

# Ultra-Efficient Fourth-Generation pTSF OLED Devices and Products

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## Abstract

*Phosphor-assisted thermally activated delayed fluorescence sensitized fluorescence (pTSF) organic light-emitting diodes (OLEDs) have attracted much attention and have been regarded as the fourth-generation OLED technology since its invention in 2022. In this work, through material optimization, device optimization, and process optimization, we fabricate a series of green pTSF OLED based devices and products, which realize high efficiency, long lifetime, and high color purity, manifesting the bright further of pTSF technology in mass production.*

## Author Keywords

multi-resonance; pTSF; energy transfer; high efficiency; high color purity

## 1. Introduction

Recently, organic light-emitting diode (OLED) has already been widely used in products, including wearable devices, mobile phones, laptops and televisions. As artificial intelligence (AI) becomes increasingly widely used in these electronic devices, the power consumption of their constituting hardware, including screens, needs to be further reduced. Besides, most OLED products nowadays adopt the Digital Cinema Initiatives Protocol 3 (DCI-P3) color gamut, which covers nearly 45% of the area of Commission Internationale de L'Eclairage 1931 (CIE1931) color space. How to further broaden the color gamut of OLED products to enhance the display quality has become another critical problem. To solve the above two problems of OLED products, materials and devices are the keys.

Fluorescent OLEDs adopting fluorescent emitters are regarded as the first-generation OLED devices [1]. They usually exhibit high color purity and long lifetime. However, extremely low external quantum efficiency (EQE) hinders their application in industry. To boost the efficiency, Ma et al. and Forrest et al. designed second-generation phosphorescent materials and fabricated phosphorescent OLEDs (PhOLEDs) in 1998 [2, 3]. With the aid of heavy atoms such as iridium and platinum, PhOLEDs can harvest 100% excitations to emit light, resulting in a four-fold increase in EQE compared to fluorescent devices. However, heavy atoms will increase the cost of PhOLEDs. Besides, deep-blue/blue PhOLEDs suffer from short lifetime, low efficiency, and severe efficiency roll-off. In 2012, Adachi et al. proposed the third generation of thermally activated delayed fluorescence (TADF) materials and devices [4]. Through skillful material design, TADF materials can capture 100% excitons through reverse internal system crossing (RISC) process and emit light. Albeit that TADF devices have achieved nearly 40% EQE in blue, green, and red devices, poor color purity and short device lifetime still make them unable to be used in products [5].

In 2014, Duan et al. proposed thermally activated delayed fluorescence sensitized fluorescence (TSF) OLEDs, which combine the advantages of TADF materials and fluorescent

materials [6]. In this system, TADF materials act as hosts and sensitizers to transport carriers and harvest excitons, and fluorescent materials act as emitters to emit light. As a result, TSF devices can realize high efficiency and long lifetime, simultaneously. Since then, various red, green, and blue devices have been developed. In 2022, Duan and Zhang further modified TSF OLED and proposed phosphor-assisted TSF (pTSF) OLED [7]. With the aid of additional assisted phosphors, pTSF OLEDs exhibit higher efficiency, longer lifetime, and higher luminance than TSF, TSP (TADF-sensitized phosphorescence), and PSF (phosphor-sensitized fluorescence) OLEDs, making pTSF OLEDs become the ultimate OLED technology in regarding of overall performance (Figure 1). Since then, we have been cooperating with Duan and developed a series of pTSF-related materials and devices [8, 9].

