

## **Study of Electroluminescence in Organic Materials by Monomolecular Recombination**

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**Abstract:** *Electroluminescence (EL), a type of luminescence, is the phenomenon of emission of electromagnetic radiation in excess of thermal radiation from condensed matter subjected to an external electric field. The types of EL are distinguished based on the mode of excitation of the radiative system of the EL devices, Basically, EL can be classified, firstly, as high-EL and, secondly as charge –carriers mediated EL. The charge – carriers mediated EL is of two types, namely, impact EL and recombination EL. Furthermore, the recombination EL can be separated either as injection controlled EL (ICEL) or volume controlled EL (VCEL). Here, we would like to discuss high-field EL, charge-carrier mediated EL, impact EL and recombination EL.*

**Keywords:** *Electroluminescence, ROPPV (2-5 – dialoxy-poly-phenylene-vinyl), light emitting diode.*

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### **1. INTRODUCTION**

The EL from organic materials observed for the first time by Bernose and co-workers in (Bernanose 1953) has long been of interest from a basic research point of view because of the insight that can be obtained regarding the electronic process in solids bonded by weak Vander Walls interaction. Many workers have discussed the physical factors affecting performance parameters of organic LEDs and the relative advantages of organic and inorganic EL (Bradly 1993, Allen 1994, Jolinat 1998). The demonstration that organic materials can serve as carriers transporting and luminescent layers in high quantum efficiency LEDs has imposed a strong demand of a comprehensive understanding the LED operation

The types of EL are distinguished based on the mode of excitation of the radiative system of the EL devices; Basically, EL can be classified, firstly, as high field EL and, secondly as charge – carriers mediated EL. The charge – carriers mediated EL is of two types, namely, impact EL and recombination EL. Furthermore, the recombination EL can be separated either as injection controlled EL (ICEL) or volume – controlled EL (VCEL). Here, we would like to discuss high-field EL, charge – carriers mediated EL, impact EL and recombination EL.

### **2. THEORETICAL APPROACH OF ELECTROLUMINESCENCE**

Here, we will discuss the basic theories of different types of EL.

#### **2.1. High – Field EL**

This type of EL is defined by the assumption that an external electric field (F) excites emitting states directly. In quantum – mechanical terms, this excitation mode is described by the probability of excitation of localized states or field ionization from the valence band (VB) (Zener effect). The probability per unit time for ionization from the VB can be (Franz 1952)

$$P_i = \left( \frac{e a_c F}{\hbar} \right) \exp(-b_i/F) \quad (1)$$

where

$$b_i = [8\pi(2m^*)^{1/2}] E_g^{3/2} / 3\hbar e \quad (2)$$

$E_g$  is the band gap,  $m^*$  is the effective mass,  $a_c$  is the unit cell dimension and  $h$  and  $e$  are the Planck's constant and elementary charge, respectively. This expression assumes ionization to be the tunneling process through a triangular barrier of height  $E_g$  and width  $E_g / eF$ . The tunneling rate goes to zero for fields, and is very small for wide-gap materials. For example,  $E_g = 1\text{eV}$ , the tunneling rate becomes significant when  $F$  approaches  $10^7 \text{ Vcm}^{-1}$  and the corresponding tunneling distance is then  $10 \text{ \AA}$ . The relaxation of the electron to the ground state (VB) produces light called high field EL. Because the energy gap for wide – gap semiconductors and insulators is substantially large, the probability of field ionization of localized states such as impurity centers (acceptor and donors) or other effective mass like species (donor – acceptor pair) may be treated in a way similar to a Heitler – London type analysis of the hydrogen molecules, the donor and acceptor may be treated as similar to the hydrogen atom modified by  $m^*$  and dielectric constant of the materials ( $\epsilon$ ) (Oppenheimer 1928). Thus,  $P_i$  may be written as,

$$P_i \propto (m^*/m)(F/\epsilon)^{1/4} \exp [-c_n(m^*/m)E_n^{3/2}/F] \quad (3)$$

