Acceptor Interlocked Molecular Design for Solution-Processed Stable Deep-Blue TADF and Hyper Fluorescence Organic LED Enabling High-efficiency

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Abstract

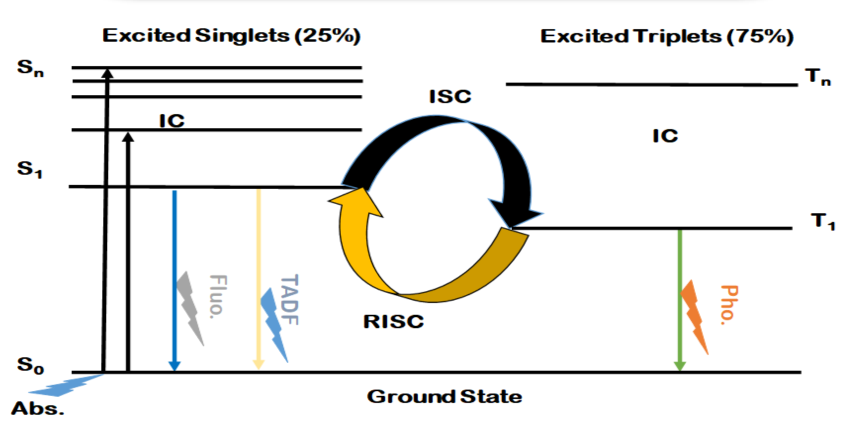
Solution processed deep-blue OLEDs with high external quantum efficiency and a long operational lifetime are constrained. In this context, we synthesized two TADF emitters utilizing new design strategy of twisted interlocked acceptor core integrated with carbazole (KCCz) and tert. butyl-carbazole (KCTBC) as donors for solution processed deep-blue TADF OLEDs. Twisting of acceptor core by two methyl groups resulted in complete separation of HOMO and LUMO along with cyanide group facilitate in generating low-lying triplet exited states as suggested by theoretical simulation. Combined effect of both resulted in tuning of emission in ultra-deep blue region through the efficient population of triplet excitons and concurrently RISC to produce highly efficient devices. A doped device based on KCTBC showed EQE_{max} of 9.0% along with low Efficiency roll-off with long operational device half lifetime of 72 minutes at initial brightness of 1,000 cd m-2, and CIE coordinates of (0.17, 0.13). In addition, with 12.5 wt% of 4CzFCN as assistant dopant/co-host to enhanced the performance of the KCTBC based device with an EQE_{max} of 13.9% and CIE coordinates of (0.18, 0.13). Further, a high-efficiency warm white OLED adopting the TADF hybrid approach is realized with EQE_{max} of 9.0 %.

Introduction

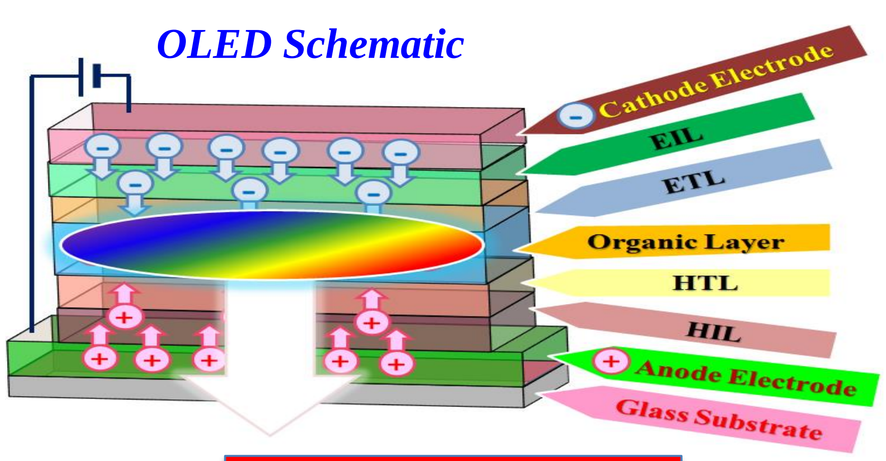
- TADF materials are gaining much interest due to vast applications in the fields of organic light-emitting diodes because of their 100% IQE.
- Organometallic counterparts need to be replaced with pure organic TADF materials which is cost effective, non toxic and have numerous structural modification scope.
- Full-color displays and white OLEDs require effective deep-blue emitters.
- Deep-blue emitters are challenging to build due to their large band gaps, uneven carrier transport, and poor energy level matching with other OLED layers.