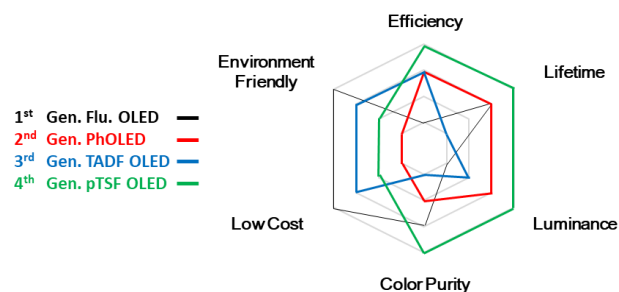


Figure 1. Comparison of fluorescent, phosphorescent, TADF, and pTSF OLEDs

## 2. Experimental Results and Discussion

**2.1 Material Optimization:** In constructing the key emitting-layers (EMLs) of pTSF OLEDs, three major materials should be taken into consideration: TADF hosts, assisted phosphors, and fluorescent emitters. Among them, TADF hosts will be responsible for carrier transport, charge recombination, and exciton utilization. Compared with traditional single molecular TADF hosts, the singlet-triplet energy gap ( $\Delta E_{st}$ ) of exciplex TADF hosts approaches zero, which can facilitate the RISC process and result in higher efficiency. Besides, exciplex hosts contain both p-type and n-type components. By adjusting the fraction of two components, the charge balance of exciplex hosts can be precisely tuned to achieve better efficiency and lower driving voltage. Based on the above concepts, we verified the performance of pTSF OLEDs employing two different kinds of hosts. As presented in Table 1, they exhibit similar emission peaks and full width at half maxima (FWHM). However, compared with the reference device using non-TADF host GH1, the pTSF device using exciplex TADF host GH2 exhibited 6% higher current efficiency (CE) and 12% longer LT95 (lifetime to the initial luminance of 95%). Additionally, the driving voltage ( $V_d$ ) of GH2 based device decreased by 0.2V, indicating more efficient charge injection and more balanced charge transport.

**Table 1.** Properties of pTSF OLEDs adopting different kinds of hosts at 10mA/cm<sup>2</sup>

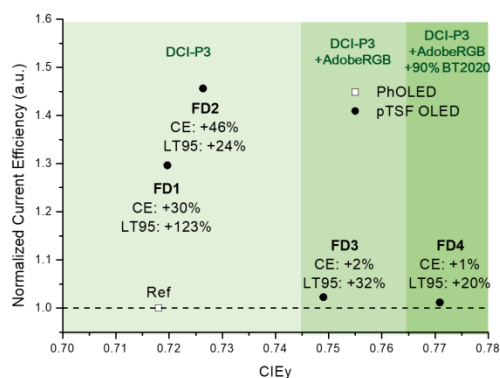
EML	Main Peak (nm)	FWHM (nm)	CE	V <sub>d</sub> (V)	LT95
GH1: PD1(5%): FD1(0.5%)	536	23	Ref.	Ref.	Ref.
GH2: PD1(5%): FD1(0.5%)	536	23	+6%	-0.2	+12%

For assisted phosphors, they will receive the energy of unused triplets of hosts through Dexter energy transfer (DET) routes and transfer their energy to the fluorescent emitters through ultrafast Förster energy transfer (FET) routes. Additionally, the external heavy atom (EHA) effect of assisted phosphor can also boost the RISC process of the exciplex host and further enhance the exciton utilization [10]. Here, we selected two different phosphorescent materials to construct pTSF OLEDs (Table 2). The CE of these two devices is insensitive to the assisted phosphors, indicating efficient energy transfer in pTSF devices. However, these two pTSF OLEDs exhibit distinct lifetime, which is similar to their corresponding PhOLEDs. Thus, selecting a stable assisted phosphor is one of the keys to constructing high-performance pTSF OLEDs.

**Table 2.** Properties of PhOLEDs and pTSF OLEDs adopting different assisted phosphors at 10mA/cm<sup>2</sup>

EML	Main Peak (nm)	FWHM (nm)	CE	V <sub>d</sub> (V)	LT95
GH2: PD1	534	24	Ref.	Ref.	Ref.
GH2: PD2	533	25	+14%	-0.1	-40%
GH2: PD1: FD1	536	23	Ref.	Ref.	Ref.
GH2: PD2: FD1	536	23	+3%	-0.1	-33%

As for fluorescent emitters, they will receive energy from both exciplex hosts and assisted phosphors through FET. Compared with traditional fluorescent materials, multi-resonance (MR) type fluorescent emitters exhibit higher photo luminance quantum yields (PLQYs), higher color purity, and better stability, making them the perfect emitters for pTSF OLEDs. Through fluorescent dopant design and selection, we fabricated a series of pTSF devices with CIE<sub>y</sub> coordinates ranging from 0.72 to 0.77 by using fluorescent emitters, namely FD1~FD4, to fulfill the requirement of DCI-P3 and AdobeRGB color gamut (Figure 2). Among them, FD4 based pTSF OLED could even satisfy the requirement of 95% BT2020 color gamut for green emission. In addition, all pTSF OLEDs exhibit higher CE and lifetime than the currently used reference PhOLED, manifesting their great potential for further application in mass production lines.