With  $E_n = m^*e^{4/2} (4\pi\epsilon_0\epsilon)h^2n^2$  where  $n = 1, 2, 3, \dots$

The electron transitions from the conduction band (CB), or hole transition from the VB to a localized level, lead to light emission and it is the high-field EL. In extrinsic semiconductors, electrostatic ionization and high – field EL are already noticeable in fields with an intensity of  $10^6 \text{ V cm}^{-1}$ . For narrow – band insulating materials a transition from the ground state,  $\{|\psi_1|\}$ , to an excited state,  $\{|\psi_2|\}$ , due to a perturbation by an external field, has been considered as a possible means to excite well localized electronic states. The probability of such a transition can be expressed as

$$P_n = [e\psi_1|r_1nF|\psi_n]^2 / (E_1 - E_n)^2 \quad (4)$$

Where  $E_1$  and  $E_n$  are the energies corresponding to the ground and  $n^{\text{th}}$  excited states, respectively.

## 2.2. Charge Carriers Mediated EL

The charge carriers accelerated by an electric field collide with luminescent centres, chemical and physical defects or the unperturbed material molecules (atoms) exciting them to higher electronic states and those produced in the opposite charge carrier recombination processes after ionization, produces impact EL.

### 2.2.1. Impact EL

At high electric fields the charge carriers can gain a substantial amount of energy from the field and this would show itself as an increase of thermodynamic temperature of the material structure. In a ballistic acceleration process an electron can attain the energy  $E_m$  required to excite or ionize a molecule already over its mean free path  $l_c$  and the probability of the electron experiencing a path greater than  $l_c = E_m / eF$  is

$$P = \exp (-E_m / eFl) \quad (5)$$

In order to accelerate electrons to 2-3 eV and allowing impact generation of excited states, the drift velocity equivalent  $10^8 \text{ cm s}^{-1}$  must be reached. This is possible at high fields with mobility's of  $(100\text{cm}^2\text{V}^{-1}\text{s}^{-1})$  (Kurl 1990, Kari 1990, Pope 1982).

### 2.2.2. Recombination EL

The emission of light by luminescent material resulting from the recombination of electric field injected, opposite sign charge carriers will be called recombination electro-luminescence. Perhaps this is most common type of EL occurring in organic materials. The essential difference between the recombination processes of free carriers in organic and inorganic solids is that the former are concerned with a superposition of energy levels for neutral excited states and ionized states, whereas the later depend strongly on the band structure. Interband optical transitions being the dominant mechanism for recombination in direct semiconductors, where phonons or impurities are required to provide momentum conservation in the recombination process (Weyrich 1978). The excitation and emission bands are often attributed to transitions between states of an impurity system called activators. The recombination processes leading to the lowest-energy excitons, as

typical for organic materials are discussed below. From a kinetic model, assuming the excited states to be produced throughout the sample by an electron-hole recombination process with arbitrary concentrations of electrons  $n_e$  and holes  $n_h$ , the photon flux per unit area from the EL cell of thickness  $d$  can be expressed as

$$\Phi_{EL} = \phi P_s \gamma n_e n_h d \quad (6)$$

Where  $\phi = k_F/k_s$  is the emission quantum yield defined by the ration of radiative ( $k_F$ ) and total ( $K_s = k_F + k_n$ ), decay rate constant for an excited state, including the non-radiative decays with the overall rate constant  $k_n$ .  $P_s$  is the probability that in a recombination event a singlet excited state will be created. In trap – free materials  $P_s = 1/4$  because due to spin statistics, three times more triplet than singlet excited states are created in the electron – hole recombination process characterized by the second – order rate constant  $\gamma$ .

In fact, there are two types of recombination EL; (i) injection – controlled electroluminescence (ICEL), and (ii) volume – controlled electroluminescence (VCEL). When the transit time ( $t_t$ ) is less than the recombination time  $t_r$ , that is  $t_t \ll t_r$ , then we get ICEL. On the other hand, when  $t_t \gg t_r$ , we get VCEL.

