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Study on scalable Coulombic degradation for estimating the lifetime of organic light-emitting devices

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Abstract
The luminance decays of organic light-emitting diodes (OLEDs) are investigated with initial luminance of 1000 to 20 000 cd m^{-2} through a scalable Coulombic degradation and a stretched exponential decay. We found that the estimated lifetime by scalable Coulombic degradation deviates from the experimental results when the OLEDs work with high initial luminance. By measuring the temperature of the device during degradation, we found that the higher device temperatures will lead to instabilities of organic materials in devices, which is expected to result in the difference between the experimental results and estimation using the scalable Coulombic degradation.

1. Introduction

Organic light-emitting diodes (OLEDs), which were first reported by Tang and VanSlyke [1] in 1980s, have received considerable attention due to their potential applications in flat panel displays and lighting. The lifetime of OLEDs is crucial for commercial applications. While the first reported multilayer OLEDs had a half-lifetime of only a few hundred hours [1], due to the rapid development of organic materials and device structures, the lifetime of OLEDs has been significantly improved. It was reported that a standard red-emitting device could work for more than one million hours, which is equivalent to over one hundred years of constant use, when it is lighted at an initial luminance of 1000 cd m^{-2} [2]. In fact, the lifetime of 100 years is derived from accelerated ageing tests. A scalable Coulombic degradation model [3–7], as shown in equation (1), has been widely used to estimate the lifetime of OLEDs.

$$L_0^n \times T_{1/2} = \text{constant}, \tag{1}$$

where L_0 is the initial luminance, $T_{1/2}$ is the time for the luminance to decrease to 50% of the initial value (so-called lifetime of OLEDs) and n is the acceleration exponent. In general, the longer lifetime of OLEDs at a low initial luminance such as 100 years was obtained by extrapolating experimentally measured lifetimes at high initial luminances using the scalable Coulombic degradation model (equation (1)). The validity of the scalable Coulombic degradation given by equation (1) is therefore very important for the estimation of lifetime of OLEDs but has not yet been fully studied. In a previous paper [8], it was found that the experimentally measured lifetime deviated from the theoretically estimated lifetime by the scalable Coulombic degradation (equation (1)) at high operating currents (high

initial luminances). The authors indicated that the higher initial luminance might introduce other new degradation mechanisms, such as acceleration decay of luminance caused by self-heating, which lead to the invalid use of the scalable Coulombic degradation (equation (1)). However, they did not provide pertinent evidence because that paper did not focus on the issue, which needs to be further investigated.

In this paper, we measured the luminance degradation and operating temperatures of OLEDs based on tris(8-hydroxyquinoline)aluminium (Alq_3) at different initial luminances. It was found that the experimentally measured lifetimes deviated from those estimated by the scalable Coulombic degradation when the OLEDs worked with a higher initial luminance, because the device temperatures exceeded the glass transition temperature (T_g) of the organic material at the given initial luminance, leading to morphological changes in the organic films. Therefore, we propose that the correlation of the validity of the scalable Coulombic degradation and initial luminance is related to the device temperature.

2. Experiments

The devices used in this work were fabricated by standard vacuum deposition onto indium tin oxide (ITO)-coated glass substrates at a base pressure of about 1×10^{-3} Pa. The device structure was ITO/hole-transporting layer (HTL) (60 nm)/Alq_3(60 nm)/LiF(0.5 nm)/Al(100 nm). Two materials with different T_g were used as the HTL. The HTL of device A was N,N'-di(naphthalene-1-yl)-N,N'-diphenylbenzidine (NPB) with T_g 95 °C, and that of device B was N,N'-diphenyl-N,N'-bis(3-methylphenyl)-[1,1′-biphenyl]-4,4′-diamine (TPD) with T_g 60 °C. These devices were encapsulated in a dry nitrogen glove box. The measurements of lifetimes of the devices were made using an adjustable constant direct current power source at room temperature.

3. Results and discussion

Figure 1 shows the luminance decay curves of devices A and B at different initial luminances, together with the fits using a stretched exponential decay (SED) [7–10], as shown in the following equation:

$$L(t) = L_0 \exp\left[-\left(\frac{t}{\tau}\right)^{\beta}\right], \qquad (2)$$

where $L(t)$ is the relative luminance, τ is the delay time and β is a dispersion factor. As seen in figure 1(a), device A showed typical luminance decay curves of OLEDs and the luminance decayed faster at higher initial luminances (higher current densities). The lifetime at an initial luminance of 19 800 cd m^{-2} was 0.8 h but it was extended to more than 1000 h at an initial luminance of 1085 cd m^{-2}. Device B also showed similar decay curves even though the lifetime of the device was different, which is shown in figure 1(b). Figure 1 also shows that the SED can be used to fit all the luminance decay curves with a high degree of accuracy.

