The Study of Luminescence Efficiency by change of OLED's Hole Transport Layer

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KEYWORDS : OLED, Monomer, Hole transport layer, Luminescence efficiency, Numerical analysis, Simulation

The OLEDs(Organic Light-Emitting Diodes) structure organizes the bottom layer using glass, ITO(indium thin oxide), hole injection layer, hole transport layer, emitting material layer, electron transport layer, electron injection layer and cathode using metal. OLED has various advantages. OLEDs research has been divided into structural side and emitting material side. The amount of emitting light and luminescence efficiency has been improved by continuing effort for emitting material layer. The emitting light mechanism of OLEDs consists of electrons and holes injected from cathode and anode recombination in emitting material layer. The mobilities of injected electrons and holes are different. The mobility of holes is faster than that of electrons. In order to get high luminescence efficiency by recombine electrons and holes, the balance of their mobility must be set. The more complex thin film structure of OLED becomes, the more understanding about physical phenomenon in each interface is needed. This paper observed what the thickness change of hole transport layer has an affection through the below experiments. Moreover, this paper uses numerical analysis about carrier transport layer thickness change on the basis of these experimental results that agree with simulation results.

Manuscript received: March 4, 2005 / Accepted: October 31, 2005

NOMENCLATURE

J_n, J_p = Electron and hole current densities
Ψ = Electrostatic potential
q = Electric charge
ε = Static dielectric constant
R = Optical recombination rate
G = Optical generation rate

1. Introduction

Emitting-light mechanism of OLEDs consists of electrons and holes injected from cathode and anode recombination in emitting material layer. The mobilities of injected electrons and holes are different. The transport velocity of holes is faster than that of electrons. In order to get effective recombination by electron-hole pairs, their mobility balance has to be set. Namely, carrier transport layer and carrier injection layer between emitting material layer and anode or cathode need to balance to set mobility. Since thin-film deposition structure of OLEDs becomes more complex gradually, I must know that effort for comprehension of physical phenomenon in each interface becomes more serious. In order to get high luminescence efficiency in specification thickness, it would be useful to observe luminescence efficiency of emitting material layer changing thickness of hole transport layer. This paper surveys such phenomenon through an experiment. Moreover, we compared simulation results about carrier transport layer thickness change on the basic structure to numerical analysis. The structure of OLEDs is generalized by the OLEDs structural improvement effort and the research of the organic materials in superior emitting light material development. However, it is very difficult to comprehend interaction of cathode and electron from electron injecting layer. I already know that energy barrier of carriers (electron or hole carrier) using ITO of anode and using metal of cathode are so high, that transport carriers injection are not smooth. I have made effort to improve luminescence efficiency of OLEDs studied steadily. The improved method puts hole transport layer and electron transport layer in basis on the device rescue and carriers can pass over the energy barrier easily. However, these methods by the electrons and the holes transport the velocity of OLEDs show difference in each other by change of voltage, so electron-hole recombination in emitting light layer harmoniously create, and luminescence efficiency is dropped. Method to set transport velocity balance of two kinds of carriers by carrier injection layer and carrier transport layer to heighten luminescence efficiency should be presented.

That is, by accomplishing high-electron-hole pairs the lower mobility of hole that expresses fast mobility by thickness adjustment of hole transport layer should be presented. It is difficult to predict or comprehend the electron that multi-layer structures become in inner organic layer phenomenon if work function is made by slow stair so that carriers may be smooth injecting of hole consists harmoniously and migration of hole transport on emitting light layer. This paper forecasted beforehand the actions of electron and hole that occur in inner OLEDs by experimental results. It also expects thickness of proper thin film using simulation results and compares each other.