Characteristics of deep-blue TADF Emitter for OLED applications

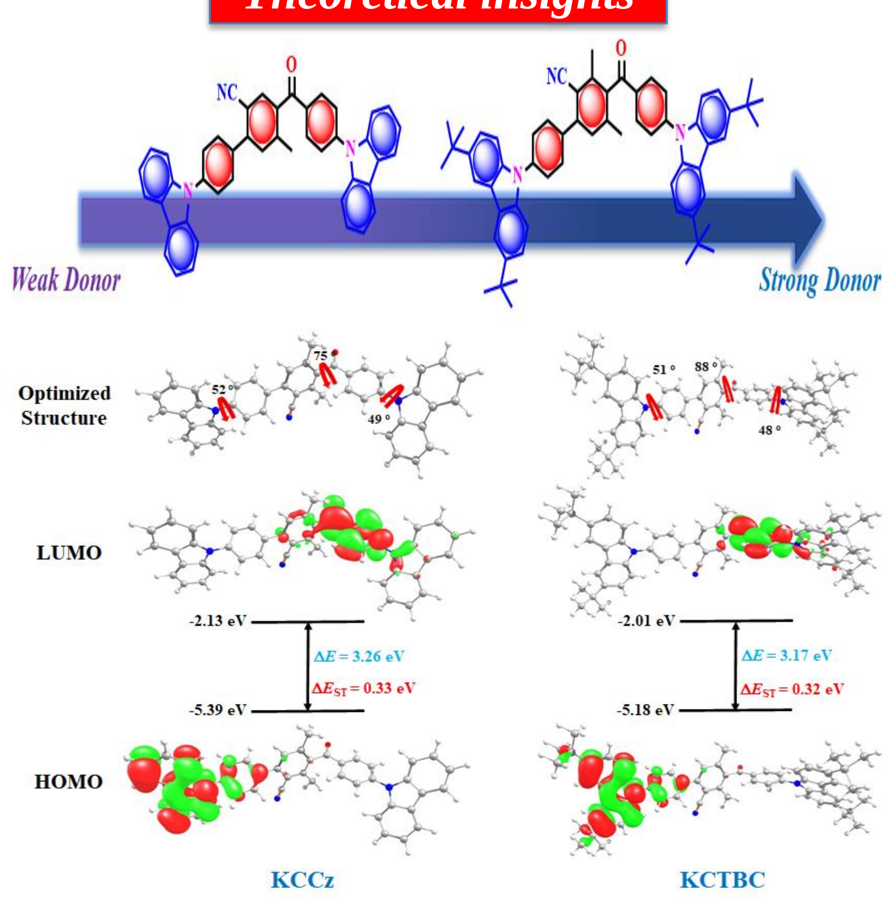
- ✓ *Twisted D-A molecular geometry.*
- ✓ *Should have small ΔE_{ST} for effective RISC.*
- ✓ *HOMO-LUMO should be well separated.*
- ✓ *Large band-gap (>3.0 eV)*
- ✓ *Should have good charge transfer property.*
- ✓ *Should have high PLQY.*



