

# Efficient exciplex emission from intramolecular charge transfer material



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## ABSTRACT

New exciplexes formed between a typical intramolecular charge transfer (ICT) material (bis[4-(9,9-dimethyl-9,10-dihydroacridine)phenyl]sulfone (DMAC-DPS)) and a series of electron donor and acceptors in donor:acceptor system have been systematically demonstrated. It is found that such ICT materials could form exciplex with both standalone electron donor and acceptor materials with itself as acceptor and donor components, which is based on the presence of both donor and acceptor species in the ICT material. The emission spectra of exciplex OLEDs based on ICT materials could be regularly tuned ranging from blue to yellow color by changing energy level alignment between ICT and standalone donor/acceptor materials. Among these exciplexes, DMAC-DPS:PO-T2T combination offered the highest exciplex EL performance, with its peak external quantum efficiency, luminance and current efficiency of 9.08%, 35,000 cd/m<sup>2</sup> and 30 cd/A, respectively. On the other hand, we also found that the exciplex efficiency was insensitive with the weight ratio between ICT material and acceptor, which means 'doping' of ICT material into the acceptor. Our finding extend the usage and selection scope of the TADF material.

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## 1. Introduction

Organic light emitting diodes (OLEDs) are anticipated as one of the next generation energy-saving and eco-friendly light sources and flat panel displays because of their merits like wide-view-angle, low driving voltages, and high efficiencies [1–3]. The traditional OLEDs are mostly based on fluorescent and phosphorescent materials and still suffer from critical disadvantages like low efficiency of fluorescent device and large efficiency roll-off and high application cost of phosphorescent OLED [4–6]. In view of the disadvantages in fluorescent and phosphorescent OLEDs, great efforts have been devoted to develop highly efficient noble metal free materials. Among these efforts, a new mechanism named thermally activated delayed fluorescence (TADF) was raised by the group of Adachi and has been attracted a lot of research interests [7,8]. OLEDs based on TADF materials are considered to have unit internal quantum efficiency because the randomly formed 75% triplet excited states can up convert into the singlet excited state and contribute to light emission [9,10]. In this mechanism, the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) of an emitting material

should be sufficiently separated to minimize the energy difference between singlet and triplet excited states [11]. This small energy difference, designed comparable with the  $K_B T$  ( $K_B$  the Boltzmann constant,  $T$  for temperature) at room temperature, enables efficient thermally activated reverse intersystem crossing of triplet states to the singlet state. Both intra molecular charge transferred (ICT) materials and exciplexes formed between electron donors and acceptors are recognized with TADF character [12–15]. Up till now, a great deal of ICT materials and exciplexes were proved have high emitting efficiencies [11,16–20]. Among which, the most impressive OLEDs based on blue and green ICT emitters performed high EQE of 19.5% and near 30% respectively [11,13,21]. However, the properties of the ICT materials in either Photoluminescence (PL) or electroluminescence (EL) situations are still not sufficiently explored. The ICT materials in EL condition can easily form exciplexes with the host materials if the energy levels are not located in the energy gap of the host materials. These exciplex emissions would damage the color purity of the ICT emission and this effect has not been paid enough attentions. On the other hand, the ICT materials have been proved of huge potential to be used in undoped (layered) devices where the ICT materials could also form exciplexes with transporting materials [22–24]. Therefore, it is of great significance to investigate the exciplex behaviors of ICT materials because it can improve the efficiency and quality of ICT

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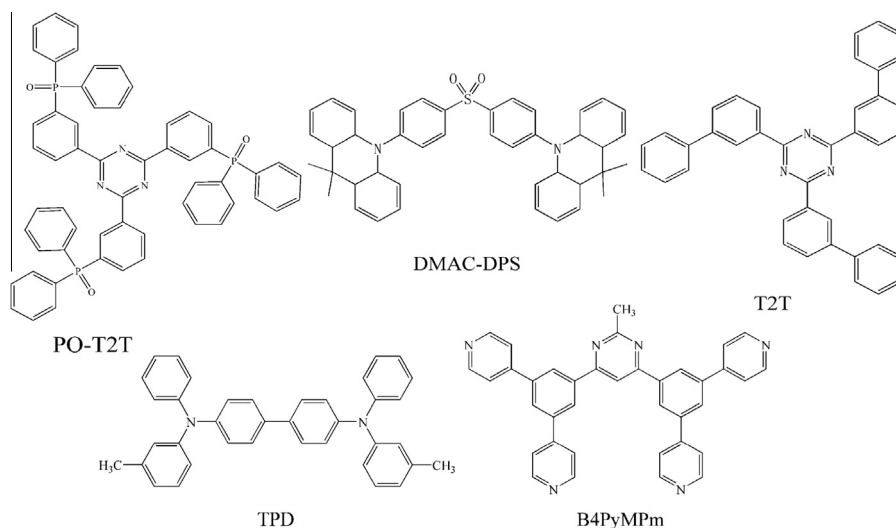
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emission and give new insight of the ICT materials. What's more, it can also be used as primary active layer in device if the ICT material based exciplex is efficient.