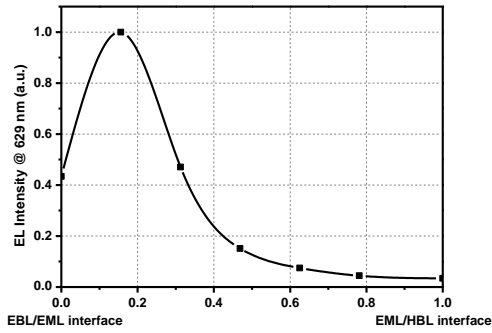
**Figure 2.** Comparison of the reference PhOLED and pTSF OLEDs employing different fluorescent emitters

**2.2 Device Optimization:** We further selected FD1 and optimized the device performance. As the concentration of PD1 increases, the CE slightly decreases, and the lifetime significantly increases (Table 3). As FD1 concentration increases, the efficiency of pTSF devices slightly increases, but the lifetime of pTSF devices dramatically decreases. This is because fluorescent dopant will trap holes, which will shorten the device's lifetime [11]. By taking both efficiency and lifetime into consideration, we selected GH2: PD1(5%): FD1(0.5%) as the EML for further investigation.

**Table 3.** Properties of pTSF devices with different PD1 and FD1 concentrations at 10mA/cm<sup>2</sup>

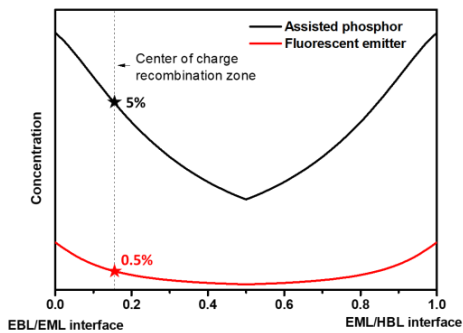
EML	Main Peak (nm)	FWHM (nm)	CE	V <sub>d</sub> (V)	LT95
GH2: PD1(5%): FD1(0.5%)	536	23	Ref.	Ref.	Ref.
GH2: PD1(3%): FD1(0.5%)	535	24	+0%	+0.0	-10%
GH2: PD1(7%): FD1(0.5%)	536	24	-2%	+0.0	+13%
GH2: PD1(5%): FD1(0.3%)	536	25	-5%	+0.0	+10%
GH2: PD1(5%): FD1(0.7%)	536	23	+2%	+0.1	-12%

We further explored the exciton generation zone of pTSF OLEDs. By inserting an ultra-thin red phosphorescent layer in different positions of EML and recording its red emission peaking at 629 nm, we acquired the distribution of the exciton generation zone throughout the EML of the pTSF device. As shown in Figure 3, the charge recombination zone of green pTSF devices is located near the interface between the electron blocking layer (EBL) and EML, which is identical to our previous research on red pTSF OLEDs [12].



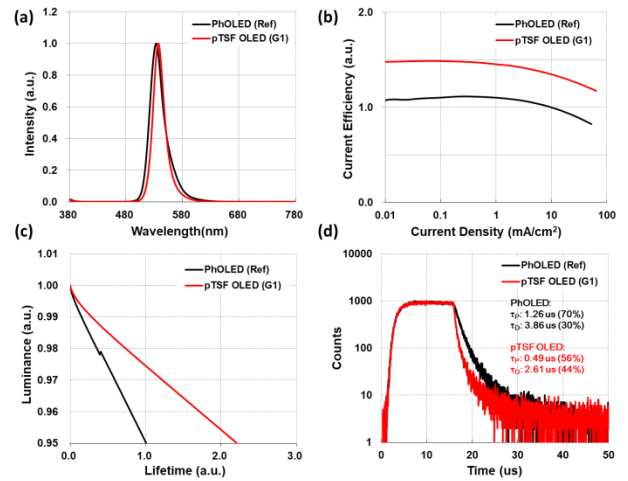
**Figure 3.** The exciton generation distribution throughout the EMLs of pTSF OLEDs.

**2.3 Process Optimization:** After acquiring the best device structure and the exciton generation distribution, we further modified the equipment and optimized the process for fabricating pTSF OLEDs in G6 mass production lines. By adjusting the position of evaporation sources, chamber height, and scan process, the best material distribution of EML in pTSF OLED is shown in Figure 4. The concentration of PD1 and FD1 at the center of the exciton generation zone in EML is 5% and 0.5%, which guarantees the best performance of pTSF OLEDs. Besides, compared with devices adopting uniform material distribution, the amount of PD1 and FD1 used in devices adopting optimized material distribution can be reduced by 10% and 8%.