The recombination EL is a ‘side effect’ of the current flow and its intensity depends on injection current. Therefore, such type of EL is called injection-controlled EL. The concept of volume-controlled limitations comes about because the doubly injected current greatly exceeds either type of single injection currents. This is obviously due to electron-hole recombination in the sample volume, and thus the accompanying optical emission is called “Volume controlled” EL (VCEL) (Vityuk 1972, Kalinowski 1997, 1996). Among various types of recombination process, the Lengevin-type recombination seems to be most appropriate to describe the recombination process in organic materials. It occurs when the mean free path for optical phonon emission  $l_m \ll r_c = q^2/4\pi\epsilon_0\epsilon kT$ , where  $r_c$  is the Coulombic capture radius, that is, a distance at which the kinetic energy ( $kT$ ) of the particle is equal to Coulombic attractive potential energy ( $q^2/4\pi\epsilon_0\epsilon r_c$ ) of the recombining counterpart. The particles are statistically independent of each other, whereby the carrier recombination is a random process and is kinetically bimolecular in which the recombination efficiency for injected carriers is given by

$$\gamma = e (\mu_e + \mu_h) / \epsilon_0 \epsilon \quad (7)$$

If the carrier lifetime becomes determined by such a process, we deal with the second order decay limit defined by  $\tau_I > \tau_{rec}$

Whereas the space – charge – limited current (SCLCs) under single carrier injection, requires that the charge injected per unit area should approach the capacitor charge  $ne, h = \epsilon_0 \epsilon F/q$ , the volume – controlled currents (VCCs) require only  $(q/n_e) - (n_h/d) = \epsilon_0 \epsilon F$ . If only one of the currents fulfils the SCL conditions (one ohmic contact), that is  $J^{e,h} > J^{h,e}$  and  $n_{e,h} > n_{h,e} \ll \epsilon_0 \epsilon F/ed$ , the EL intensity appears to be a linear function of the injection-limited current (ICL).

### **3. INJECTION ELECTROLUMINESCENCE OF ORGANIC MATERIALS**

It has been reported by many workers (Kurl 1990, Kalinowski 1997, Friend 1990) that in OLED bimolecular recombination of injected charge carriers takes place. However, when the number of trapped charge carrier will be more than the number of free charge carriers, then there is a possibility of monomolecular recombination. Considering the fact, for the kinetics of transient behavior of OLED, we would like to develop the theory based on on both the monomolecular recombination as well as the bimolecular recombination.

#### **3.1. Monomolecular Recombination – Injection Electroluminescence of Organic Materials**

An organic light emitting diode (OLED), is a device in which single layer of organic material is sandwiched between in two electrodes, one of which is transparent. When a single layer of organic materials is sandwiched between two electrodes it is called single layer OLED. When two layers of organic materials is sandwiched between two layer, it is called double layer OLED. On other hand, if more than two layers is between two electrodes, then it is called multilayer OLED.

The light emission in OLEDs takes place due to the radiative decay of excitons produced during the recombination of oppositely charged carriers injected from the cathode and anode of OLED. The monomolecular recombination will be applicable if the number of trapped charge carriers will be more than number of free charge carriers. In the following, the EL emission based on the monomolecular recombination of charge carriers is explored.

Let us consider an OLED through which a current,  $I$ , is flowing because of the carriers injected from electrodes. As current is the rate of flow of charge, the rate of injection of charge carriers is given by,  $I/q$ , where  $q$  is the electronic charge. If,  $S$ , is the cross-sectional area and,  $J$ , is the current density, then,  $I = JS$ , and the rate of injection of charge carriers can be expressed as  $JS/q$ . If  $d$  is the thickness of OLED, then the rate of injection of charge carriers per unit volume will be  $J/qd$ . Thus, the rate of generation,  $g$ , of charge carriers in per unit volume of OLED can be expressed as (Kalinowski 1997).