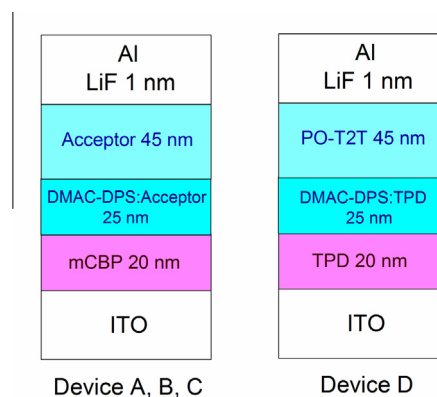
## 2. Experimental section

To study the interaction between ICT materials and other electron donors and acceptors, we chose bis[4-(9,9-dimethyl-9,10-dihydroacridine)phenyl] sulfone (DMAC-DPS) as a typical ICT material because DMAC-DPS is one of the most famous blue ICT dopant with high EQE of near 20% [13]. Generally, a highly efficient TADF material have a complete separation between its HOMO and LUMO electron cloud, which are mostly populated at the electron donating and electron accepting fragments respectively [11]. As a result of this separation, the ICT material can form new exciplexes with both electron donor and acceptor materials. The HOMO and LUMO levels of DMAC-DPS are located at 2.9 eV and 5.9 eV respectively [13]. These modest energy levels enable us to design serial devices with consecutive individual emitting spectra. Three electron acceptors, including 2,4,6-tris (biphenyl-3-yl)-1,3,5-triazine (T2T), bis-4,6-(3,5-di-4-pyridyl-phenyl)-2-methylpyrimidine (B4PyMPm) and PO-T2T and one electron donor N,N'-diphenyl-N,N'-bis(3-methylphenyl)-1,1'-biphenyl-4,4'-diamine (TPD) were chosen to form exciplex with DMAC-DPS to modulate the emitting spectra [18,25,26]. The molecular structures of DMAC-DPS, T2T, B4PyMPm, PO-T2T and TPD are given in Fig. 1.

OLED devices are fabricated using pre-cleaned indium tin oxide (ITO) coated glass substrates with a sheet resistance of  $15 \Omega \text{ cm}^{-2}$  and ITO thickness of 150 nm. They were patterned so that the OLED devices had a pixel size of about  $12 \text{ mm}^2$ . The small molecule and cathode layers were thermally evaporated using the multiple-source organic molecule deposition method. To fabricate the device, the devices were prepared in vacuum at a pressure of  $5 \times 10^{-4} \text{ Pa}$ . The  $\text{MoO}_3$ , hole-transporting materials mCBP and TPD, electron-transporting material T2T, B4PyMPm, and PO-T2T were thermally evaporated at a rate of  $0.1 \text{ nms}^{-1}$ . Blends of DMAC-DPS:acceptor and TPD:DMAC-DPS were deposited at a rate of  $0.1\text{--}0.2 \text{ nms}^{-1}$ . After the deposition of organic film, a 0.8 nm layer of LiF and a 150 nm layer of aluminum were thermally evaporated onto the organic surface. The four device structures are indicated in Fig. 2.



**Fig. 1.** Molecular structures used in this work. PO-T2T, T2T and B4PyMPm were used as electron acceptor and electron transporting layer. TPD was used as electron donor and hole transporting layer.



**Fig. 2.** Device architectures used in this work. The structure of device A, B and C is: ITO/ $\text{MoO}_3$  (3 nm)/mCBP (20 nm)/DMAC-DPS:acceptor (25 nm)/acceptor (40 nm)/LiF (0.8 nm)/Al with acceptor as T2T, B4PyMPm and PO-T2T; The structure of device D is: ITO/ $\text{MoO}_3$  (3 nm)/TPD (20 nm)/TPD:DMAC-DPS (25 nm)/PO-T2T (45 nm)/LiF (0.8 nm)/Al.

PL spectra of all the films were measured using Hitachi F7000 spectrometer. PL decay times were determined by FLS980 Fluorescence spectrometer. Current–voltage–brightness characteristics were measured by using a Keithley source measurement unit (Keithley2400) with a calibrated silicon photodiode. The EL spectra were measured by a Spectra scan PR650 spectrophotometer. All the EL measurements were carried out at room temperature under ambient condition.

## 3. Results and discussion

To achieve high emitting efficiency, the ICT materials with TADF character are mostly doped into a host material to reduce its self-quenching events [27]. Usually, the HOMO and LUMO of an ICT dopant should lower and higher than or equal to that of the host. If this requirement is not satisfied, exciplex formed between the host and the ICT dopant would appear [28]. This phenomenon can also be observed in the traditional fluorescent emitter case, like  $\text{AlQ}_3$  with m-MTDATA [29]. This unexpected exciplex in the traditional fluorophore case were firstly regarded as a negative effect that would reduce the quality of emitting color [30]. But

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