2. Experiment

OLEDs are sensitive device in temperature and humidity. Usually, ITO-coated glass was used as the substrate for OLEDs. The sheet resistance of ITO anode was about 120 $[\Omega / \square]$. The base chamber pressure was under 3 x 10^{-6}[Torr]. Chambers are consisted of metal and organic. Organic materials were prevented from being exposed to air by blowing nitrogen atmosphere in each process. The deposition rates were controlled by quartz oscillating thickness monitor to be 0.1 ~ 0.2 [nm/s] for the organic materials, 1.0 [nm/min] for Al electrode respectively. OLEDs having the structure of ITO/NPB/ Alq_3/Cathode were fabricated multiple organic molecular beam deposition method. Fig. 1 shows the typical structure of the OLEDs and chemical structure of the organic materials. ITO transparent electrode was inserted on glass substrate, and NPB was used as hole transport layer, and Alq_3 was used as emitting material. Vacuum evaporation in organic material Chamber ends and moves by metal Chamber in possible similar vacuum environment and inserted Al by anode.

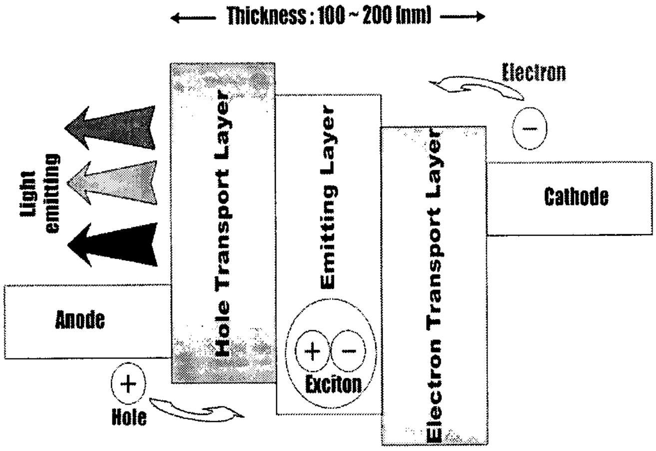

Fig. 1 Basics structure of OLED

Hole injection layer (HIL) and hole transport layer (HTL) experiment manufacturing basic thickness by 50 [nm] and measure these and represent result this to thickness of each 10/20/30/40, 40/30/20/10. Hole transport layer used NPB and hole injection layer used CuPc. By emitting light layer (Emitting Layer) Alq_3 40 [nm] do and manufactured basic device. And cathode and anode electrode used Al, ITO respectively.

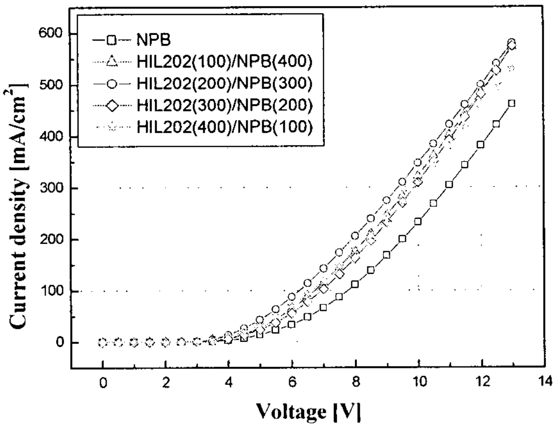

Fig. 2 Characteristics of OLEDs current density vs applied voltage

OLEDs helps hole injection layer using transparency electrode such as ITO (Indium Tin Oxide) to anode to make light which is emitted light in emitting layer be emitted outside. and lower work function uses metal to cathode electrode and turn out electron injection layer of that is smooth.

Fig. 2 shows manufactured OLEDs voltage – current density. As see result that measure in Fig. 2 could know that the best characteristic of voltage and current density appear thickness of hole transport layer is 30 [nm]. This certifies that fast mobility of hole structurally could make pair of electron and hole by hole injection layer and thickness adjustment of transport layer most efficiently.

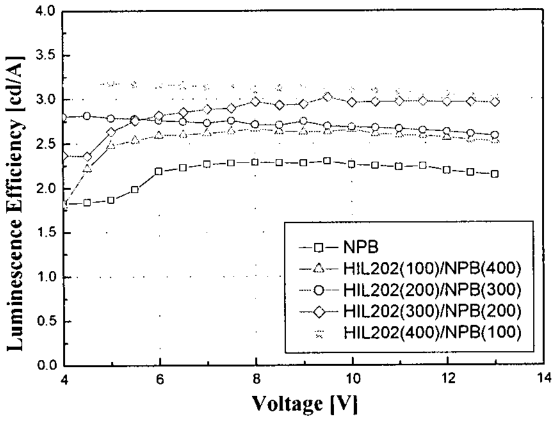

Fig. 3 luminescence efficiency of OLEDs

As we can observe characteristics of OLEDs applied voltage anu current density in Fig. 3, the variety in the layer of OLEDs has influences on the luminescence efficiency.