$$g = \frac{J}{qd} \quad (8)$$

The carrier transit time  $t$ , which is dependent on the carrier mobility,  $\mu$  and electric field,  $F$ , operating in the sample, is given by

$$t_t = \frac{d}{\mu F} \quad (9)$$

Thus, from equation (8) and (9), we get

$$g = \frac{J}{qt_t\mu F} \quad (10)$$

### 3.1.1. Expression for the Temporal Characteristics of Organic Electroluminescence Devices

When electrons and holes are injected in the emissive layer of an organic electroluminescence from electrodes, then recombination of opposite charge carriers takes place, some of the recombinations create excitons and rest of the recombinations takes place without the creation of excitons. If  $\beta_1$  is the rate constant for the monomolecular recombinations producing no excitons, then we can write the following rate equation:

$$\frac{dn}{dt} = g - \beta_1 n - \beta_2 n \quad (11)$$

$$\frac{dn}{dt} = g - \beta n$$

Where  $n$  is the number of charge carriers recombining in the emissive layer at any time  $t$  and  $\beta = (\beta_1 + \beta_2)$ . Here,  $1/\beta$  is the recombination time of charge carriers. It is to be noted that  $\beta = \gamma n_t$  where  $\gamma$  is the recombination coefficient for charge carriers, and  $n_t$  is the density of opposite charge carriers in traps.

When a voltage-pulse is applied at  $t=0$ , the EL emission takes place after a delay time of  $t_d$ . As,  $t_d$  is related to the transport of charge carriers, the recombination and subsequent excitons formation will take place after  $t_d$  (Lchikawa 2003)

Thus, taking  $n = 0$ , at  $t = t_d$  and integrating eq. (11), we get

$$n = \frac{g}{\beta} [1 - \exp\{-\beta(t - t_d)\}] \quad (12)$$

Now, the rate of generation of excitons can be expressed as

$$G = \beta_1 n = \frac{\beta_1 g}{\beta} [1 - \exp\{-\beta(t - t_d)\}] \quad (13)$$

The rate of generation of excitons is  $\beta_1 n, a$  and the rate of recombination of charge carriers is  $\beta n$ . Thus, the probability,  $P_{ex}^m$ , of the formation of excitons during the monomolecular recombination is given by

$$P_{ex}^m = \frac{\beta_1}{\beta} = \frac{\beta_1}{(\beta_1 + \beta_2)} \quad (14)$$

If  $\delta_1$  and  $\delta_2$  are the rate constants for the radiative and non- radiative decay of excitons, then we can write the following rate equation.

$$\frac{dN_{ex}}{dt} = G - \delta_1 N_{ex} - \delta_2 N_{ex}$$

or

$$\frac{dN_{ex}}{dt} = \beta_1 \frac{g}{\beta} [1 - \exp\{-\beta(t - t_0)\}] - \delta N_{ex} \quad (15)$$

Where  $N_{ex}$  is the number of excitons at any time  $t$  and  $\delta = (\delta_1 + \delta_2)$ . Here  $1/\delta$  is the lifetime of excitons.

Taking  $N_{ex} = 0$  at  $t = t_d$  and integrating equation (15), we get

$$N_{ex} = \frac{\beta_1 g}{\beta(\beta - \delta)} [\exp\{\beta(t - t_d)\} - \exp\{-\delta(t - t_d)\}] + \frac{\beta_1 g}{\beta \delta} [1 - \exp\{-\delta(t - t_d)\}] \quad (16)$$

The lifetime of excitons is much shorter than the recombination time of charge carriers, that is  $\delta \gg \beta$ , therefore, eq. (16) can be written as

$$N_{ex} = \frac{\beta_1 g}{\beta \delta} [1 - \exp\{-\beta(t - t_d)\}] \quad (17)$$

As the radiative decay of excitons gives rise to light emission, the photons emitted from a unit volume area of a single layer organic electroluminescence can be expressed as

$$\psi_{EL} = \delta_1 N_{ex} = \frac{\beta_1 \delta_1 g}{\beta \delta} [1 - \exp\{-\beta(t - t_d)\}] \quad (18)$$

