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Correlation of doping concentration, charge transport of host, and lifetime of thermally activated delayed fluorescent devices



Thermally activated delayed fluorescent (TADF) organic light-

emitting diodes (OLEDs) are attractive as high efficiency OLEDs

challenging a theoretical 100% internal quantum efficiency (IQE) of

phosphorescent OLEDs [1-6]. Although the emission process of

TADF OLEDs is different from that of phosphorescent OLEDs in that

the TADF emission is from singlet excitons rather than triplet ex-

citons of phosphorescent emission [7–9], the TADF OLEDs can

reach the same IQE level of phosphorescent OLEDs by complete

utilization of all singlet and triplet excitons for singlet emission

[10,11]. It was already proven that the QE of the TADF OLEDs is as

high as that of the phosphorescent OLEDs [12]. In particular, (4s,

6s)-2,4,5,6-tetra(9H-carbazol-9-yl)isophthalonitrile (4CzIPN) TADF

lifetime green emitter in addition to the high OE [13]. In previous

works, the lifetime of the 4CzIPN device was similar to that of

common green phosphorescent OLEDs. Adachi et al. reported long

lifetime in the 4CzIPN device at a high doping concentration of 15% using 3,3-di(9*H*-carbazol-9-yl)biphenyl (mCBP) as the host mate-

rial [13]. However, optimum doping concentration of the TADF

The 4CzIPN emitter was also verified to have a potential as a long

emitter achieved a high QE above 30% as a green emitter [3].

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1. Introduction

ABSTRACT

Origin of doping concentration dependence of lifetime of thermally activated delayed fluorescent (TADF) devices was examined using a TADF emitter doped in a hole transport type and a bipolar host material. Lifetime of the hole transport type host based TADF device was increased according to doping concentration of TADF emitter, while that of the bipolar host based TADF device was decreased according to doping concentration of TADF emitter. The doping concentration dependence of the lifetime could be correlated with recombination zone of the emitting layer. Broad recombination zone at high doping concentration in the hole transport type host and at low doping concentration in the bipolar host was proposed as the main contributor of the doping concentration dependence of the lifetime.

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emitters was relatively high compared with that of the phosphorescent emitters in the lifetime evaluation of the two emitters. Typical doping concentration range of TADF emitters for optimized lifetime was around 20%, but that of phosphorescent emitters was below 10%. However, it is not clear why the optimum doping concentration of TADF emitter is higher than that of phosphorescent emitter in terms of lifetime. Therefore, it would be helpful to understand the origin for the optimum doping concentration of TADF emitters.

Herein, the doping concentration dependence of the lifetime of the TADF devices was elucidated by examining the emission zone of the emitting layer according to doping concentration of 4CzIPN using a hole transport type host and a bipolar host. The doping concentration could be correlated with emission zone of the emitting layer and the lifetime was optimized at the doping concentration showing broad emission zone. It was verified that strong electron trapping by the 4CzIPN emitter is the main parameter affecting the doping concentration dependent emission zone change.

2. Experimental

The 4CzIPN devices for lifetime measurement had the device structure of organic materials stacked on indium tin oxide (ITO, 120 nm) substrate. *N*,*N*'-diphenyl-*N*,*N*'-bis-[4-(phenyl-*m*-tolyl-amino)-phenyl]-biphenyl-4,4'-diamine (DNTPD) was stacked on

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the ITO as a hole injection material, *N*,*N*,*N*'*N*'-tetra[(1,1'-biphenyl)-4-yl]-(1,1'-biphenyl)-4,4'-diamine (BPBPA) was deposited as a hole transport material, 9,9-dimethyl-10-(9-phenyl-9H-carbazol-3-yl)-9,10-dihydroacridine(PCZAC) was deposited as a buffer material, 3,3-di(9H-carbazol-9-yl)biphenyl (mCBP):4CzIPN or 4,6-di(9Hcarbazol-9-vl)isophthalonitrile (DCzIPN):4CzIPN was co-deposited as an emitting layer, and 1, 3, 5-tris(*N*-phenylbenzimidazole-2-yl) benzene (TPBI) was evaporated as an electron transport laver. Cathode was a double layer of LiF and Al. For emission zone measurement, 4CzIPN devices with a 2 nm thick red sensing layer at 5, 15, and 25 nm from the hole transport layer were fabricated. Sensing material in the emission zone sensing device was Bis[2-(1isoquinolinyl-*N*)phenyl-C](2,4-pentanedionato-O2,O4)iridium(III) (Ir(piq)₂acac). Lifetime measurement was carried out by fixing the current of the devices. Luminance was traced to obtain lifetime data of the 4CzIPN devices.

3. Results and discussion

The devices studied in this work were the 4CzIPN doped DCzIPN and mCBP devices. The DCzIPN was a host having bipolar charge transport characterics and the mCBP was a unipolar host. The two host materials were selected to analyze the cause of lifetime dependence on dopant concentration of 4CzIPN. Molecular structures of organic materials used in the device fabrication are shown in Fig. 1. Energy levels of the DCzIPN, mCBP and 4CzIPN were compared to estimate the light emission process of the DCzIPN:4CzIPN and mCBP:4CzIPN devices. As can be seen from Fig. 2, the HOMO/LUMO levels of 4CzIPN and DCzIPN were -6.08/ -3.06 eV and -6.26/-3.56 eV. The HOMO and LUMO level differences between the two materials were 0.18 eV and 0.40 eV, respectively, suggesting that energy transfer would dominate the light emission of the DCzIPN:4CzIPN device. In the case of mCBP:4CzIPN, the LUMO level difference between mCBP and 4CzIPN was as high as 2.4 eV, implying that electron trapping would be the main light emission process.

The different electron trapping effect in the two devices had an effect on the current density of the mCBP and DCzIPN devices



Fig. 2. Energy level diagram of materials in the TADF OLED device.

according to 4CzIPN doping concentration, Fig. 3(a) is a current density-voltage graph of the mCBP:4CzIPN devices at 4CzIPN dopant concentrations of 10%, 20% and 30%. When the doping concentration of 4CzIPN was 10% in the mCBP:4CzIPN device, the 4CzIPN acted as a trap for the electron carrier because of large LUMO level difference between 4CzIPN and mCBP. The trapped electrons by 4CzIPN should be carried by the 4CzIPN molecules, but the large distance between the 4CzIPN molecules at 10% doping concentration hindered the electron transport and reduced the current density. The molecule to molecule distance is reduced at high doping concentration, which increased the current density at 20% and 30% doping concentrations. The current density of the DCzIPN:4CzIPN device was also examined according to 4CzIPN doping concentrations. As shown in Fig. 3(b), since the electron trapping by 4CzIPN of the DCzIPN:4CzIPN device would not be so strong as that of the mCBP:4CzIPN device, the doping concentration dependence of the current density was not significant. Luminance of each device was in line with the current density.

Lifetime of the mCBP:4CzIPN devices with different 4CzIPN doping concentrations was measured to check doping



Fig. 1. Chemical structure of emitter materials used in the device fabrication.

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