3. Simulation

Fig. 4 shows the simulation of OLED structure. The transport of electrons and holes in organic device can be solved by the continuity equation, with a drift-diffusion equation, coupled to Poisson's equation. Energy level discontinuities at the organic hetero-junction can be used to produce an energy barrier that blocks charge transport across the structure.

$$\frac{dJ_n}{dx} = -q(G - R) \qquad (1)$$

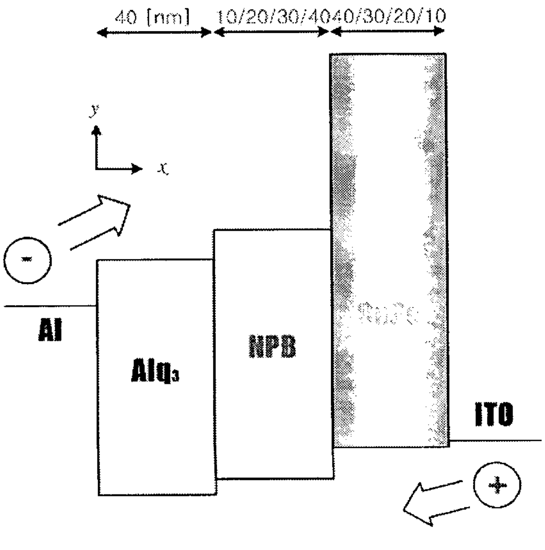

Fig. 4 Schematic of OLED structure

$$\frac{dJ_p}{dx} = q(G-R) \tag{2}$$

$$\frac{d^2\psi}{dx^2} = -\frac{q}{\varepsilon}(p-n) \tag{3}$$

where, J_n and J_p are the electron and hole current densities, respectively. Electrostatic potential ψ are function of the length of the device, q is the electric charge and ε is the static dielectric constant. The optical recombination rate R is given by $R = \gamma\ n\ p$, where $\gamma = 4\pi q\mu_R / \varepsilon$ is Langevin recombination coefficient. The generation of electron hole pair (EHP) is given by $G = \gamma\ n_e p_e$, where n_e, p_e is thermal equilibrium electron and hole carrier density. Effective recombination mobility μ_R taken to be larger either the hole mobility μ_p or the electron mobility μ_n.

In equilibrium, the electron density n and the hole density p is represented to n_e and p_e using Maxwell-Boltzmann statistics,

$$n_e = n_0 \exp\left(\frac{q\psi - q\phi_F + \chi_C}{kT}\right) \tag{4}$$

$$p_e = p_0 \exp\left(-\frac{q\psi - q\phi_F + \chi_C + E_g}{kT}\right) \tag{5}$$

where, ϕ_F is the Fermi level in equilibrium and χ_C is the electron affinity. T is the temperature in Kelvin and k is the Boltzmann's constant. n_0 is molecule's density of state and E_g is energy gap.

The drift-diffusion equations defining the electron and hole currents, J_n and J_p are;

$$J_n = q\ \mu_n \left(n\ E + \frac{kT}{q}\frac{dn}{dx}\right) \tag{6}$$

$$J_p = q\ \mu_p \left(p\ E - \frac{kT}{q}\frac{dp}{dx}\right) \tag{7}$$

where, the electric field is given by $E = -d\psi / dx$, the electron and hole mobility is given by $\mu_n = \mu_{n0} \exp(E/E_0)$ and $\mu_p = \mu_{p0} \exp(E/E_0)$, respectively. μ_{p0} and μ_{n0} are hole and electron mobility in zero electric field, respectively. The equations are solved numerically using a Sharfetter-Gummel spatial discretization method,

