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#### ABSTRACT

The influence of the host molecules on the photoluminescent (PL) and electroluminescent (EL) properties of organic light-emitting diode (OLED) emitters showing efficient thermally-activated delayed fluorescence (TADF) has yet to be investigated in detail. Here we demonstrate that the choice of host can cause large variations in the PL quantum yield ( $\Phi_{PL} \sim 15-70\%$ ) and delayed PL transient decay ( $\tau_{del} \sim 2-70$  ms) of a spiro-acridine-based TADF guest. We show that the effect of exciplex formation on  $\Phi_{PL}$  must be considered even at low concentrations of the TADF guest. Using the same TADF guest but changing the host layer, we are able to greatly vary the PL transient decay time from ~4 to ~70 ms while maintaining a high  $\Phi_{PL} \sim 70\%$ , which can lead to new applications. Detailed spectral characterization during PL decay reveals a gradually increasing singlet-triplet energy gap ( $\Delta E_{ST}$ ) as the origin of these observations. The time-varying  $\Delta E_{ST}$  is explained based on dipole interactions between the host and guest molecules. Finally, PL and electrical considerations for host selection are discussed based on the performance of OLED devices.

#### 1. Introduction

Thermally-activated delayed fluorescence (TADF) occurs in organic aromatic molecules in which the lowest singlet-triplet energy splitting ( $\Delta E_{ST}$ ) is small enough to allow thermal activation from triplet to singlet excited states via reverse intersystem crossing (RISC), resulting in delayed fluorescence [1]. To promote highly efficient RISC,  $\Delta E_{ST}$  should be less than a hundred meV [2]. However, most conventional molecules possess a large  $\Delta E_{ST}$  and low RISC efficiency, so the application of TADF to emitter materials saw limited interest. Recently, by tailoring the

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http://dx.doi.org/10.1016/j.orgel.2014.05.027 1566-1199/© 2014 Elsevier B.V. All rights reserved. molecular design to achieve a small  $\Delta E_{ST}$ , we have demonstrated many classes of highly efficient TADF emitters. These new emitters are a promising cost-effective alternative to the best phosphorescent organometallic emitters that are widely used today in organic light-emitting diodes (OLEDs) for displays and illumination [2,3].

When neat films of the emitter molecules are used as the emitter layer in OLEDs, performance is often poor because of concentration quenching, unwanted excimer emission, aggregation, and other effects [4]. These losses can often be reduced and performance improved by doping the emitter molecules, also called the guest, into a film comprised of appropriate host molecules. The design of host–guest systems demonstrating highly efficient delayed luminescence originating from triplet excitons has been shown to require careful consideration of the intermolecular interactions between host and guest molecules [5]. While the effects



of host molecules on the photophysical processes of phosphorescent compounds have been studied thoroughly in the past ten years [6], only a limited number of similar studies concerning TADF emitters exist [7].

The spiro-acridine compound 10-phenvl-10Hspiro[acridine-9,9'-fluorene]-2',7'-dicarbonitrile (ACRF). which we recently reported as a TADF emitter, is an ideal candidate for investigating how the host can affect the TADF behavior of guest molecules because the triplet excitons in ACRF have rather long transient lifetimes [8]. When ACRF is doped into a small-molecule host layer, intense TADF can be observed in an oxygen-free environment, and an external quantum efficiency ( $\eta_{ext}$ ) of ~10% was achieved by using the doped films in OLEDs [8]. On the other hand, neat films of ACRF result in a very weak TADF component due to the concentration quenching-effect [8]. Thus, the choice of host can greatly impact the emission properties, and understanding the interaction between guest and host molecules is important for the further development of devices based on TADF.

In this study, we investigate how the host layer affects the photoluminescence (PL) properties of the TADF emitter ACRF in solid-state films. We carefully analyze the host dependence of the TADF emission in ACRF based on transient radiative decays, absolute PL quantum yields ( $\Phi_{\rm PI}$ ), and changes in PL spectra. We show that the host can impact both  $\Phi_{\rm PI}$  and the lifetime of the delayed emission, and we discuss the various mechanisms that can account for these changes. We propose a mechanism to explain how some hosts can lead to much longer delayed lifetimes based on shifts in the emission spectra and their temperature dependence. Furthermore, we study the OLED performance of these host-guest films as emitters and the factors affecting  $\eta_{ext}$ . Through these comprehensive PL and electroluminescence (EL) studies, we clarify some of the criteria for the selection of host molecules to enhance and tailor specific properties of TADF.

#### 2. Results and discussion

#### 2.1. Photoluminescence lifetime of ACRF:host layers

To explore the host-guest interactions, we used a series of small-molecule host matrices with a variety of chemical structures and energy gaps. Similar to when phosphorescent emitters are used, the T<sub>1</sub> level of the host molecule  $(T_1^H)$  should be higher than that of the guest  $(T_1^G)$  to prevent back-energy transfer from the TADF guest to the host [9]. Therefore, hosts with  $T_1^H$  close to or higher than  $T_1^G$  were chosen. The host molecules used in this study were triphenyl-(4-(9-phenyl-9Hfluoren-9-yl)phenyl)silane (TPSi-F), which we used as an efficient host in our previous study [8], 1,3,5-tri(m-pyrid-3-yl-phenyl)benzene (TmPyPB) [10], 2,4,6-tris(biphenyl-3-yl)-1,3,5-triazine (T2T) [11], 2,6dicarbazolo-1,5-pyridine (PYD2) [12], 9-(4-tert-butylphenyl)-3,6-bis(triphenylsilyl)-9H-carbazole (CzSi) [13], and bis(2-(diphenylphosphino)phenyl)ether oxide (DPEPO) [14]. The chemical structures and  $T_1$  levels are shown in Fig. 1. The T<sub>1</sub> levels were determined from the onset of the phosphorescence spectra in neat films for the hosts (Fig. S1) and in a 2-methyltetrahydrofuran solution for ACRF (detailed experimental methods are in the Supplementary information (SI)). The molecules TmPyPB, TPSi-F, and T2T have very similar  $T_1^H$  of 2.80, 2.84 and 2.86 eV, respectively, which are slightly higher than that of ACRF ( $T_1^G = 2.62 \text{ eV}$ ). The other host molecules possess higher  $T_1$  levels. The  $T_1^H$ is 3.03 and 3.09 eV in PYD2 and CzSi, respectively, and DPEPO has the highest  $T_1^H$  of 3.38 eV.

Figure 2 shows the emission decay profiles of the various host–guest thin films measured under vacuum over a time range of 50 ms. The doping concentrations of the guest molecules in the host layers were 2 or 6 wt% to exclude any effect from concentration quenching between ACRF molecules, as verified by separate  $\Phi_{PL}$  measurements. Remarkable differences in the length of the decay were



**Fig. 1.** The molecular structures and T<sub>1</sub> levels of ACRF and the hosts used in this study. The error bars of ±0.02 eV represent the uncertainty in determining the onset of the phosphorescence spectra.

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