transport material's positive polarons and the singlet excitons degrades the HTL/ETL interface, contributing to the voltage increase during stressing [18]. The degradation of the bulk active layer causes the decrease of the photoluminescent quantum efficiency (PLQE) due to either the chemical aging [19,20], or the aggregation-induced quenching [21,22].

The most popular, and probably the only approach to study the OLED lifetime, is setting the device at the constant current while monitoring the luminance and the operation voltage. Under the constant-current driving mode, VanSlyke found that the luminance decay rate is directly proportional to the injection current density, which is referred as the coulombic degradation [23]. Parker revealed that the lifetime of the OLEDs depends on both the total charge passing through the device and the temperature [24]. Fery set up the stretched exponential decay model to precisely simulate the whole aging process of OLED under low current density, and showed that the annihilation of the emission centers is the main mechanism responsible for the OLED degradation [25].

In actual applications, the constant current driving mode is the most widely used operation mode in the active matrix OLED

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displays [26–28]. The half-life time $T_{1/2}$, defined as the time for the luminance to decrease to half of its initial value, is universally accepted as the standard to describe the lifetime of OLEDs [29]. However, from the view of the users, a display keeping its brightness constant over the display's lifespan is more desirable. Any brightness drop of the display hurts the users' viewing experience. Even for the application of solid-state lighting, a light source with constant brightness is better than a light source keeping dimming over time. In practice, optical feedback driver circuits have been proposed to compensate the luminance drop in both OLED displays and large-area OLED lighting panels [30–33]. Unfortunately, few people have studied the OLED aging process under the constant-brightness driving mode. Herein, we report the constant-brightness aging process of OLED in our contribution.

During the operation of the OLEDs, we keep the OLED device's brightness constant by using real-time feedback to compensate the current needed for the constant-brightness aging, while monitoring the increase of both the current and the voltage. The time-dependent current and voltage exhibit similar behavior, in which a long period of linear increase is followed by an exponential increase before the eventually catastrophic failure featured by a vertical increase. The device lifetime in constant-brightness driving mode is defined as the time for the OLED to reach the end of its first linear aging stage. The newly defined lifetime T_{th} fits the well-known lifetime-brightness relation found in the constant-current driving mode, i.e. $L^n \times T_{th}$ = Const., where L is the brightness, and n is the acceleration exponent. During the whole aging process, not only does the charge injection efficiency continues to decrease, but also the PLQE of the active layer keeps dropping. Both mechanisms contribute to the OLEDs' degradation under the constant-brightness driving mode.

2. Experimental

2.1. Device fabrication

Green OLED device is fabricated in the configuration of ITO/MeO-TPD: F4-TCNQ (100 nm, 4%)/NPB (15 nm)/TCTA (5 nm)/PT604: Ir(dmppy)$_2$(ppymp) (30 nm, 3%)/TmPyPb (25 nm)/LiF (1 nm)/Al (150 nm). Red OLED device is fabricated in the configuration of ITO/MeO-TPD: F4-TCNQ (100 nm, 4%)/NPB (20 nm)/Bepq$_2$: Ir(MDQ)$_2$acac (40 nm, 5%)/Bepq$_2$ (25 nm)/LiF (1 nm)/Al (200 nm). In both devices, MeO-TPD (N,N,N′,N′-Tetrakis(4-methoxyphenyl)-benzidine) doped with 4 wt% F4-TCNQ (2,3,5,6-Tetrafluoro-7,7,8,8-tetracyano-qinodimethane) is employed as the hole injection layer. NPB (N,N′-Bis(naphthalene-1-yl)-N,N′-bis(phenyl)-benzidine) is used as the hole transport layer. TCTA (Tris(4-carbazoyl-9-ylphenyl)amine) is used as the electron and the exciton blocking layer in the green device. TmPyPb (1,3,5-Tri[(3-pyridyl)-phen-3-yl]benzene) and Bepq$_2$ (Bis(10-hydroxybenzo[h]quinolinato)beryllium) are employed as the electron transport layers in the green device, and the red device, respectively. The host PT604 (unknown molecular structure) doped with 3 wt% Ir(ppy)$_2$(m-mbppy) (Bis(2-phenylpyridinato)[2-methyl-5-(2-pyridinyl)-1,1′-biphenyl] iridium(III)), and the host Bepq$_2$ doped with 5 wt% Ir(MDQ)$_2$acac (Bis(2-methyl-dibenzo[f,h]quinoxaline)(acetylacetonate)iridium(III)) are used as the emission layers of the green and the red devices, respectively. All the organic materials are purchased from Luminescence Technology Corp., and used as received. To fabricate the devices, the ITO glass substrate was pre-patterned by photolithography to give an effective device size of 9 mm^2. Prior to the deposition of the organic layers and the cathode, the ITO substrates were thoroughly cleaned in sequence in ultrasonic bath of acetone, isopropanol, detergent, de-ionized water, isopropanol, and dried in an oven. The entire organic layers were thermally evaporated onto the substrate, followed by the cathode deposition, in a vacuum chamber with a base pressure of 2×10^{-7} Torr. The devices were encapsulated with a glass cap in a nitrogen filled glove-box right after they were taken out from the vacuum chamber.