For the organic electroluminescence of thickness  $d$ , and surface area  $S$ , the EL brightness can be expressed as

$$\Phi_{EL} = S d \psi_{EL} \quad (19)$$

Thus from eqs. (18) and (19), we get

$$\Phi_{EL} = \frac{s d \beta_1 \delta_1 g}{\beta \delta} [1 - \exp\{-\beta(t - t_d)\}] \quad (20)$$

### 3.1.2. Rise of the Organic EL Brightness

For low values of  $(t - t_d)$ , eq.(20) can be written as

$$\Phi_{EL} = \frac{s d \beta_1 \delta_1 g}{\delta} (t - t_d) \quad (21)$$

The above equation indicates that when the voltage will be applied to organic electroluminescence, then initially  $\Phi_{EL}$  should increase linearly with  $(t - t_d)$ .

It is evidence from eq. (20) that in monomolecular recombination,  $\Phi_{EL}$  should attain a satiations, value for larger values of  $(t - t_d)$  and it is given by

$$\phi_{EL}^{sm} = \frac{sd\beta_1\delta_1g}{\beta\delta} \quad (22)$$

or

$$\phi_{EL}^{sm} = \frac{S\beta_1\delta_1J}{q\beta\delta}$$

Now, eq. (20) can be expressed as

$$\Phi_{EL} = \phi_{EL}^{sm} [1 - \exp\{-\beta(t - t_d)\}] \quad (23)$$

It follows from eq. (20) that the recombination time,  $t_{rec}$ ,  $1/\beta$ , of the charge carriers can be determined from inverse of the slope of plot  $(\phi_{EL}^{sm} - \Phi_{EL})/\phi_{EL}^{sm}$  between  $\ln$  and  $(t-t_d)$ .

It is seen from eq. (22) that,  $\phi_{EL}^{sm}$  is proportional to the injection current density,  $P_{ex}^m = \beta_1/\beta$  through a linear relation has been found between EL brightness and J for certain organic electroluminescence devices (Tang 1989, Zhang 1993, Braun 1992) in many cases nonlinear correction has been reported. The nonlinear relation may occur due to several  $P_{ex}^m = \beta_1/\beta$  ping of charge carriers in organic materials, (i) dependence of the probability, f the formation of excitons during the monomolecular recombinations on the current density J, (ii) dependence of  $\eta_1 = \delta_1/\delta$  efficiency, for  $P_{ex}^m$  radiative decay of excitons on the current density J and so on. It has been reported that, increases with the current (or voltage) (Kalinowaski 1997, Friend 1999, Bozano 1999). It has been shown that the EL brightness increases with current (or voltage) due to the contribution from traps which are distributed exponentially (Kalinowaski 1997, 1998, 1994, Burrows 1996).

Thus more accurately eq. (20) may be written as:

$$\phi_{EL}^{sm} = \frac{H_0 S \beta_1 \delta_1 J^p}{q \beta_1 \delta} \quad (24)$$

Where p is power factor and  $H_0$  is a constant taking account of the inclusion of p, in eq. (24)

Equation (24) indicates that the dependence of  $\phi_{EL}$  on J should follow the power – type relation. It is to be noted that Kalinowaski (1997) and Rieess (1997) have reported the power – type relation between  $\phi_{EL}$  and J thus their results are in accord with eq. (24).

### 3.1.3. Decay of Organic EL brightness

When the applied voltage will be switched off at  $t = t_c$ , then g will become zero at  $t=t_c$  and from eq. (11), we get

$$\frac{dn}{dt} = -\beta n \quad (25)$$

Equation (12) shows that for higher values of  $(t_c-t_c)$  we can take  $n=g/\beta$  at  $t = t_c$ . Thus, taking  $n= n_0$ , at  $t = t_c$  the integration of eq. (25) gives

$$n = n_0 \exp[-\beta(t - t_c)] \quad (26)$$

In this case, the rate of generation of excitons will be  $G = \beta_1 n$  and thus we can write the following rate equation

$$\frac{dN_{ex}}{dt} = G - \delta N_{ex}$$

or

$$\frac{dN_{ex}}{dt} = \beta_1 n - \delta N_{ex}$$

or

$$\frac{dN_{ex}}{dt} = \beta_1 n_0 \exp[-\beta(t - t_c)] - \delta N_{ex} \quad (27)$$