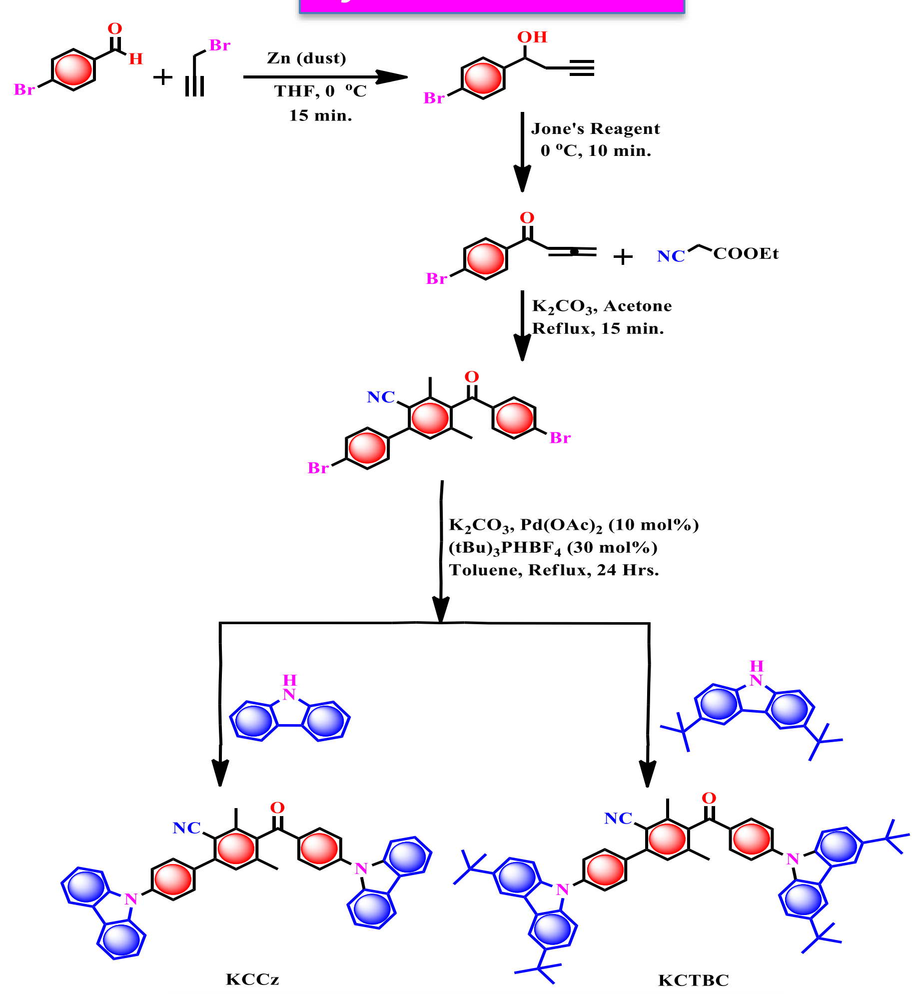
OLED Schematic



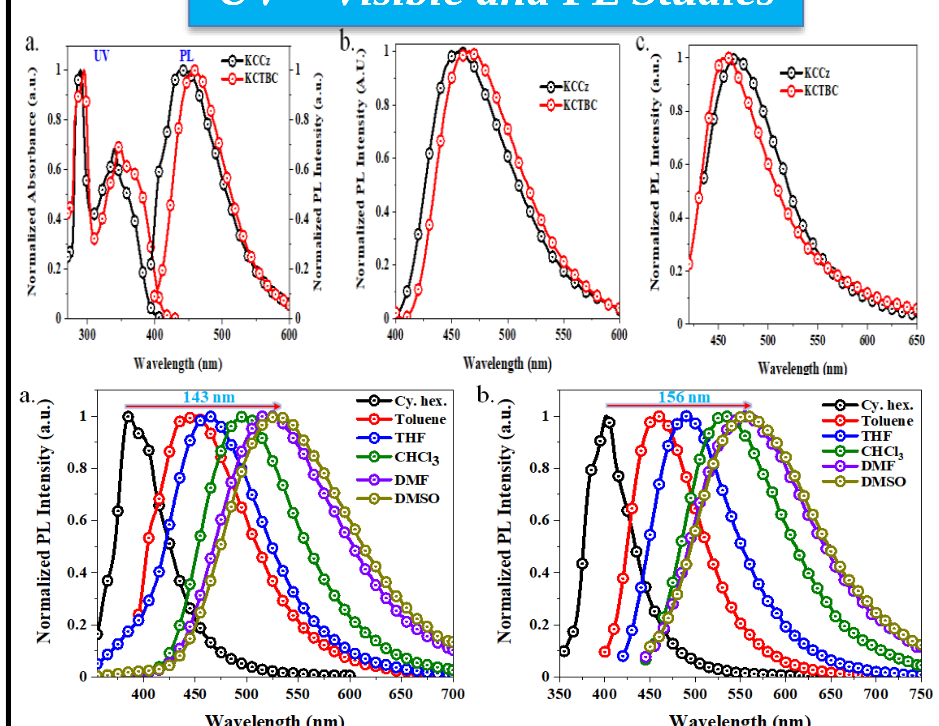
Theoretical insights



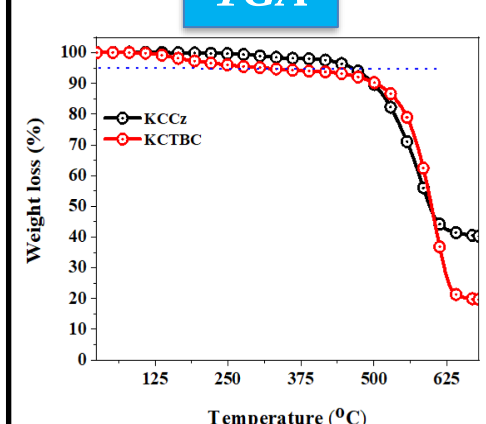
Synthetic scheme



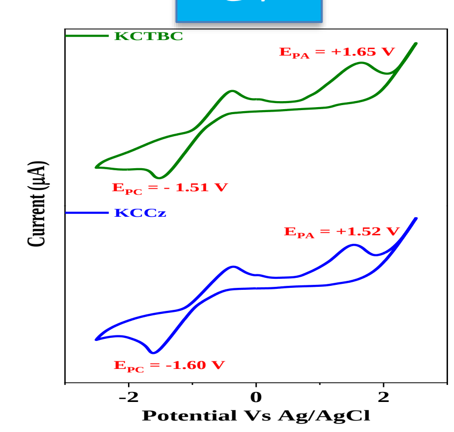
UV – Visible and PL Studies



TGA



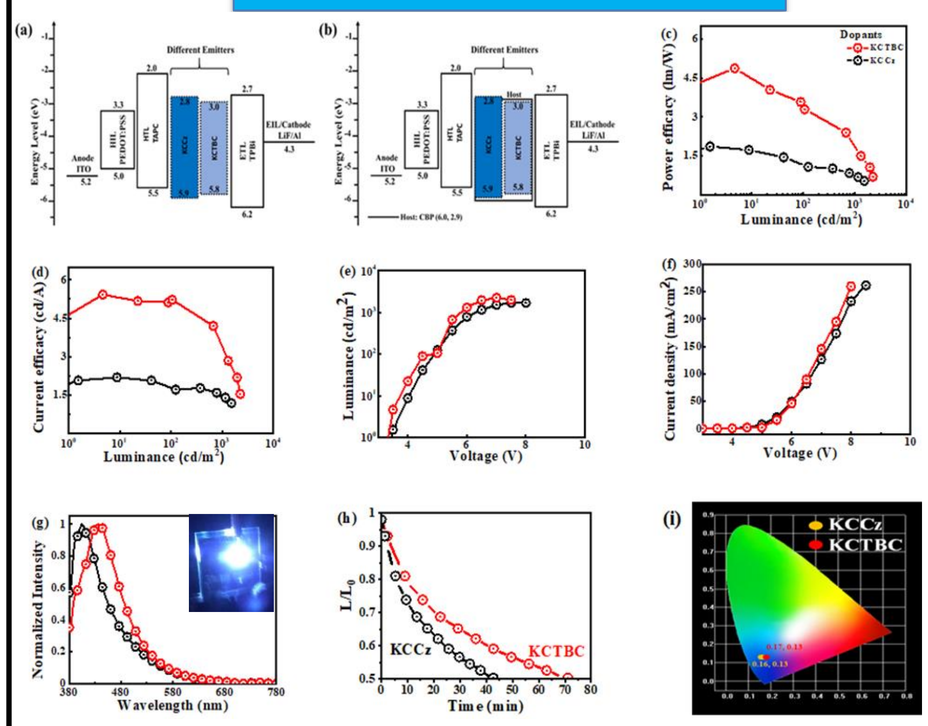
CV



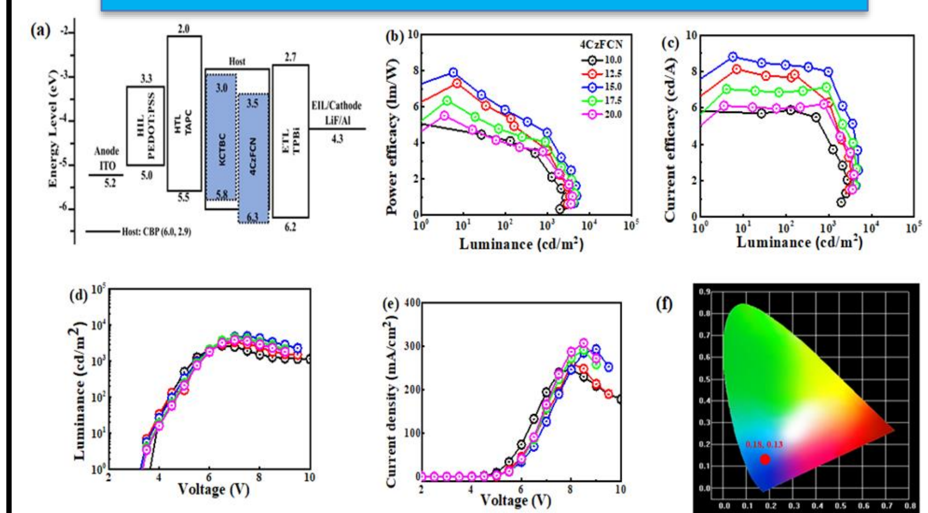
