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Lifetime and Electric Characteristics of Encapsulated Organic Light-Emitting Devices

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Organic light-emitting devices (OLEDs) with tris-(8-hydroxyquinoline)aluminum (Alq₃), which is typically a green emitter, have been studied by many researchers because of their potential for facilitating the development of flexible electronics. In this paper, the luminance characteristics, current, capacitance, and dispersion factor for degraded OLEDs, which were obtained by applying various bias currents ($0.5 \leq I_{\text{Bias}} \leq 9$ mA), are studied. The current dependences of lifetime were divided at approximately 2 mA, and they represented nearly linear behaviors but had different slopes in a logarithmic plot of lifetime versus ubias current. With OLEDs emitting, the anomaly of capacitance, as shown in the capacitance–voltage curve, occurred because of two factors, polarization in the bulk of organic materials and the interface between the metal and organic layers. In decayed OLEDs that had lower bias currents of less than 2 mA, it was found that the degradation of luminance was related to both the decrease in polarization and to the lowering of the injection barrier. [DOI: 10.1143/JJAP.45.7766]

KEYWORDS: organic light-emitting devices (OLEDs), luminance, lifetime, encapsulation, spectrum, capacitance, barrier height

1. Introduction

Recently, organic electronic devices such as transistors and electroluminescence (EL) devices have attracted considerable attention owing to their potential in flexible electronics.^{1–5} In particular, the characteristics of organic light-emitting devices (OLEDs) have made such devices feasible for applications requiring large-area coverage, low driving voltage, and low overall cost.^{6,7} However, Tang and VanSlyke have reported a fatal shortcoming in these devices: OLEDs that use well-known EL materials such as tris-(8-hydroxyquinoline)aluminum (Alq₃) have shown a decrease of up to 50% in their initial EL intensities after 100 h.¹ Moreover, Hamada *et al.* have found that operating OLEDs in air resulted in a 99% loss of EL intensity in as short as 150 min.⁸ This very short device lifetime is a very serious problem that must be overcome for OLEDs to be used to their full potential under conditions where moisture and oxygen are present.^{9,10} In addition, the lifetime of OLEDs can also be negatively affected by other factors, such as the OLED structure, substrate treatment (especially for substrate-coated indium–tin oxide, ITO), and doping in the hole transport layer or emission layer.¹¹

In this paper, we report on the lifetime properties of OLEDs encapsulated by a glass cap. This encapsulation method has been conventionally used for the passivation of OLEDs based on glass substrates and is composed of a glass can containing desiccants.¹² The glass cap blocks the entrance against air effectively but it cannot prevent the progress of the internal degradation of OLEDs induced by a high DC current. The decay of luminance and the change in capacitance for encapsulated OLEDs were investigated in terms of their lifetime and electrical properties, respectively.

2. Experimental Methods

Small molecular OLEDs were fabricated by vacuum deposition and their configuration was as follows: ITO (150 nm)/N,N'-bis-(1-naphthyl)-N,N'-diphenyl-1,1'-biphenyl-4,4'-diamine (NPB, 50 nm)/Alq₃ (60 nm)/LiF (1 nm)/Al (100 nm) on a glass substrate. ITO-coated glass substrates with a surface resistance of 10–20 Ω/sq were cleaned in

ultrasonic baths of acetone and methanol. The NPB and Alq₃ were the hole transporting layer and the emitting layer, respectively. The cathode was the bilayer of LiF and aluminum. The layers of NPB and Alq₃ as well as the metallic cathode were all deposited by thermal evaporation (below 10^{-3} Pa). The area of the test pixel was approximately 4 mm². The devices were encapsulated by a conventional method using a glass can and desiccants. Glass encapsulation was carried out in a nitrogen glove box. To evaluate the effect of glass encapsulation, OLEDs without encapsulation and with the glass cap were prepared and the degradations of their emission efficiency and luminescence were examined. The electrical, capacitance, and lifetime properties were measured at room temperature using a source measure unit (Keithley 238), an impedance analyzer (HP4195), and photometers (Minolta LS-100 and CS-1000), respectively. Emission images of the OLEDs were taken throughout the lifetime measurement using a black-and-white charge-coupled device (CCD) camera. All the temperature-dependent data were obtained by a low-temperature system using a cryostat chamber and a temperature controller (Lake Shore 340).