**Figure 4.** Optimized PD1 (black) and FD1 (red) distribution throughout the EML.

To confirm the device performance, we further fabricated FD1 based green pTSF OLED in the mass production line and compared it with the currently used reference PhOLED. As can be seen from Figure 5, the CE and lifetime of pTSF OLED are 32% and 121% higher than the reference PhOLED. Such high efficiency and lifetime can significantly reduce power consumption and prolong the lifetime of screens. To gain a deeper insight into this phenomenon, we also measured the transient EL decay curves of pTSF OLED and PhOLED. Compared with the reference PhOLED with the prompt lifetime ( $\tau_p$ ) and delayed lifetime ( $\tau_D$ ) of 1.26  $\mu$ s and 3.86  $\mu$ s, respectively, the  $\tau_p$  and  $\tau_D$  of pTSF OLED significantly decrease to 0.49  $\mu$ s and 2.61  $\mu$ s (Figure 5d). Short exciton lifetime can significantly alleviate the triplet-triplet annihilation (TTA), triplet-singlet annihilation (STA), and triplet-polaron quenching (TPQ) phenomenon in pTSF OLEDs, which will enhance device efficiency and reduce efficiency roll-off [13].



**Figure 5.** (a) EL spectra, (b) CE, (c) lifetime (at the initial luminance of 100,000 nits), and (d) transient EL decay curves of PhOLED and pTSF OLED fabricated in G6 line

**2.4 Product and Performance:** Based on the above results, we eventually fabricated two high-performance green pTSF OLEDs based products, namely products A and B for proof of concept (Figure 6). Products A and B adopted FD1 and FD3 as fluorescent emitters to construct green pTSF OLEDs, respectively.



**Figure 6.** Pictures of products A (left) and B (right).

For comparison, we also select a standard mass-production product using green PhOLED as the reference. As shown in Table 4, compared with the reference product, product A adopting an ultra-efficient green pTSF device exhibits 12% lower power consumption. As for lifetime, product A exhibits more than 15% higher LT95 at a standard white luminance of 500 nits. Due to significantly reduced exciton lifetime and reduced efficiency roll-off, pTSF OLEDs are expected to exhibit an even longer device lifetime than PhOLEDs in high luminance [7]. Additionally, we carried out a series of reliability tests for 240 h, including high temperature operation (HTO) at 70 °C, high temperature storage (HTS) at 80 °C, 60°C+90% humidity test, and 85°C+85% humidity test. The results of the reference product and product A are comparable.

As for product B, with the aid of high color purity green emission, it can cover both 99.5% DCI-P3 color gamut and 99.5% AdobeRGB color gamut, and exhibits a much more vivid picture than product A (Figure 6). Additionally, product B exhibits 6% lower power consumption and 5% longer lifetime, manifesting the advantages of pTSF OLED in realizing high efficiency, long lifetime, and high color gamut OLED products. Besides, the reliability test results of product B are comparable to those of the reference product.

**Table 4.** Properties of the reference product, product A, and product B.

Product		Reference Product	Product A	Product B
Adopted Green OLEDs		PhOLED	pTSF OLED (FD1)	pTSF OLED (FD3)
Power Consumption @500nits		Ref.	-12%	-6%
LT95 @500nits		Ref.	≥+15%	≥+5%
Color Gamut in CIE1931	Coverage Fraction of DCI-P3	≥99.5%	≥99.5%	≥99.5%
	Coverage Fraction of AdobeRGB	-	-	≥99.5%
Reliability Test @240hrs	HTO	Ref.	Comparable	Comparable
	HTS	Ref.	Comparable	Comparable
	60 °C+90% Humidity Test	Ref.	Comparable	Comparable
	85 °C+85% Humidity Test	Ref.	Comparable	Comparable

### 3. Impact and Outlook

In this article, we introduced our recent advances in designing ultra-efficient fourth-generation pTSF OLED devices and products. Through skillful material optimization, device optimization, and process optimization, we finally fabricated two green pTSF OLED based products A and B, exhibiting low power consumption, long lifetime, and high color gamut, simultaneously. Our results manifest the practical application of pTSF OLEDs and opened a new era for the OLED industry. After the successful application of green pTSF devices, we are now working on developing ultra-efficient fourth-generation red and blue pTSF OLEDs. According to our current results, the bottleneck of red pTSF OLEDs is the lack of stable and efficient red MR emitters. Whereas for blue pTSF OLEDs, the bottleneck is the lack of stable blue assisted phosphors. We believe that, through strong cooperation with universities, materials manufacturers, and terminal customers, we can solve the bottlenecks soon.

### 4. Acknowledgments

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