

# **Navigating CIE Space for Efficient TADF Downconversion WOLEDs**

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## **ABSTRACT**

**High efficiency orange and green emitting perylene dyes have been synthesised and dispersed in an inert polymer host to form an optical downconversion layer. To avoid dye aggregation and allow controlled colour tuning, this layer was deposited in multiple low-concentration spin-coating steps, directly on top of a high performance blue thermally activated delayed fluorescence (TADF) organic light emitting diode (OLED). The orange downconversion layer partially absorbs the blue OLED emission, while emitting a complementary orange to give white light. However, as energy transfer between the TADF and perylene down converter is based on emission and reabsorption, absorptive filtering of the blue OLED emission band necessitates the inclusion of an additional green-emitting perylene top-layer to achieve optimal white balance. The optimised white OLED fabricated in this way displayed excellent white colour balance (CIE  $x, y$ ; 0.33, 0.33) with perfect stability, good colour rendering (CRI 80), and a high maximum efficiency (maximum EQE 17.2%) with minimal losses compared to the base blue OLED. This approach is widely applicable for generating white emission from any kind of blue OLED, and is compatible with a wide range of downconverting dyes and host materials.**

## **INTRODUCTION**

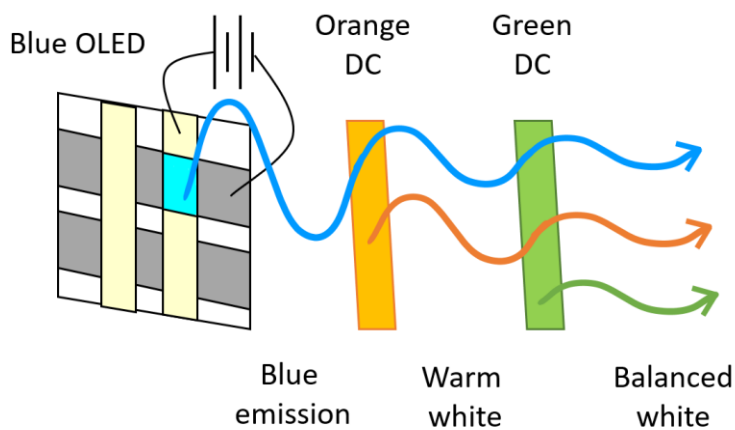
Organic Light Emitting Diodes (OLEDs) are highly attractive for future innovation in the lighting and display industries. This appeal is due to their potential to deliver low cost materials and fabrication processes, and their compatibility with mechanically flexible substrates that enable entire new classes of consumer products. However, while monochromatic OLEDs are already well-developed, there are multiple competing strategies to achieve efficient white OLEDs (WOLEDs).

For display applications, individually addressable red, green, and blue pixels are required to access a complete colour space. Lighting applications are generally less technically demanding, as they require only a single white colour to be generated – although at much higher practical brightnesses. This simpler colour requirement means that bicolour white emission, (such as orange/cyan mixtures, in comparison to tricolour red/green/blue) is often suitable, as long as the colour components are spectrally broad enough to ensure satisfactory colour rendering.

To achieve white emission in OLEDs, the two most commonly reported approaches are to deposit multiple emission layers (EMLs) for different component colours in the white mixture [1], or to co-deposit multiple chromophores in a single EML [2–4]. In the first approach, each emissive material produces electroluminescence independently. The white colour balance is then determined by the relative brightness of each EML section, which can be controlled, for example, by tuning their thicknesses or dopant concentration. In the second approach, direct electrical recombination in the EML is accompanied by short range energy transfer from the higher energy to lower energy chromophores via concentration dependent Förster or Dexter processes. As the proportion of emission from each colour component can no longer be tuned independently, the relatively simple device architectures of single EML devices are somewhat offset by their more

difficult colour tuning. Other recently reported TADF materials can also exhibit dual emission mechanisms through multiple conformers that in combination give white emission, but here too controlling the white balance can be challenging as it relies on molecular engineering of the emitter[5].