3. Results and Discussion

Figure 1(a) shows the decay curves of normalized luminance for 4 mm²-encapsulated OLEDs on a glass substrate. The devices were tested at room temperature and ambient pressure under the condition of DC bias currents. The magnitude of the bias current was between 0.5 and 9 mA. Initial luminances for all devices were between 6.8×10^2 and 1.25×10^4 cd/m², as shown in the inset of Fig. 1(a). The emitting efficiency was approximately 5.5 cd/A. The operational lifetime was measured at 5 min intervals from the initial luminance to a degradation of 60%. As the bias current increased, the lifetime at $x\%$ ($x < 100$) of the starting value decreased. This result indicates that the lifetime of OLEDs might depend on not only the effects of moisture and oxygen but also on the magnitude of the bias currents. For comparison, the lifetimes of devices with and without glass cap encapsulation were investigated under the same bias current of 0.5 mA. The devices without encapsu-

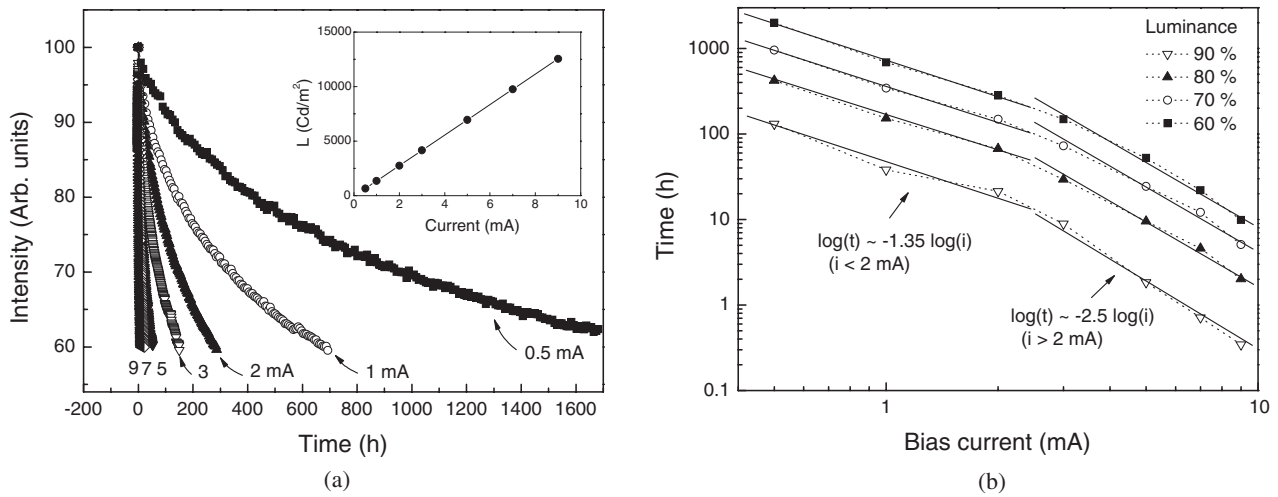


Fig. 1. (a) Decay curves of normalized luminance for different bias currents and (b) logarithmic plot of lifetime versus bias current in encapsulated OLEDs. The inset of Fig. 1(a) shows the luminance–current (L – I) characteristics of fabricated OLEDs.

lation had a very short lifetime of approximately 75 h and their images exhibited many dark spots after the lifetime measurement at atmospheric pressure. Figure 1(b) shows the logarithmic plot of lifetime versus bias current for the encapsulated OLEDs, which was obtained from the data of luminance decay shown in Fig. 1(a). The 60, 70, 80, and 90% points of the initial luminance were introduced into this figure. The current dependence of lifetime was changed to approximately 2 mA. Specifically, the current dependence of lifetime in the lower bias current region of less than 2 mA ($I_{\text{Bias}} \leq 2$ mA) represented a nearly linear behavior; that is, $\log t \propto n \log i$, where t is the lifetime, n the proportional factor, and i the bias current. The analogous tendency was also observed in the region higher than 2 mA ($I_{\text{Bias}} > 2$ mA). The factors n were approximately -1.35 and -2.5 in the regions of $I_{\text{Bias}} \leq 2$ mA and $I_{\text{Bias}} > 2$ mA, respectively. The lifetimes measured in the region of $I_{\text{Bias}} \leq 2$ mA were smaller than the values extrapolated from the linear fitting of data in the region of $I_{\text{Bias}} > 2$ mA.