Emitter	λ_{abs} (nm) Sol[a]	λ_{em} (nm) (Sol[a]/film[b]/solid[c])	HOMO/LUMO (eV)[d]	E_{H-L} (eV)[e]	Quantum yield[fgh] (%)	τ_p/τ_d (ns[i]/ms[j])	S_1/T_1 (ΔE_{ST}) (eV)[k]	T_{onset} (°C)[l]
KCCz	290, 340	443/456/468	-5.92/-2.80	3.12	7.09/60.0/48.2	2.62/1.5	3.0/2.8 (0.20)	470
KCTBC	295, 348	459/465/457	-6.05/-2.89	3.16	16.8/70.0/78.5	1.86/0.89	2.96/2.74 (0.22)	324

[a]For toluene solution, [b]For neat film [c]For solid powder. [d]HOMO/LUMO and [e]Optical band gap (E_{H-L})measured from the CV. Absolute PLQY of the [f]neat films, [g]CBP doped film and in [h]$CHCl_3$ measured using an integrating sphere at room temperature. [i]Prompt and [j]delayed fluorescence decay time of thin-films KCTBC doped in CBP at 300 K. [k]Calculated from onset of fluorescence and Phosphorescence spectra [l]Temperature corresponds to 5% weight loss (T_{onset}).