Although already having reached commercial maturity and extensively researched in inorganic LED pumped systems [6–8], the use of external downconverters (DCs) remains surprisingly rare in WOLED research [9–12]. This approach is shown in Figure 1, in which an external material is used to absorb some of the emission from the electrically driven device, and itself then emits the remaining colours necessary to produce white light. In the example of fluorescent tubes, UV emitting plasma discharge tubes excite a white-emitting phosphor powder DC, while in inorganic white LEDs an orange emitting DC layer is coated on top of an underlying blue LED. In the case of inorganic white LEDs this makes colour tuning extremely simple through the thickness or concentration of the DC topcoat, enabled by the availability of cheap and efficient blue LEDs based on GaN junctions.



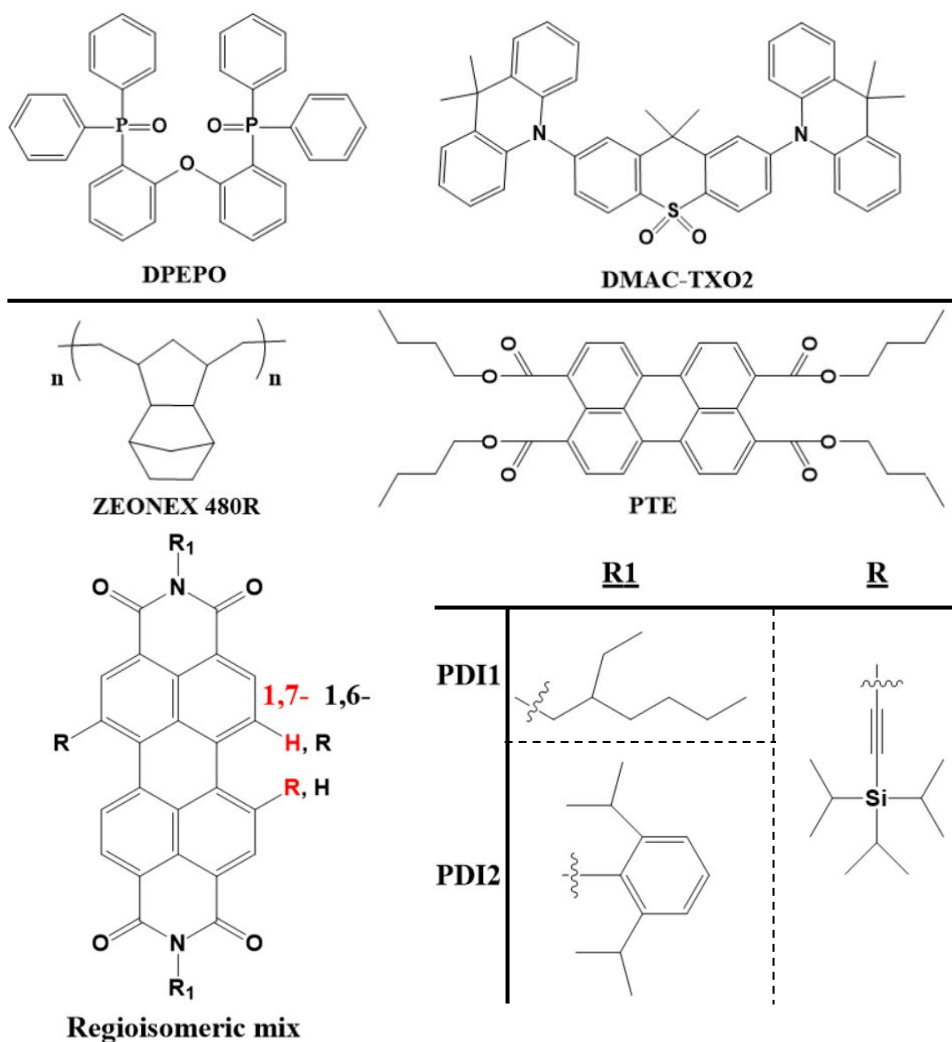
**Figure 1:** Schematic of DC WOLED approach. A blue OLED (left) is pumped electrically, with emitted light absorbed and attenuated through orange and green DC layers. These layers are

depicted detached from the OLED for clarity, and reemit at colours complementary to the base OLED to produce white light.