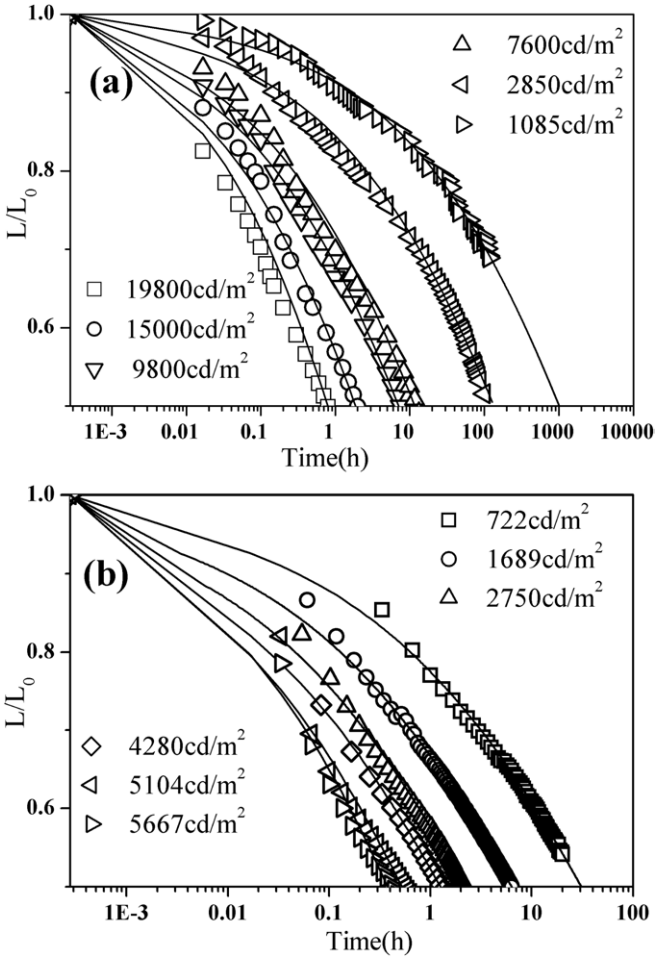

Figure 1. Luminance decay curves of devices at various initial luminances: (a) device A; (b) device B. Black lines are the fitting lines using the SED.

As the estimated results of the scalable Coulombic degradation given by equation (1), the logarithm of L_0 and $T_{1/2}$, are linear, shown by the solid lines in figure 2(a), the degradation event resulting from charge injection is cumulative and irreversible [11]. However, the measured lifetimes (symbols in figure 2(a)) of the devices deviated from the estimation using equation (1) when the devices worked with high initial luminances (i.e. device A: 15 000 cd m^{-2}, 19 800 cd m^{-2}; device B: 5104 cd m^{-2}, 5667 cd m^{-2}). Figure 2(b) shows the fitting parameter β of the SED at different initial luminances. The coefficient β is almost a constant at low initial luminances, and it increases at high initial luminances, just at which the measured lifetimes deviated from those estimated by equation (1). The results reveal that the scalable Coulombic degradation (equation (1)) is valid under the condition that the coefficient β of SED (equation (2)) is a constant at various initial luminances. In a previous paper [7], it was reported that the coefficient β of SED (equation (2)) and the acceleration exponent n of the scalable Coulombic degradation (equation (1)) depend on the materials as well as on the device architecture. In addition, the coefficient β has been demonstrated to be an accurate indicator of glassy properties [12]. Increased β indicates that there may be glassy properties (crystallization) in organic materials