Figures 2(a) and 2(b) show the black-and-white CCD images of encapsulated OLEDs operating at 1 and 7 mA, respectively. In the range of high luminance above 120 cd/m², a CCD image was observed through an optically neutral filter. The photographs of 40%-decayed OLEDs under the constant currents of 1 and 7 mA are shown in Figs. 2(c) and 2(d), respectively. In the decayed images [Figs. 2(c) and 2(d)], the deviation of brightness was less than 5%, and no dark spots or edge shrinkages appeared. It at first seemed possible that encapsulation with the glass cap effectively protected the devices from moisture and oxygen and would prolong the lifetime of the device. However, the lifetime of the OLEDs is also greatly affected by the bias current, so the lifetime at the I_{Bias} of 7 mA was approximately 30 times shorter than at the I_{Bias} of 1 mA.

The capacitance–voltage (C – V) measurements were performed for the diode structure of OLEDs by applying a small AC amplitude of 10 mV and a frequency of 10 kHz. The sweep rate was 1.5 V/s. The area of the capacitor and the total thickness of organic layers are 4 mm² and 110 nm, respectively. The C – V and dispersion factor–voltage ($\tan \delta$ – V) characteristics for the different luminance decays

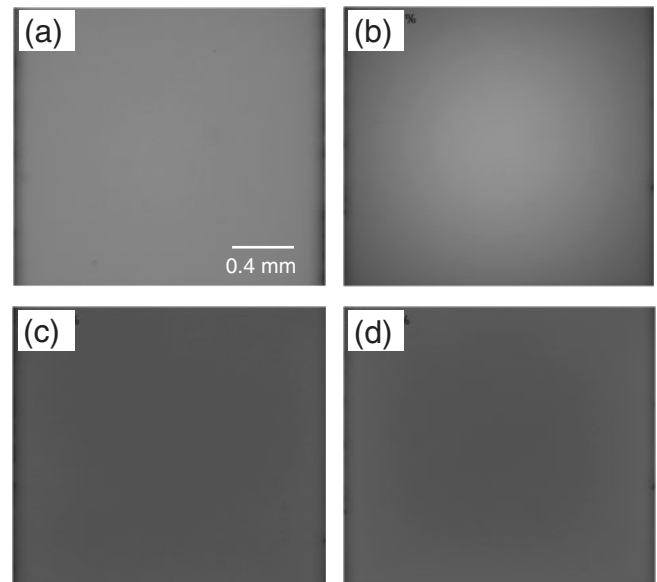
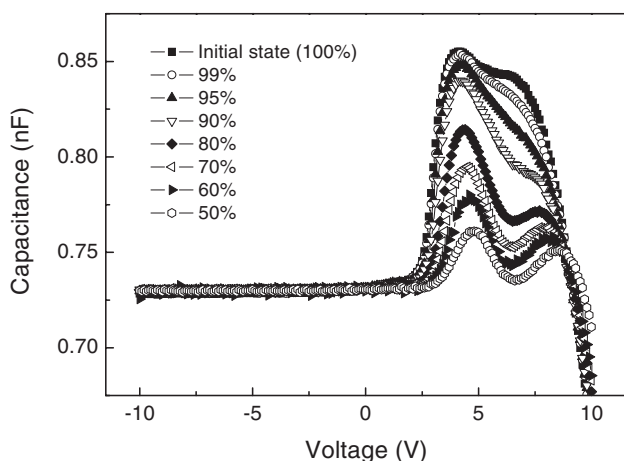