Despite their commercial successes, limitations of existing inorganic phosphors and inorganic LEDs make it appealing to apply the same approach in organic systems. In the context of WOLEDs, inorganic CeYAG based DC materials can be replaced by higher efficiency, lower cost, and optically tunable organic chromophores. Additionally, the narrow emission bands of inorganic blue OLEDs can be replaced with much broader OLED emission profiles, leading to enhanced colour rendering. This broad emission also increases the range of materials that can be used as DCs, as their absorption spectra are not as tightly restricted by the emission profile of the driving OLED. Indeed, recently perylene diimides (PDIs) and perylene tetraester (PTE) derivatives have been successfully incorporated in DC systems for inorganic LEDs [7,8], as these materials displaying excellent optical properties as well as high thermal stability [13–18]. Although previous work in this direction has been successful in generating white emission, overall WOLED efficiency is generally limited by poor conversion efficiency in the DC layer[10–12].

In this study, we have applied the DC approach to develop a high efficiency, fully organic WOLED. As the active components of the DC layer, bay-substituted perylene diimides (PDI-1 and PDI-2) and previously reported perylene-3,4,9,10-tetracarboxy tetrabutylester (PTE) were synthesized and characterized by  $H^1$ ,  $C^{13}$ -NMR, LC/MS mass spectrometry and optical spectroscopy. Solutions of both PDIs and PTE, having orange and green emission, respectively, were deposited by spin coating together with an inert polymer (zeonex) to give high clarity blue-to-orange DC films. To prevent dye aggregation and quenching in the DC, multiple spin coated layers were sequentially deposited at low concentrations. By depositing directly on top of a blue

thermally activated delayed fluorescence (TADF) OLED, the colour of the overall emission could be tuned from blue to white without introducing significant External Quantum Efficiency (EQE) losses compared to the underlying blue OLED.



**Figure 2:** Chemical structures of DMAC-TXO2, DPEPO, ZEONEX 480R, PTE, PDI1 and PDI2. Materials above the horizontal line are internal to the OLED, while those below are used to fabricate the external DC layers.

## **EXPERIMENTAL**

### **Synthesis of perylene derivatives and preparation of blue TADF OLED**

Chemical structures of all materials are shown in Figure 2. The green emitting PTE was synthesized according to literature procedures [7]. Bay substituted TIPS-functionalised PDIs with different imide groups have been previously reported [19]. The steps used to obtain mixtures of 1,6 and 1,7-di(triisopropylsilylacetylene)-N,N'-di[2-ethylhexyl]perylene-3,4,9,10-tetracarboxylicdiimide (PDI1) and mixtures of 1,6 and 1,7-di(triisopropylsilylacetylene)-N,N'-di[2,6-diisopropylphenyl]perylene-3,4,9,10-tetracarboxylicdiimide (PDI2) are given in Supporting Information (Figure S1-S8) with full structural and thermal characterisation. The bulky TIPS units were included to help preventing  $\pi$ - $\pi$  stacking and aggregation associated luminescence quenching in these dyes [17,20]. The optical properties of PDI1 and PDI2 were later found to be identical (Figure S9). In both cases the final materials were mixtures of both regioisomers, and used as prepared.

The blue OLEDs used as substrates for the DC layers were produced as a single batch (nine individual devices) and characterised as reported elsewhere [21], using the high performance TADF material DMAC-TXO2 co-evaporated at 30% in DPEPO as the emissive layer. All other organic layers were as previously reported, with full stack: ITO | NPB (40 nm) | TSBPA (10 nm) | DMAC-TXO2:DPEPO (30 nm) | DPEPO (10 nm) | TPBi (40 nm) | LiF (1nm) | Al (100nm).

