

Efficient inter-molecular energy transfer via dye-dopants in poly(methylphenylsilane) based electroluminescent devices [☆]

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Abstract

Organic electroluminescent devices, comprised of polymer host (poly(methylphenylsilane)) and dye dopants (perylene and 4-dicyanomethylene-6-cp-julolidinostyryl-2-tert-butyl-4H-pyran; DCJTB), have fabricated and characterized to investigate the inter-molecular energy transfer. The device has configuration of indium–tin-oxide (ITO) anode a buffer layer of poly(3,4-ethylenedioxythiophene):poly(4-styrenesulfonate) (PEDOT:PSS) (30 nm), emitting layer of PMPS 1 wt% of THF: dye dopant with a mole ratio of 0.1–1.0% (60 nm) and an electron transporting layer of tris(8-hydroxyquinoline) aluminum (Alq₃) (20 nm) and LiF (0.5 nm) Al cathode (100 nm). Energy transfer mechanism between PMPS and dye dopants was studied by absorption, photoluminescence and electroluminescence spectra. Current–voltage–luminance (*I*–*V*–*L*) characteristics of the devices were also discussed. The polysilane based organic electroluminescent diodes exhibited a maximum external quantum efficiency of 0.87% and a maximum luminous efficiency of 0.36 lm/W and luminance of 890 cd/m² (at a driving voltage of 21 V). The excitation energy was transferred from the polysilane to the dye dopants efficiently and the energy matching between polymer and dopants was the most important factor in fabricating the EL devices through the simple wet processes for the dye-dopants electroluminescent devices.

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1. Introduction

Organic light-emitting devices (OLEDs) have attracted much attention due to their advantages for the full color flat panel displays application, such as low cost, superior viewing performance and excellent operating characteristics (wide viewing angle, fast response time, high contrast etc.) [1–3]. In recent years, significant developments have been made through the multi layer and dye doping structure to modify the emission range and to enhance the efficiency and lifetime. The high color purity and improved efficiency could be obtained

by the proper choice of energy-well-matched hosts and dopants [4].

In host–guest system such as polymer host and dye guest, excited energy from the host are transfers to the luminescent dye via a Förster and Dexter energy transfer. Förster energy transfer is a long range dipole–dipole coupling, thus its available range is up to 50–80 Å. These mechanisms only transfer the singlet state. Dexter transfer which is dominant mechanism in phosphorescent material, is a short-range process and the energy transfer range is about 10–15 Å. In contrast to Förster transfer, Dexter transfer permits both singlet–singlet and triplet–triplet transfer [5,6].

Polysilanes are well known as a new type of σ -conjugated polymers which exhibit characteristics such as photoconductivity with high hole mobility, large non-linear optical effects and effective light emission due to one-dimensional exciton.

In this paper, we investigated the electrical and optical characteristics of the polysilane based

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electroluminescent devices fabricated with the dye molecules, perylene and 4-dicyanomethylene-6-cp-julolidinostyryl-2-tert-butyl-4H-pyran (DCJTB) as the emitting dopants. In addition, the inter-molecular energy transfer between polymer host and dye dopants was also discussed.

2. Experimental

Prior to organic layer deposition, the ITO (indium–tin–oxide, sheet resistance of $10 \Omega/\square$) coated substrates were degreased in ultrasonic solvent baths of toluene, isopropyl alcohol, acetone and methanol consecutively. And then, air plasma treatment was followed to remove the surface spikes in the substrates. PEDOT:PSS, used as a buffer layer to decrease OLEDs leakage current [7] and to increase OLED fabrication yield [8], was spun onto the ITO and baked for 15 min at 120°C , attaining an approximate thickness of 30 nm. All devices fabricated with the same concentration of 1 wt% PMPS and various concentration of perylene (0.1–1.0 mol%), DCJTB (0.1–1.0 mol%). This polymer and dye dopants solution was spin-coated onto the PEDOT:PSS layer to form a 60 nm thickness. All molecular organic layers and metal electrode were sequentially deposited without breaking vacuum by thermal evaporation at the base pressure of 1×10^{-5} Torr. Deposition began with a 20 nm-thick tris(8-hydroxyquinoline)aluminum (Alq_3) electron transporting layer with the deposition rate of 0.5 Å/s. The cathode consisted of 0.5 nm of LiF followed by 100 nm of Al [9].