Taking  $N_{ex} = N_{exp} = N_{ex}^0$  and integrating eq. (27), we get

$$N_{ex} = \frac{\beta_1 n_0}{\delta - \beta} [\exp\{-\beta(t - t_c)\} - \exp\{-\delta(t - t_c)\}] + N_{ex}^0 \exp[-\delta(t - t_c)] \quad (28)$$

As  $(t_c - t_c)$  is sufficiently large, where saturation of  $\phi_{EL}$  is occurred, from eq. (17) the value  $N_{ex}^0$  can be expressed as

$$N_{ex}^0 = \frac{\beta_1 g}{\beta \delta} \quad (29)$$

Now, substituting the values  $n_0$  and  $N_{ex}$  in eq. we get

$$N_{ex} = \frac{\beta_1 g}{\beta(\delta - \beta)} [\exp\{-\beta(t - t_c)\} - \exp\{-\delta(t - t_c)\}] + \frac{\beta_1 g}{\beta \delta} \exp[-\delta(t - t_c)] \quad (30)$$

Using eq. (24), the decay of organic electroluminescence brightness can be expressed as

$$\phi_{EL} = sd\delta_1 N_{ex}$$

$$\phi_{EL} = \frac{sd\beta_1 \delta_1 g}{\beta(\delta - \beta)} [\exp\{-\beta(t - t_c)\} - \exp\{-\delta(t - t_c)\}] + \frac{sd\beta_1 \delta_1 g}{\beta \delta} \exp[-\delta(t - t_c)]$$

or

$$\phi_{EL} = \frac{sd\beta_1 \delta_1 g}{\beta(\delta - \beta)} [\exp\{-\beta(t - t_c)\} - \frac{\beta}{\delta} \exp\{-\delta(t - t_c)\}] \quad (31)$$

As  $\delta \gg \beta$ , eq. (26) can be written as

$$\phi_{EL} = \frac{sd\beta_1 \delta_1 g}{\beta \delta} [\exp\{-\beta(t - t_c)\}]$$

$$\phi_{EL} = \phi_{EL}^{sm} [\exp\{-\beta(t - t_c)\}] \quad (32)$$

Equation (32) indicates that the organic electroluminescence brightness should decay exponentially with  $(t - t_c)$  where the inverse of the slope plot between  $\ln(\phi_{EL})$  and  $(t - t_c)$  will give the lifetime,  $1/\beta$  of the charged carriers.

#### 4. CONCLUSION

A very high electric field is required for high field EL, it seems that the EL in organic material like ROPPV, MEH – PPV and family member of PPV polymer is related to the high – field EL. Theoretical study has been made to the injection of electroluminescence in monomolecular. The theoretical approach to the injection of charge in organic materials is dealt with, on the basis of the monomolecular recombination of injected charge carriers, considering that in organic material, light emission takes place during the radiative decay of singlet excitons produced during the ionization or excitation or recombination between electrons injected from cathode and holes injected from anode, expressions have been derived for the injection of charges on organic materials. For the monomolecular EL brightness found voltage – pulse is applied at  $t = 0$  on thickness,  $d$  and surface area  $S$ , taking  $n = n_0$  at  $t = t_c$ . The EL brightness in case of monomolecular recombination based injection electroluminescence of organic materials has been derived as

$$\phi_{EL} = \frac{sd\beta_1 \delta_1 g}{\beta \delta} [1 - \exp\{-\beta(t - t_d)\}]$$

Where  $\beta$  is the rate constant for monomolecular recombination,  $t_d$  is delay time,  $\beta_1$  is the recombination coefficient for the radiative.

Rise of the organic EL brightness ( $\phi_{EL}$ ) at any time (t) in case of monomolecular recombination based injection electroluminescence of organic materials has been derived at delay time of

$$\phi_{EL} = \phi_{EL}^{sm} [1 - \exp -\beta(t - t_d)]$$

$\phi_{EL}^{sm}$  = saturated EL brightness

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