### **Deposition, colour tuning, and evaluation of downconversion layer**

Stock solutions of PDI1, PDI2 or PTE (5mg/mL) and Zeonex polymer (100mg/mL) in toluene were prepared individually and dissolved by mixing at 50°C for 24 hours. The two stocks of Zeonex and one of the dyes (PDI1, PDI2, or PTE) were then mixed in the correct volume ratios

to give solutions with solid mass fractions of 0.1% perylene in Zeonex. DC layers were then deposited by spin-coating 100uL aliquots of these solutions at 3000 rpm, with 1 minute of continued spinning between successive depositions to allow the polymer film to adequately dry. Deposition in this fashion was found to give high clarity and uniform films in the first instance (when deposited directly onto glass). Because the film was allowed to dry completely, and because zeonex dissolves only very slowly in toluene (on a timescale of hours), additional depositions could be made in the same fashion directly atop the first layer without causing solvent damage to the dried film underneath. Furthermore, perfect refractive index matching of the first and subsequent polymer layers –on account of their identical composition- meant that the thicker films from multiple depositions retained high clarity, with top surface uniformity preserved at each spin-coating step. Repeated depositions allowed films with high optical absorbance to be formed while always keeping the concentration of the dyes low in both solution and solid states.

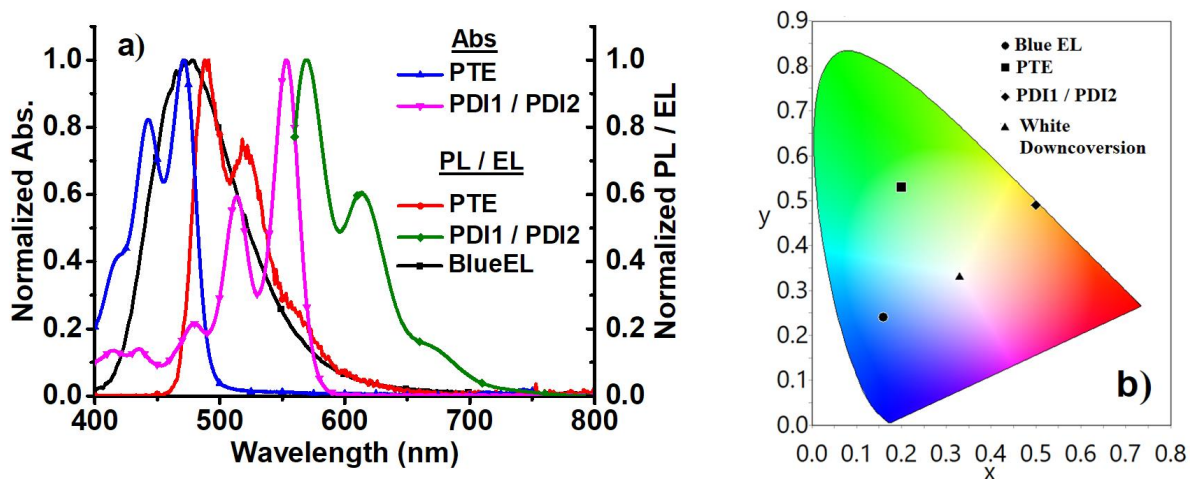
To gauge the appropriate number of layers needed to achieve good WOLED colour balance, a single OLED pixel (4 per device) was repeatedly driven at 5, 5.5 and 6V as the DC layer number (a proxy for total DC layer thickness) was increased. The OLED with DC layer was mounted inside a calibrated 12 inch integrating sphere (labsphere) when electrically driven (Keithley 2400 or Agilent 6632B SMU), and the integrated emission spectra recorded with a fibre spectrometer (Ocean Optics) from which the CIE coordinates could be calculated. Once suitable CIE coordinates had been achieved, an unstressed pixel on the same device was then swept from 0-12V to obtain full opto-electrical performance data.



## RESULTS & DISCUSSION

### Optical Characterisation of Perylenes

All three perylene dyes show strong absorption and vibronic emission bands in chloroform due to highly conjugated  $\pi$  systems. The absorption and photoluminescence (PL) of PTE is blue shifted compared to PDI1 and PDI2, due to its absence of imide groups and lower exciton delocalisation length (Figure 3a). In contrast, the blue electroluminescence (EL) of the TADF OLED is very wide and unstructured (typical of charge-transfer state TADF emitters [22–25]) and overlaps well with the absorption of both PTE and of PDI1/2. Therefore, the TADF OLED can excite these perylenes and they are broadly compatible for DC WOLED applications. Furthermore, the CIE-1931 colour coordinates of the blue EL and orange PDI1/2 PL are colinear with the pure white coordinate (0.33,0.33), indicating that these two materials should be able to achieve balanced white emission (Figure 3b).