2.2. Device characterization

The constant-brightness aging was tested in our home-made lifetime test system [34]. A silicon photodiode in front of the device monitored the brightness continuously, and sent the feedback signal to the power supplying unit. The device current was adjusted based on the feedback signal in real time to keep the brightness constant. The current density–voltage–luminance characteristics were obtained using a Keithley 2400 source meter and a silicon photodiode calibrated by a Konica Minolta Chroma Meter CS-2000. The photoluminescence spectra were measured by a Fluoromax-4 spectrofluorometer (HORIBA Scientific). The excitation wavelength was 380 nm. The photoluminescence quantum efficiency (PLQE) was measured by the FluoroSENS Fluorimeter (Gilden Photonics Ltd.) using the same excitation wavelength. Since the actual device with thin emission layer could not yield reliable PLQE data, the samples for PLQE measurement were prepared by evaporating 150 nm PT: Ir(dmppy)$_2$(ppymp) (3%) for the green device and 150 nm Bepq$_2$: Ir(MDQ)$_2$acac (5%) for the red device on the glass slides. The PLQE of the stressed devices are scaled by integrating the absolute PL spectrum.

3. Results and discussion

The green and the red devices were stressed at different brightness. A typical current density (J)–time (t) curve could be divided into three stages as illustrated in Fig. 1. AB segment represents the first aging stage, in which the current density increases slowly and linearly for a long period. BC segment represents the second aging stage, in which the current density rises exponentially over time. CD segment represents the third aging stage, in which the current density vertically rises, featuring the catastrophic failure of the device. As shown in Fig. 2, the voltage rise over time follows a similar trend to the current density. Under the constant-brightness driving mode, the lifetime of the OLED, T_{th}, could be defined as the time it takes for the device to finish the first aging stage. The threshold current density J_{th} is defined as the current density at the end of the first aging stage. Once the current density goes

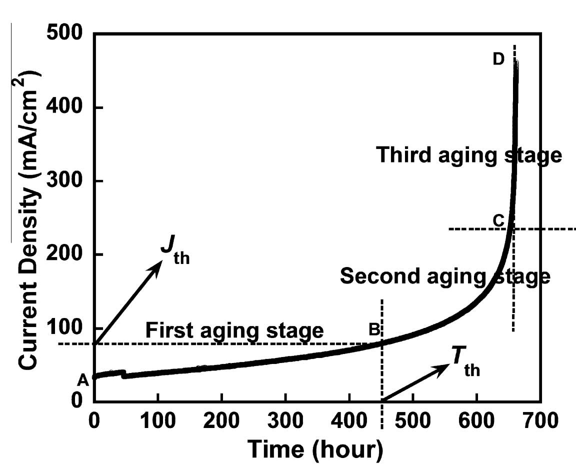

Fig. 1. The typical current density–time curve of the OLED device under the constant-brightness driving mode. T_{th} is the end of the first linear aging stage, which is defined as the lifetime. J_{th} is the threshold current density to start the second aging stage.