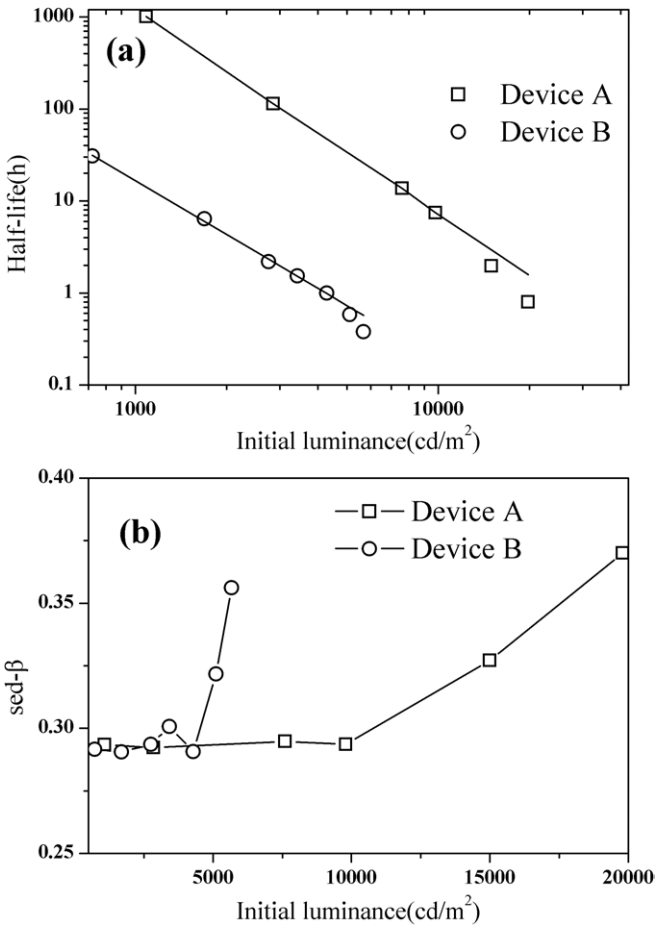

Figure 2. (*a*) Measured lifetimes as a function of initial luminance for two devices; solid lines correspond to the estimated lifetime by equation (1). (*b*) Fitting parameters β of the SED versus initial luminance of devices A and B.

due to high device temperatures caused by self-heating, which is crucial at high current densities (high initial luminances). The self-heating is mainly due to the Joule heat generated by current injection and transport when the thermal conductivity of the substrate and organic films is poor. The Joule heat accumulates in active areas, leading to a substantial increase in the device temperature. Serious self-heating may induce thermal expansion, interlayer diffusion and crystallization of the organic layers, accelerating the degradation of devices. In figure 2, we also found that device B demonstrated similar behaviours as device A with a lower initial luminance, which may be caused by the lower T_g of TPD. Thus, it is necessary to measure the temperature of the OLEDs during degradation.

As the temperature of the device during operation [13, 14], the operating cathode temperature of the device was measured by thermocouples on the unencapsulated cathode surface close to the contacts at room temperature under ambient atmosphere. A thermal grease was used between the cathode and thermocouples to ensure efficient contacts. Figure 3 shows the relation of device temperature and working time at different initial luminances for device A. Device A achieved thermal equilibrium for a short time, and the equilibrium time varied inversely with the initial luminance.

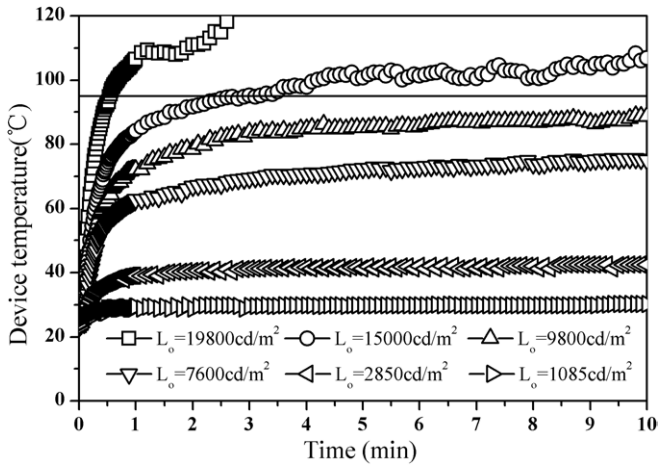

Figure 3. Correlation between device temperature and time at various initial luminances for device A. The solid line indicates T_g of NPB (95 °C).

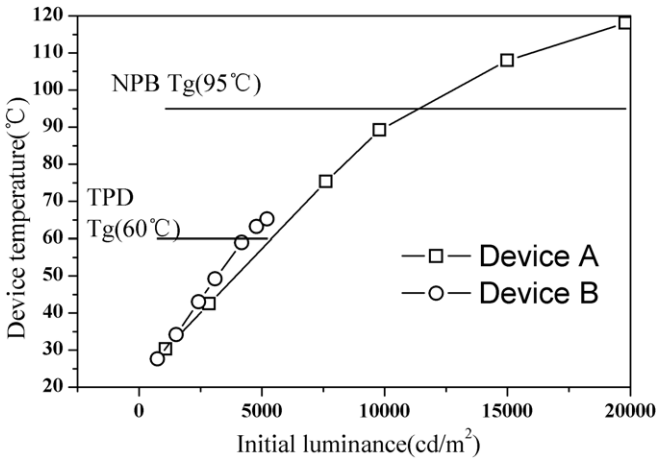

Figure 4. Device temperature versus initial luminances after the devices have worked for 10 min.