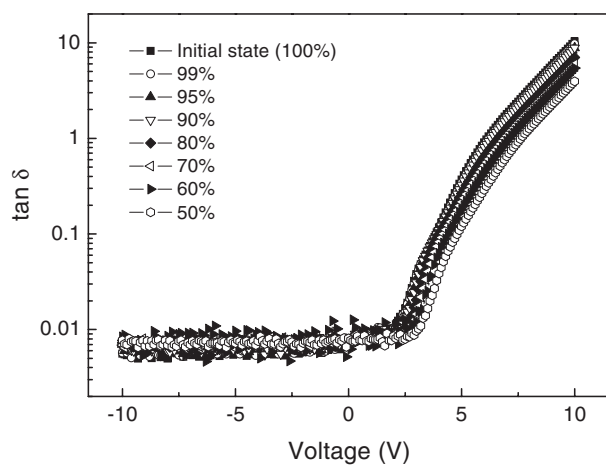
Fig. 2. Luminance images observed using black-and-white CCD camera. (a) and (b) are the initial images of OLEDs operating at 1 and 7 mA, respectively. (c) and (d) are the images of 40%-decayed OLEDs under the constant currents of 1 and 7 mA, respectively.

between 0 and 50% are shown in Figs. 3(a) and 3(b), respectively. The bias current was 1 mA. A diode behavior was observed featuring a steep increase in $\tan \delta$ in the forward direction. This indicates the efficient charge-carrier injection and transport above a certain voltage called a turn-on voltage. Above the turn-on voltage, anomalies of capacitance occurred; these might be related to the formation and alignment of dipole moments in the bulk of organic materials and the interface between the metal and organic layers.¹³⁾ Figure 3(c) shows the voltage dependence of the accumulated charge (ΔQ) for the different luminance decays. The ΔQ is obtained by integrating the anomaly of capacitance as

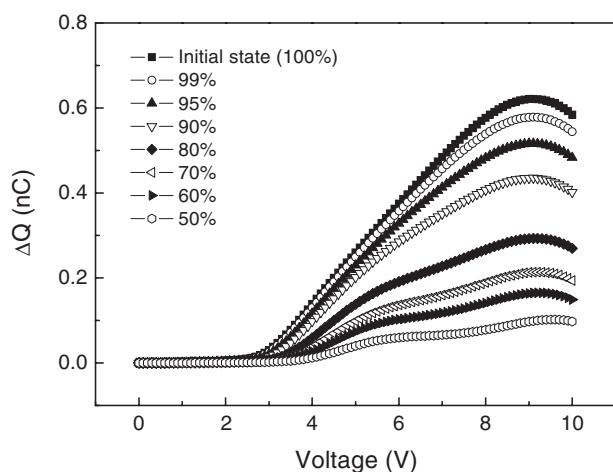
$$\Delta Q = \int_0^V (C(V) - C_0) dV, \quad (1)$$



(a)



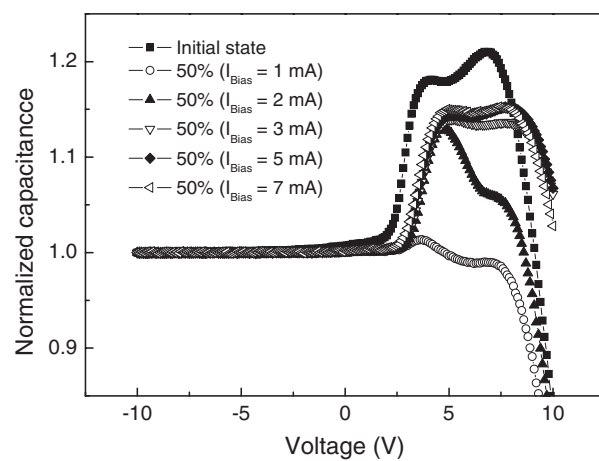
(b)



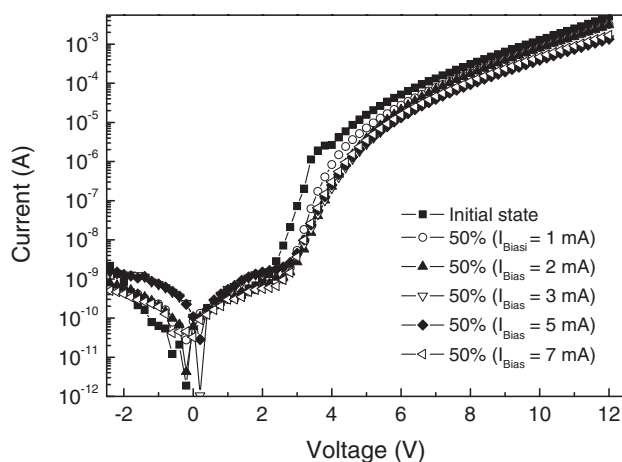
(c)

