Delayed Electroluminescence in Organic Light Emitting Diodes

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Abstract

The present paper reports an analytical approach to the delay time of organic light-emitting diodes (OLEDs). The delay time of OLEDs decreases with decreasing value of the threshold voltage or work function of the cathode, increases with the device area and thickness of the electron or hole transporting layer and decreases with increasing value of the applied electric field. Whereas $t_{inj}$ depends linearly on the device area, the value of $t_{run}$ for $\tau << t_d$ and $\tau << t_{inj}$, and $\tau >> t_d$ and $\tau >> t_{inj}$, becomes independent of the device area. The values of threshold voltage, $(V_{th})$, time constant $(\tau)$ of OLED, zero-field charge carrier mobility $(\gamma)$, and the electric field coefficient $(\mu_0)$ to the mobility can be determined from the measurement of the dependence of delay time on the strength of applied electric field.

Introduction

The response time of organic light emitting diodes (OLEDs) when addressed by a step voltage is an essential criterion for their application in optoelectronic displays. It has been found that there is a time lag called delay time, $t_{del}$, between the time of application of a voltage pulse to OLED and the onset of EL emission. Considering the importance in performance of OLEDs, significant attention has been paid to the investigation of time delay of OLEDs [1 - 6]. Several workers have reported that the delay time to be dependent on device area [2 - 5] and many workers have reported it to be independent of device area [1, 6]. The present paper reports a theoretical model for the injection and transport of charge carriers in pulsed organic light emitting diodes.
Theoretical Approach to the Delay Time of OLEDs

Basically, the EL delay time should depend on two components: (i) the charge injection time, $t_{inj}$, and (ii) the charge running delay, $t_{run}$. Thus, the EL delay time may be expressed as

$$ t_d = t_{inj} + t_{run} \tag{1} $$

Considering the equivalent circuit of the OLED device to be a series connected circuit of a resistor and a capacitor (consisting of the anode and cathode of the OLED), the voltage difference at a time (t) can be expressed as an exponential growth function in the following way

$$ E = E_0 \left[ 1 - \exp \left( \frac{-t}{\tau} \right) \right] \tag{2} $$

where $E_0$ is the electric field strength of the applied pulse voltage, and $\tau$ is the time constant of the OLED device with the drive system.

Using eq. (2), the injection delay can be expressed as

$$ t_{inj} = -\tau ln \left( 1 - \frac{E_{th}}{E_0} \right) = \tau_i S \frac{E_{th}}{E_0} = (R + R_0) C_i S \frac{E_{th}}{E_0} \quad \text{(for } E_{th} \ll E_0) \tag{3} $$

where, $E_{th}$ is the threshold electric strength for charge injection. $\tau_i$ is the specific time constant per unit area of the time constant $\tau$, and $S$ is the cross-sectional area of OLED, and $R$, $R_0$, and $C_i$ represent the series resistance of OLEDs mainly caused by ITO (Indium-Tin-Oxide), the additional series resistance of the drive system and the specific capacitance per unit area of the OLED, respectively.

As such, the transit time, in which the carriers will travel a distance, $d$, which is thickness of electron or hole transporting layer, can be obtained from the following equation

$$ d = \int_{t_{inj}}^{t_{inj}+t_{run}} \mu E \, dt \tag{4} $$

where $E$ is the strength of electric field at any time $t$, and $\mu$ is the mobility of the charge carriers.

In organic semiconductors, the charge mobility is electric-field dependent and it can be expressed as

$$ \mu = \mu_0 \exp(\gamma \sqrt{E}) \tag{5} $$

where, $\mu_0$ is the zero-field mobility and $\gamma$ is the electric-field coefficient to the mobility.