Figure 4 shows the characteristic of device temperature versus initial luminance after the devices worked for 10 min. The device temperatures with lower initial luminances (device A: 1085, 2850, 7600 and 9800 cd m^{-2}; device B: 722, 1689, 2750 and 4280 cd m^{-2}) were always below the T_g of the HTL (device A: NPB 95 °C; device B: TPD 60 °C). At high initial luminances such as 15 000 and 19 800 cd m^{-2}, however, as shown in figure 3, the device temperature exceeded the T_g of NPB and increased again after a short-term thermal equilibrium. This can be explained by the fact that crystallization and morphological changes occur in organic films when they are at high temperatures above T_g of the organic materials. Figure 5 shows the optical microscopic images of the organic films at different temperatures. It clearly indicates that the morphological stability of organic films is retained when the device temperature is below the T_g of the organic materials, whereas device temperatures above the T_g of the organic materials induce morphological changes in the organic films. As a result, the high device temperature caused by the high current density (high initial luminance) may introduce crystallization or other morphological changes

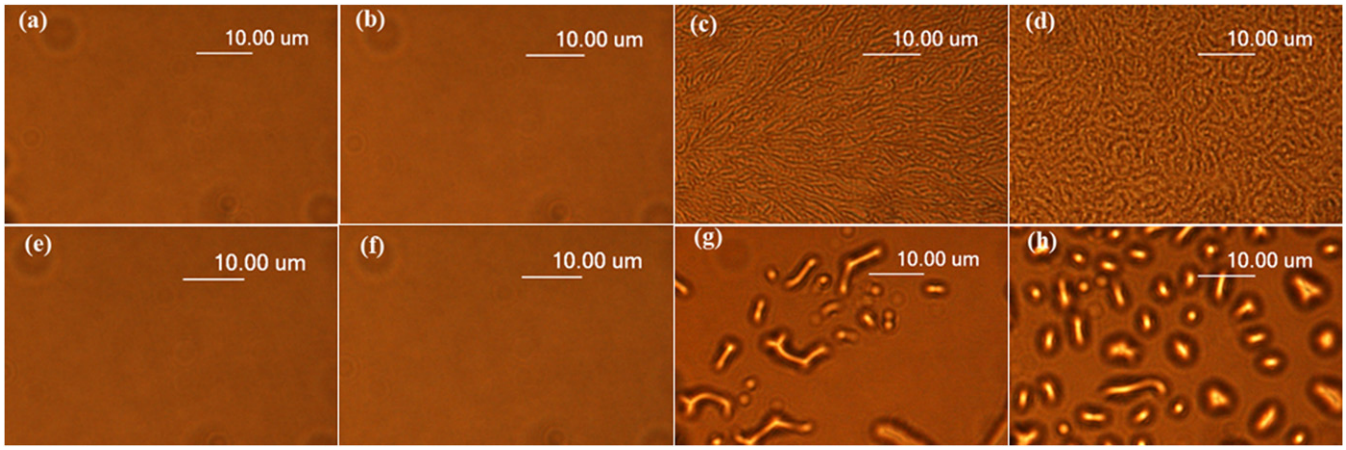

Figure 5. Optical microscopic images of organic films under different temperatures. (*a*)–(*d*) correspond to NPB film under 0 °C, 89 °C, 108 °C and 118 °C annealing, respectively; (*e*)–(*h*) correspond to TPD film under 0 °C, 58 °C, 64 °C and 66 °C annealing, respectively. (This figure is in colour only in the electronic version)

in the organic material, leading to the fracture of the organic film, accelerating the luminance decay of the devices, and thus resulting in the invalid use of the scalable Coulombic degradation as equation (1).

4. Conclusion

In conclusion, we investigated the correlation of the validity of the scalable Coulombic degradation (equation (1)) and initial luminance in OLEDs with TPD and NPB as the hole-transporting layer. We found that the validity of the scalable Coulombic degradation of the OLEDs with various initial luminances depends on the device temperature. A high initial luminance leads to a higher device temperature (exceeding the T_g of the organic materials), and results in the instability of the organic materials (crystallization or other morphological changes) in devices, which is expected to invalidate the use of the scalable Coulombic degradation for lifetime estimation. Our study will be helpful in accurately estimating the device lifetime of OLEDs at different initial luminances.

Acknowledgments

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