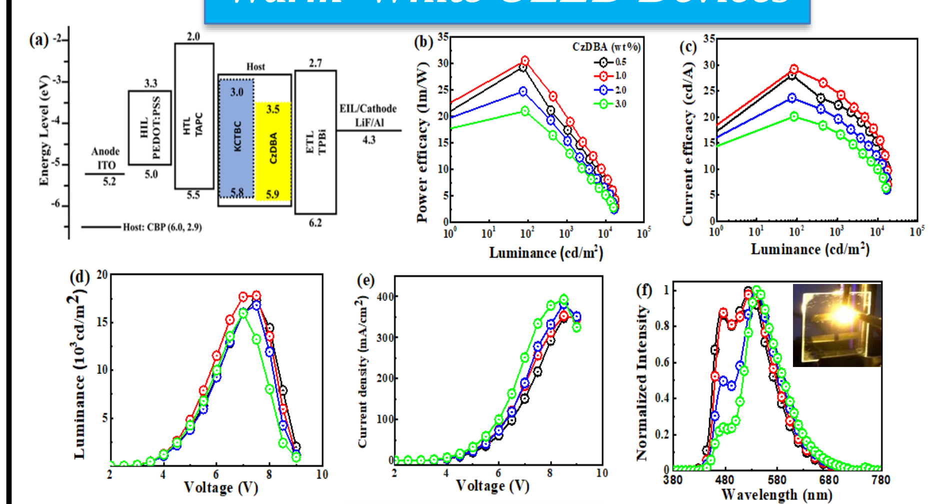
Deep-blue OLED Devices



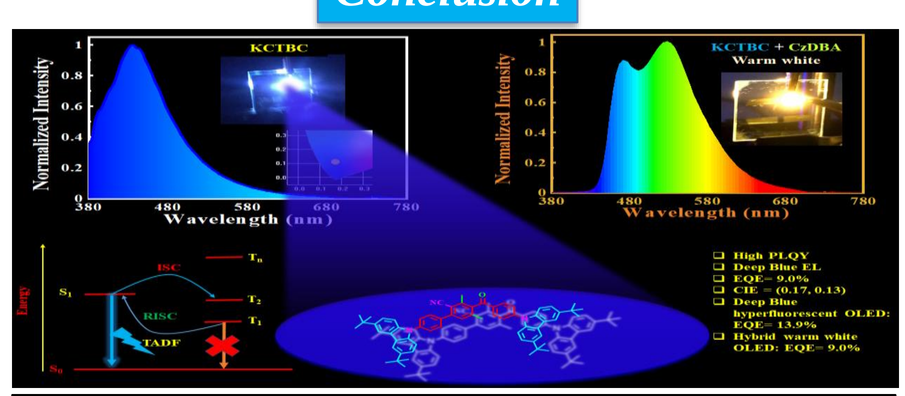
Hyperfluorescent Deep-blue Devices



Warm White OLED Devices



Conclusion



- ✓ Successfully designed, synthesized and characterized two new TADF deep-blue emitters.
- ✓ Theoretical and experimental study suggested small ΔE_{ST}.
- ✓ KCTBC based device showed better EL performances among the two.
- ✓ Solution processed cheap deep-blue (EQE_{max} = 9%) as well as warm white OLEDs (EQE_{max} = 9%) were fabricated.
- ✓ A deep-blue hyperfluorescent OLED based on KCTBC doped with 4CzFCN with EQE_{max} = 14% was also fabricated.

References

1. .Li, Y.; Gecevicius, M.; Qiu, *J. Chem. Soc. Rev.* **2016**, 45, 2090.
2. Kabe, R.; Notsuka, N.; Yoshida, K.; Adachi, C. *Adv. Mater.* **2016**, 28, 655.
3. Chen, W.C.; Yuan, Y.; Zhu, Z.L.; Ni, S.F.; Jiang, Z.Q.; Liao, L.S.; Wong, F.L.; Lee, C.S. *Chem.Commun.* **2018**, 54, 4541.

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