Using eqs. (2), (4), and (5), and neglecting higher orders of $t$ and $\tau$, we get...
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\[
d = \mu_0 E_0 \left[ t_{\text{inj}} + t_{\text{run}} + \tau \exp \left( -\frac{(t_{\text{inj}} + t_{\text{run}})}{\tau} \right) \exp \left( \sqrt{\frac{E_0}{\tau}} \left( 1 - \exp \left( -\frac{(t_{\text{inj}} + t_{\text{run}})}{\tau} \right) \right) \right) \right]
\]

\[-\mu_0 E_0 \left[ t_{\text{inj}} + \tau \exp \left( -\frac{t_{\text{inj}}}{\tau} \right) \exp \left( \sqrt{\frac{E_0}{\tau}} \left( 1 - \exp \left( -\frac{t_{\text{inj}}}{\tau} \right) \right) \right) \right] \tag{6}\]

Now, we consider the following three cases:

**Case I :** \( \tau \ll \left(t_{\text{inj}} + t_{\text{run}}\right) \) or \( \tau \ll t_{\text{inj}} \)

In low field regime and for low value of the time constant \( \tau \) of OLED, \( \tau \ll \left(t_{\text{inj}} + t_{\text{run}}\right) \) or \( t_d \) and, \( \tau \ll t_{\text{inj}} \), and, therefore, for low value of \( \tau \)

\[
t_d = \frac{d}{\mu_0 E_0} = \frac{d^2}{\mu_0 v_0^2} \tag{7}\]

where, \( v_0 = E_0 d \), is the applied voltage.

**Case II :** \( \tau \gg \left(t_{\text{inj}} + t_{\text{run}}\right) \) or \( t_d \) and \( \tau \gg t_{\text{inj}} \)

High field regime, \( t_{\text{inj}} \) and \( t_{\text{run}} \) may become low as compared to \( \tau \), and thus for \( \tau > \left(t_{\text{inj}} + t_{\text{run}}\right) \) and \( \tau > t_{\text{inj}} \) an therefore for \( \tau \gg \left(t_{\text{inj}} + t_{\text{run}}\right) \)

\[
t_d = t_i S \frac{E_{\text{th}}}{E_0} \tag{8}\]

**Case III :** \( \tau \) comparable with \( t_{\text{inj}} \) and \( t_{\text{run}} \)

In this case, from eq. (6), \( t_d \) may be expressed as

\[
t_d = \frac{d}{\mu_0 E_0} + t_i S \left[ \exp \left( \frac{t_{\text{inj}}}{\tau} \right) \exp \left( \sqrt{\frac{E_0}{\tau}} \left( 1 - e^{-\frac{t_{\text{inj}}}{\tau}} \right) \right) \right]
\]

\[+ t_i S \left[ -\exp \left( -\frac{t_d}{\tau} \right) \exp \left( \sqrt{\frac{E_0}{\tau}} \left( 1 - e^{-\frac{t_d}{\tau}} \right) \right) \right] - \left( \ln \left( 1 - \frac{E_{\text{th}}}{E_0} \right) \right) \tag{9}\]

Thus, for \( E_{\text{th}} < E_0 \) and \( \tau < t_d \), eq. (9) may be expressed as

\[
t_d = mS + t_{d0} \tag{10}\]

where,

\[
m = t_i \left[ e^{\sqrt{E_{\text{th}}}} - e^{\left( \frac{t_d}{\tau} \right) e^{\sqrt{E_0}}} \right] \tag{11}\]

is the slope between \( t_d \) versus \( S \) plot, and

\[
t_{d0} = \frac{d}{\mu_0 E_0} \tag{12}\]

is the intercept on \( t_d \) axis.

**Experimental Support to the Proposed Theory**

Fig. 1 shows that the plot between \( t_d \) and \( 1/E_0 \) for ITO/TPD/Alq3/Mg/Ag OLED having Alq3 thickness 60 and 30 nm [1], is a straight line with positive slope, whereby the delay time increases with increasing value of \( d \). These results are also in accord
with eq. (7) Fig. 2 illustrates dependence of EL delay time on the device area for ITO/NPD/Alq3/BCP/Ca and ITO/NPD/Alq3/BCP/Mg/Ag OLEDs [3]. It is seen that in this case the delay time increases with device areas, and it is higher for Mg:Ag electrodes as compared to Ca electrode. As the work function is less for Ca as compared to Mg/Ag, $E_{th}$ is less for Ca and it has lower value of $t_d$. These results are in accord with eq. (9) The value of $V_{th}$ is determined by interpolating Fig. 3 of ref. [5], at which the steep rise of $t_d$ with applied voltages takes place and it is found to be 3.0 V for the ITO/α–NPD/Alq3/LiF/Al double layer OLEDs. This value of $V_{th}$ is comparable with that reported by Ichikawa et al. [3] for similar OLEDs.