The samples were only exposed to air while being tested. The deposition rate and thickness of the organic and inorganic materials were monitored by quartz crystal oscillator thickness monitor and the thickness was confirmed by α -step. Fig. 1 shows the organic materials used in the fabrication of the OLEDs in this study.

The PMPS, used as a host material, was synthesized by Wurtz-type reaction of dichloromethylphenylsilane with metal sodium. The detailed condition of the synthesis was reported elsewhere [10–12]. The molecular weight of PMPS was 24,000 by gel permeation chromatography using polystyrene as the standard. The structure of PMPS was confirmed by ^1H NMR, ^{29}Si NMR at 400 MHz (Varian Inity INOVA400) and UV–vis spectra (RF-540PC). The Alq_3 and LiF were purchased from Aldrich and used without any other purification.

The configurations of the OLEDs were shown in Fig. 1. The structure of the multi-layer OLED were shown as follows; ITO coated glass substrate/buffer layer (PEDOT:PSS 30 nm)/emitting layer (PMPS:Perylene:DCJTB 60 nm)/electron transporting layer (Alq_3 20 nm)/electron injection layer (LiF 0.5 nm)/Al

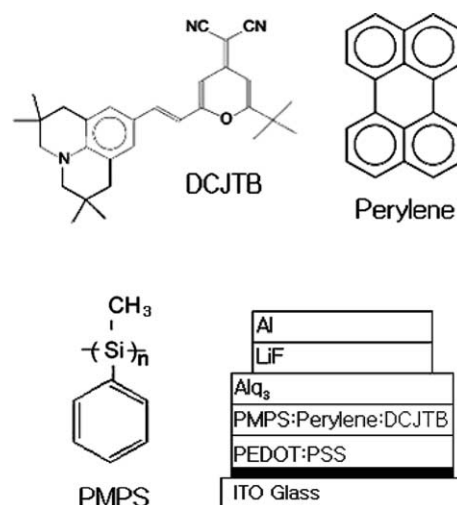


Fig. 1. Molecular structure and device configuration of perylene, 4-dicyanomethylene-6-cp-julolidinostyryl-2-tert-butyl-4H-pyran (DCJTB) and poly(methylphenylsilane) (PMPS).

(100 nm). The emissive layer of the device is 6 mm^2 ($2 \times 3 \text{ mm}$).

The characteristics of the device were studied by measuring the photoluminescence (PL), electroluminescence (EL) spectra and current density–voltage–luminance (J – V – L) curves at room temperature. The PL spectra were measured with a Shimadzu Spectrofluorophotometer (RF-5301PC). The J – V – L and EL characteristics were measured using a Keithley 2000 multimeter equipped with a calibrated Si photodiode (Hamamatsu Photonic).

3. Results and discussion

The probability per unit time of resonant energy transfer from excited energy donor to energy acceptor depends on the energy overlap of the emission spectrum of donor and absorption spectrum of acceptor.

As shown in the Fig. 2, the fluorescence spectrum of PMPS host overlaps with the absorption of perylene and DCJTB dopants. Thus, excitation energy of the PMPS can be transferred to the perylene and DCJTB, and the excitation energy of the perylene can also be efficiently transferred to the DCJTB because of the large spectral overlap.

The photoluminescence spectra of the PMPS host material and mixed materials of PMPS, perylene and DCJTB films are shown in Fig. 3. The maximum peak of PMPS film doped with perylene and DCJTB are observed at 446, 470, 505 nm originated from perylene and 565 nm originated from DCJTB. As the doping concentration of perylene and DCJTB peak intensity increases, PMPS peak decreases gradually. This indicates the excitation energy transferred from PMPS to the dye

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