$$J_{p_{i+\frac{1}{2}}} = \frac{kT\mu_p}{\Delta x}\left[p_i B\left(\frac{q\psi_{i+1} - q\psi_i}{kT}\right) - p_{i+1} B\left(\frac{q\psi_i - q\psi_{i+1}}{kT}\right)\right] \tag{8}$$

$$J_{n_{i+\frac{1}{2}}} = \frac{kT\mu_n}{\Delta x}\left[n_{i+1} B\left(\frac{q\psi_{i+1} - q\psi_i}{kT}\right) - n_i B\left(\frac{q\psi_i - q\psi_{i+1}}{kT}\right)\right] \tag{9}$$

where, $\Delta x = x_{i+1} - x_i$ is differential mesh size, $B(y) = y/(\exp(y) - 1)$ is function of Bernoulli.

At the metal-organic-metal contact, there are boundary conditions. Firstly, total current is sum of thermionic current from $x = 0$ to $x = L$ and back-flowing interface recombination current and FN (Fowler-Nordeim tunneling) current,

$$J_p(0) = -qv_{rp}\left(p_e[E(0)] - p(0)\right) - \left(J_{tp}\big|_{x=0} - J_{tp0}\big|_{x=0}\right) \tag{10}$$

$$J_n(L) = qv_{rn}\left(n_e[E(L)] - n(L)\right) + \left(J_{tn}\big|_{x=L} - J_{tn0}\big|_{x=L}\right) \tag{11}$$

where, electron and hole's effective recombination velocity are $v_{rn} = 16\pi\varepsilon\mu_n (kT)^2 / q^3$, $v_{rp} = 16\pi\varepsilon\mu_p (kT)^2 / q^3$ respectively. And the hole and electron density of quasi-equilibrium in $x = 0$, $x = L$ is

$$p_e[E(0)] = n_0 \exp\left(-\frac{\varphi_{bp} - \Delta\varphi_{bp}}{kT}\right) \tag{12}$$

$$n_e[E(L)] = n_0 \exp\left(-\frac{\varphi_{bn} - \Delta\varphi_{bn}}{kT}\right) \tag{13}$$

where, Schottky hole and electron electric potential barrier are φ_{bp}, φ_{bn}. If φ_{bp} and φ_{bn} are negative electric potential, there are barrier lowering by image force. In this case the model also incorporates image force lowering of the barrier at contacts.

$$\Delta\varphi_{bp} = \sqrt{qE(0)/4\pi\varepsilon} \tag{14}$$

$$\Delta\varphi_{bn} = \sqrt{qE(L)/4\pi\varepsilon}\ . \tag{15}$$

The Fowler-Nordheim currents take the form

$$J_{tp}\big|_{x=0} = C_p E(0)^2 \exp\left(-\frac{\kappa_p}{E(0)}\right) \tag{16}$$

$$J_{tn}\big|_{x=L} = C_n E(L)^2 \exp\left(-\frac{\kappa_n}{E(L)}\right) \tag{17}$$

where, constant coefficient is given by

$$C_p = 3q^2 / 8\pi h\left(\varphi_{bp} - \Delta\varphi_{bp}\right) \tag{18}$$

$$\kappa_p = 8\pi\sqrt{2qm}\left(\varphi_{bp} - \Delta\varphi_{bp}\right)^{3/2} / 3h \tag{19}$$

$$C_n = 3q^2 / 8\pi h\left(\varphi_{bn} - \Delta\varphi_{bn}\right) \tag{20}$$

$$\kappa_n = 8\pi\sqrt{2qm}\left(\varphi_{bn} - \Delta\varphi_{bn}\right)^{3/2} / 3h \tag{21}$$

The position independent of the total current $J = J_p + J_n$ is used to verify that steady state has been reached. At steady state, one can obtain the recombination current $J_r = J_p(0) - J_p(L)$. These quantities are related to the quantum efficiency $\eta_q = QJ_r / J$, and power efficiency $\eta_p = Q(J_r / J)(E_g / V)$ by multiplying the ratio of radiation to total recombination. The ratio of radiation to total recombination is $Q = 1/4$ because of a quarter of the excitons forms are singlets.