**Figure 3:** a) Electroluminescence of blue OLED, as well as absorption and emission spectra of PDI1, PDI2 and PTE in chloroform solution. b) CIE coordinates of the emission from PDI1/2, PTE and blue EL of OLED.

Both PDI1 and PDI2 were found to have excellent photoluminescence quantum yields (PLQYs) in both solution and Zeonex films (Table 1). The solution PLQY of PTE is similarly high from previous reports (98% in dichloromethane) [26]. However, while films prepared at 0.1 wt% exhibited the same PL spectrum as in solution and similar high PLQYs, at even 0.5 wt% PDI concentrations, the intensity of the first vibronic emission peak was diminished compared to the second, and the PLQY was decreased (Figures S10 and S11). These deleterious effects at higher concentrations were primarily attributed to molecular aggregation during film drying, despite the inclusion of bulky TIPS groups. Alternatively or additionally, inner filter effects could alter the spectral shape of waveguided light emerging from the edge of the zeonex films (spin coated on sapphire substrate) in the PLQY sphere (Horiba Quanta-Phi). Inner filter effects on forwards emission were discounted on the basis of the small film thickness.

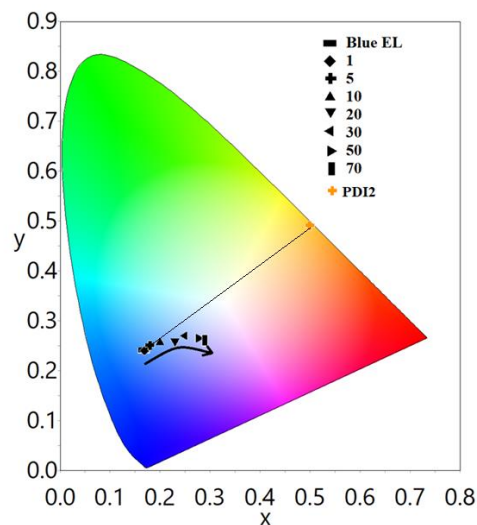
Although waveguided inner filter effects would not be observed in DC OLED applications (as the edges of the OLED are masked to prevent collection of waveguided emission, Figure 6), to strictly avoid any aggregation in DC applications it was therefore necessary to always keep the concentration of PDI stock solutions low. This restriction necessitated the multiple-step film fabrication procedure outlined above to achieve suitable DC film thicknesses and optical densities, despite the need to maintain low PDI concentrations.

**Table 1:** PLQY properties of Perylene derivatives

	Solution Phase ( $5 \times 10^{-6}$ M, $\text{CHCl}_3$ )	Film phase (Spin coated with Zeonex from toluene)	
		0.1 wt%	0.5 wt%
PDI1	92	94	87
PDI2	93	90	79