Fig. 3. (a) C - V , (b) $\tan \delta$ - V , and (c) ΔQ - V characteristics for different luminance decays of encapsulated OLEDs. The bias current was 1 mA.

where $C(V)$ is the capacitance at a voltage V and C_0 is the capacitance at $V = 0$ V. The charge density in the initial state was calculated to be approximately 10^{11} cm^{-2} , which was less than the surface charge densities (10^{12} – 10^{13} cm^{-2}) between Alq_3 and metals reported by Lee *et al.*¹⁴⁾ With decreasing luminance, the threshold voltage shifted toward a higher voltage and the maximum of ΔQ decreased from 0.62 to 0.09 nC, similar to the results reported by Kondakov



(a)



(b)

Fig. 4. (a) C - V and (b) I - V characteristics before and after lifetime measurement for different bias currents. The level of bias currents was from 1 to 7 mA.

*et al.*¹⁵⁾ They found that the decreasing luminance efficiency was linearly correlated with an accumulation of immobile positive charge at the hole-transport layer (HTL)/electron-transport layer (ETL) interface, and that the magnitude of the charge was comparable to the total charge at that interface when a new device was operated.¹⁵⁾

Figure 4(a) shows the C - V characteristics of OLEDs before and after the lifetime measurement for the different bias currents. The level of bias currents ranged from 1 to 7 mA and the capacitance of the devices was measured at half of the initial luminance. In the C - V curves, the anomalies of capacitance for 3-, 5-, and 7-mA-biased samples were similar to one another, but the anomaly for the 1-mA-biased sample was much smaller than for the others. The shrinkage of anomaly indicated a decrease in polarization generated in the luminous state of OLEDs. Figure 4(b) shows a plot of $\log I$ versus V for the same devices. No significant difference in the I - V curves for new or degraded OLEDs was found, except in the reduction of currents.

The normalized EL spectra and the peak positions of OLEDs after the lifetime measurement for the different bias current are shown in Figs. 5(a) and 5(b). The bias currents were between 1 and 9 mA, and the spectra were measured in the wavelength range from 380 to 780 nm. The spectra of the

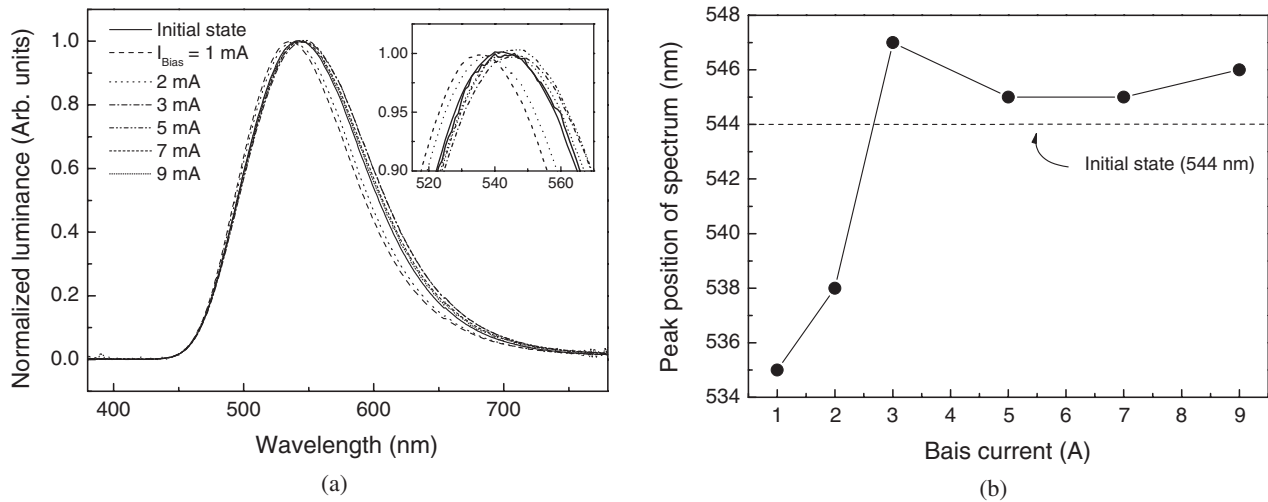


Fig. 5. (a) Spectrum curves and (b) peak positions for new and 50%-decayed OLEDs. The bias currents were between 1 and 9 mA.