**Figure 1:** Electric field dependence of the delay times for ITO / TPD / Alq3 / Mg : Ag cell. Open and closed squares show the transit times for the cells with the Alq3 thickness of 60 and 30 nm, respectively (after Hosokawa et al., ref. [1]).

**Figure 2:** Device area dependence of EL delay time for OLEDs using various cathode metals at applied voltage of 32 V (after Ichikawa et al., ref. [3]).
Using eq. (3), the value of $\tau$ is determined from the slope of $t_i$ versus $\ln[1-(E_{th}/E_0)]$ plot. The value of $t_i$ is determined from the value $t_d$ given in Fig. 3 of ref. [5], in which it is mentioned that at an applied voltage of 10 V, only 2% and 6.3% of total delay time is due to the carrier injection, while at 40 V, the share of injection time reached 14.1% and 17.0% for a device area of 0.52 mm$^2$ and 2.11 mm$^2$, respectively. The value of $\tau$ was found to be 0.39 ns and 1.62 ns for the ITO/$\alpha$–NPD/Alq$_3$/LiF/Al double layer OLEDs of area 0.52 mm$^2$ and 2.11 mm$^2$, respectively. These values of $\tau$ are comparable with that reported by Ichikawa et al. [3] for similar OLEDs.

Fig. 3 shows that the plot between $t_d$ and device area $S$ is a straight line with a positive slope and positive intercept on $t_d$ axis, in which the values of slope and intercept decrease with increasing value of the applied voltage or field. This is in accordance with eq. (10). Fig. 4 shows the plot between the intercept, $t_{do}$, and $1/E_0$, is a straight line, in which the slope is equal to $d/\mu_o$ (eq. 12). From the known values of slope between $t_{do}$ and $1/E_0$, and thickness $d$ (60 nm) of the hole–transporting layer, the value of zero–field mobility, $\mu_o$, is determined and it is found to be 4.01 cm$^2$/Vs, which is comparable with that 3.39 cm$^2$/Vs, determined by other method [5]. Using eq. (9) and substituting the value of $t_d$, $d$, $\mu_o$, $E_0$, $\tau$, $E_{th}$ and $t_{ini}$, the value of electric field coefficient, $\gamma$, to the mobility is determined and it is found to be 0.00154, which is comparable with its value 0.00153 determined by experimental method [5].

**Figure 3**: Dependence of $t_d$ on the device area for ITO/$\alpha$–NPD (60 nm)/Alq$_3$ (50 nm)/LiF/Al double layer OLED for different voltages (after the data given in Fig. 3) by Wei et al., ref. [5] ($\alpha$–NPD stands for 4,4’–bis[N–(1–napthyl)–N–phenyl–amino]–biphenyl).
Figure 4: Plot of $t_{do}$ versus $1/E_o$ for ITO/α-NPD (60 nm)/Alq$_3$ (50 nm)/LiF/Al double layer OLEDs.

Conclusions
The delay time of OLEDs should decrease with decreasing value of the threshold voltage or work function of cathode, increase with device area, increase with thickness of the electron or hole transporting layer, and decrease with increasing value of the applied electric field. It is shown that the values of threshold voltage ($V_{th}$), time constant ($\tau$) of OLEDs, zero-field charge carrier mobility ($\mu_0$) and the electric field coefficient ($\gamma$) to the mobility can be determined from the measurement of the dependence of delay time on the strength of applied electric field. A good agreement is found between the theoretical and experimental results.

References