4. Results

This paper presents calculation of change of hole transport layer in organic light emitting diode characteristics using two carrier device models that include charge injection, transport, FN tunneling and space charge effects in the organic material. I applied the device model to organic material device using structure of Al/ Alq$_3$/NPB/CuPc/ITO in which the hole is majority carrier. The calculations include carrier diffusion and field dependent mobility therefore it is hard to reduce the way of simple analytic form to describe the limited regime of the space charge to be derived for field independent mobility and neglecting diffusion. It was considered that the energy barrier is much smaller than the hole and it dominates the current flow in the device. The temperature in all calculations is also considered at room temperature. The model calculations show a good description of the measured I –V characteristics over a wide current range in both structures. I can control the optimum thickness of hole transport layer in high luminescence efficiency effect handful.

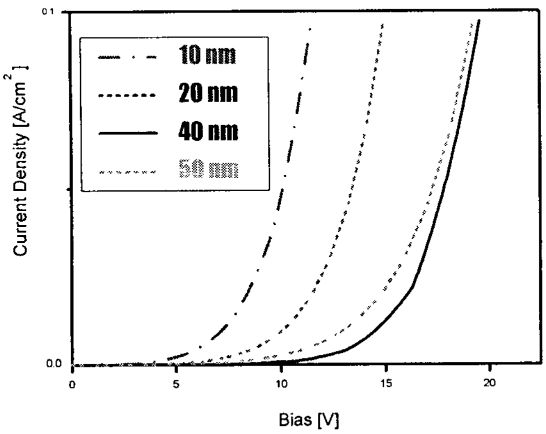

Fig. 5 Al/Alq_3/NPB/CuPc/ITO characteristics of current density vs applied voltage in simulation result of OLED structure

5. Discussions

The experiment shows that, the mobility of hole, is faster by 1~2 order degrees than the mobility of electron. In order to increase the luminescence efficiency, many electron-hole pairs must be made and the mobility of electron must be made faster. Therefore, it is important to search for the optimized thickness to make many electron-hole pair as possible. Specially, thickness adjustment of hole transport layer is required more urgently in case of manufacturing organic light emitting diode by multi-thin-film layer structure. I could know the thickness of hole transport layer that express maximum luminescence efficiency, which was in good agreement with simulation result in Fig. 5.

ACKNOWLEDGEMENT

This research was supported by the 2004 Hongik University Academic Research Support Fund.

REFERENCES

1. Martin, Simon J., Lupton, J. M., Samuel, I. D. W. and Walker, Alison B., “Modeling temperature-dependent current-voltage characteristics of an MEH-PPV organic light emitting device, " J. Phys., Vol. 14, No. 42, pp.9925-9933, 2002.
2. Malliaras, G. G. and Scott, J. G., "Roles of injection and mobility in organic light emitting diodes," Appl. Phys., Vol. 83, No.10, pp.5399-5403, 1998.
3. Davids, P. S., Kogan, Sh. M., Parker, I. D. and Smith, D. L., "Charge injection in organic light-emitting diodes: Tunneling into low mobility materials," Appl. phys. Lett., Vol. 69, pp.2270-2272, 1996.
4. Kepler, R. G., Besson, P. M., Jacobs, S. J., Anderson, R. A., Sinclair, M. B., Valencia, V. S. and Cahill, P. A., "Electron and hole mobility in tris(8-hydroxyquinolinolato-N1,O8) aluminum," Appl. Phys. Lett., Vol. 66, pp.3618-3620, 1995.
5. Pay, D. M., "Transient Photoconductivity in Poly(N-vinylcarbazole)," J. Chem, Phys., Vol. 52, pp.2285-2291, 1970.
6. Crone, B. K., Davids, P. S., Cambell, I. H. and Smith, D. L., "Device model investigation of single layer organic light emitting diodes, " J. Appl. Phys., Vol. 87, No. 2, pp.833-842, 1998.
7. Blades, C. D. J. and Walker, Alison B., "Simulation of organic light emitting diodes, " Synthetic Metals., Vol. 111-112, pp.335-340, 2002.
8. Conwell, E. M. and Wu., M. W., "Contact injection into polymer light-emitting diodes," Appl. phys. Lett., Vol. 70, pp.1867-1869, 1989.