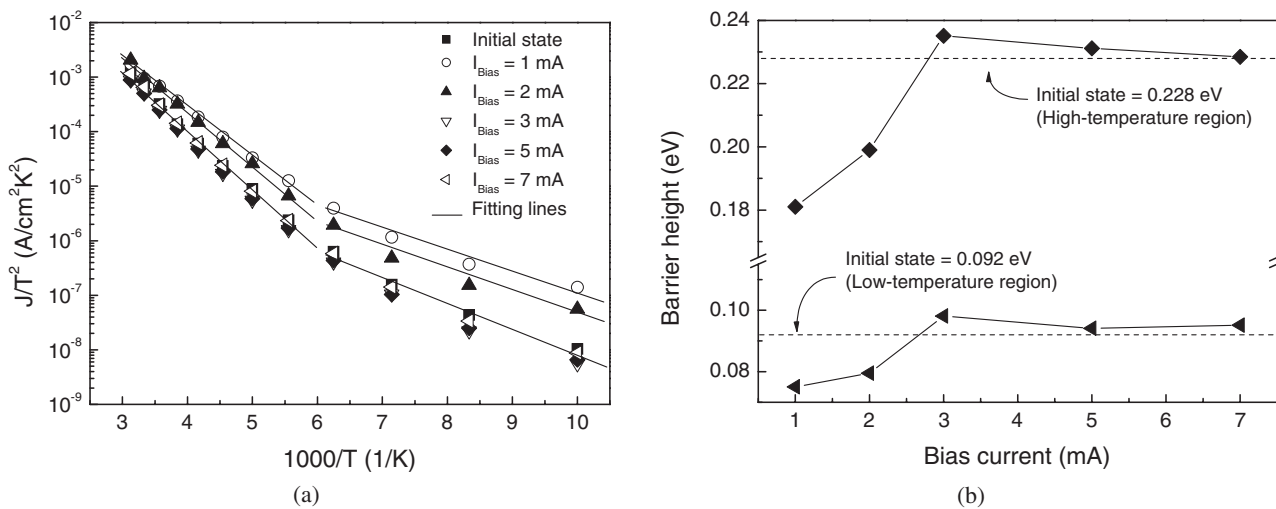


Fig. 6. (a) Temperature-dependent current curves and (b) barrier heights for new and 50%-decayed OLEDs. The level of bias currents was from 1 to 7 mA.

OLEDs at half of the initial luminance were compared with the spectrum of a new device. In the initial state, the peak of the spectrum corresponding to Alq₃ was clearly observed at 544 nm. By decreasing the bias current, the peak position in the degraded devices gradually shifted to a short wavelength.

To study the characteristics of the injection barrier for new and 50%-decayed OLEDs, the temperature dependence of static currents was investigated in the region of low temperatures. With an external voltage of 15 V, the static current was measured at a constant temperature. Figure 6(a) shows a plot of J/T^2 versus $1000/T$, where J is the current density and T is the temperature. Two distinguishable tendencies of the static current were evident in the range of 100 to 330 K. The height of the injection barrier, commonly called a Schottky barrier, in the diode structure of the OLEDs was calculated using the field-enhanced Schottky emission model.^{16–18} As given in Fig. 6(b), the barrier heights at higher and lower temperatures in the initial state of OLEDs were 0.228 eV ($T > 167$ K) and 0.092 eV ($T < 167$ K), respectively. The presence of a barrier to electron injection in the Alq₃/cathode interface at room

temperature has been studied by Barth *et al.*¹⁸) and Hung *et al.*¹⁹) The height in $T > 167$ K agreed with the reported values (0.2–0.3 eV).^{18,19}) As the bias current decreased in the region of $I_{\text{Bias}} \leq 2$ mA, the barrier heights were reduced to 0.181 eV and 0.075 eV for $T > 167$ K and $T < 167$ K, respectively. The lowering of the barrier height caused reduced polarization in the interface between the metal and organic layers. It might also have affected the internal field and the recombination zone of electrons and holes, thus generating weak EL and, moreover, accelerating the decay of luminance. This result coincided with other findings, such as the C - V curves and spectrum data for the different bias currents. Finally, we found that the state of the degraded OLEDs under the condition of $I_{\text{Bias}} \leq 2$ mA, in contrast to that for $I_{\text{Bias}} > 2$ mA, affected the decrease in polarization, the blue shift of the spectrum (or the change in the recombination zone), and the lowering of barrier height.

4. Conclusions

In this study, we investigated the fabrication of encapsulated OLEDs on a glass substrate and their electric

characteristics by C - V , I - V , and spectral measurements. The operational lifetime of OLEDs was measured from the initial luminance to a 50% degradation at a constant current. The luminance decreased after the lifetime measurement in accordance with the magnitude of the bias currents. Above the turn-on voltage, the anomaly of capacitance in a C - V curve was observed by the formation of polarization in the bulk and in the organic/metal interface. Phenomena such as the diminution in polarization, the shift of the spectrum, and the lowering of barrier height were discovered in the degraded OLEDs under bias currents of $I_{\text{Bias}} \leq 2$ mA.

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- 1) C. W. Tang and S. A. VanSlyke: Appl. Phys. Lett. **51** (1987) 913.
- 2) K. Nagayama, T. Yahagi, H. Nakada, T. Tohma, T. Watanabe, K. Yoshida and S. Miyaguchi: Jpn. J. Appl. Phys. **36** (1997) L1555.
- 3) G. Parthasarathy, P. E. Burrows, V. Khalfin, G. G. Kozlov and S. R. Forrest: Appl. Phys. Lett. **72** (1998) 2138.
- 4) C. D. Dimitrakopoulos, S. Purushothaman, J. Kymissis, A. Callegari and J. M. Shaw: Science **283** (1999) 822.
- 5) Y. S. Yang, S. H. Kim, J. Lee, H. Y. Chu, L. Do, H. Lee, J. Oh and T. Zyung: Appl. Phys. Lett. **80** (2002) 1595.
- 6) G. Gu, P. E. Burrows, S. Venkatesh, S. R. Forrest and M. E. Thompson: Opt. Lett. **22** (1997) 172.
- 7) M. S. Weaver, J. J. Brown, R. H. Hewitt, S. Y. Mao, L. A. Michalski, T. Ngo, K. Rajan, M. A. Rothman, J. A. Silvermail, W. E. Bennet, C. Bonham, P. E. Burrows, G. L. Graff, M. E. Gross, M. Hall, E. Mast and P. M. Martin: Inf. Disp. **17** (2001) 26.
- 8) Y. Hamada, C. Adachi, T. Tsutsui and S. Saito: Jpn. J. Appl. Phys. **31** (1992) 1812.
- 9) H. Aziz, Z. D. Popovic, C. P. Tripp, N. Hu, A. Hor and G. Xu: Appl. Phys. Lett. **72** (1998) 2642.
- 10) K. K. Lin, S. J. Chua and S. F. Lim: J. Appl. Phys. **90** (2001) 976.
- 11) H. Aziz, Z. D. Popovic, N. Hu, A. Hor and G. Xu: Science **283** (1999) 1900.
- 12) P. E. Burrows, V. Bulovic, S. R. Forrest, L. S. Sapochak, D. M. McCarty and M. E. Thompson: Appl. Phys. Lett. **65** (1994) 2922.
- 13) G. G. Malliaras, Y. Shen, D. H. Dunlap, H. Murata and Z. H. Kafafi: Appl. Phys. Lett. **79** (2001) 2582.
- 14) S. T. Lee, X. Y. Hou, M. G. Mason and C. W. Tang: Appl. Phys. Lett. **72** (1998) 1593.
- 15) D. Y. Kondakov, J. R. Sandifer, C. W. Tang and R. H. Young: J. Appl. Phys. **93** (2003) 1108.
- 16) S. M. Sze: *Physics of Semiconductor Devices* (Wiley, New York, 1969) pp. 492–501.
- 17) Y. S. Yang, S. J. Lee, S. H. Kim, B. G. Chae and M. S. Jang: J. Appl. Phys. **84** (1998) 5005.
- 18) S. Barth, U. Wolf, H. Bässler, P. Müller, H. Riel, H. Vestweber, P. F. Seidler and W. Rieß: Phys. Rev. B **60** (1999) 8791.
- 19) L. S. Hung, L. S. Liao, C. S. Lee and S. T. Lee: J. Appl. Phys. **86** (1999) 4607.