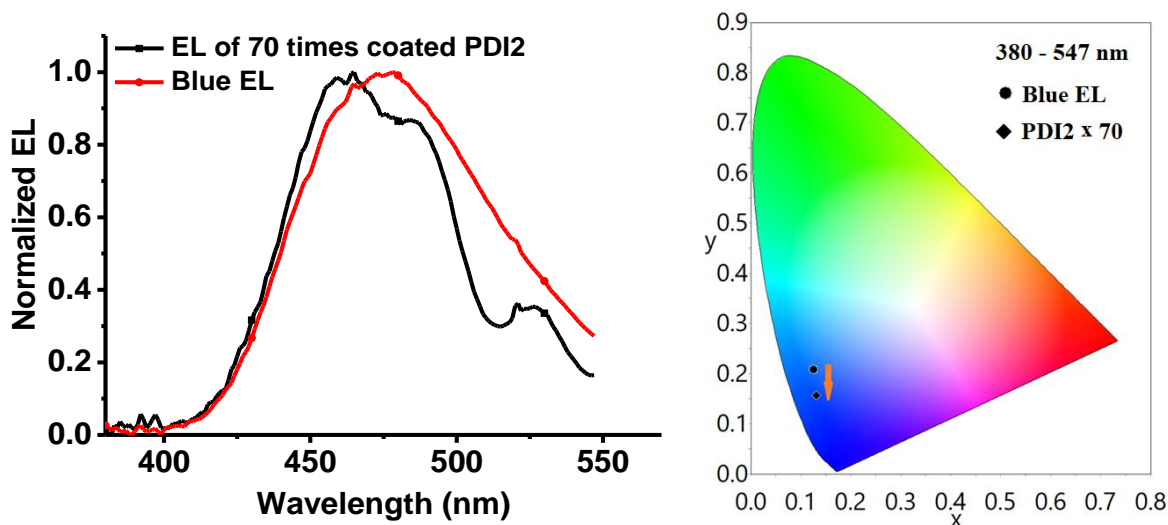
## Optimisation of DC OLED colour purity

PDI2 (0.1% concentration in zeonex) was first spin coated on a blue TADF OLED, with the emission colour recorded as the overall DC layer number (a proxy for total DC layer thickness) was increased. Inspired by other WOLED reports and the collinearity of the blue OLED EL, orange DC PL and white coordinate, we expected to be able to reach balanced white emission at the correct DC thickness. Figure 4 shows the actual trajectory through CIE coordinate space that this approach produced. Instead of continuing along a straight line, the CIE coordinates remained blue-ish, and refused to approach the desired balanced white. Despite this, a blue-white coordinate of (0.29, 0.26) with good efficiency performance was still achieved, Table 2. Once the best white coordinate had been achieved a single unstressed pixel on this device was also fully tested, with performance data included in Table 2 and Figure 7, and data and spectra in Table S1 and Figure S12, respectively.



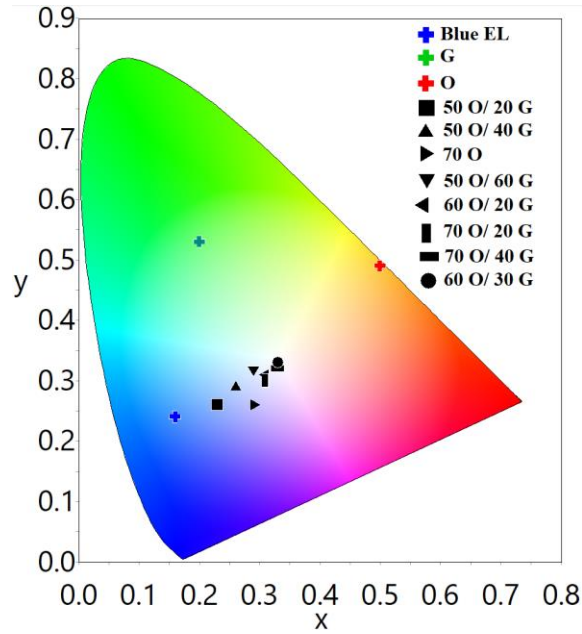
**Figure 4:** CIE trajectory of PDI2 DC WOLED with different thicknesses of DC layer (number of film depositions). The black line joining the blue OLED EL and PDI2 PL indicates the expected path through CIE space.

From the EL spectra of the DC WOLED we determined that this unexpected path through CIE space was due to absorptive filtering of the blue OLED EL emission. Figure 5 compares the short wavelength EL of the uncoated OLED with that of the DC WOLED with 70 layers of PDI2/zeonex. Bands subtracted from the OLED EL spectrum match the positions of the vibronic PDI2 absorption peaks, Figure 3a. This filtering effect means that the CIE coordinate of the blue ‘starting position’ shifts downwards as additional DC layers are added, as shown in Figure 5b. As a result, the trajectory through CIE space moves along a curve rather than a straight line. This result is due to the emission-absorption-reemission operating mechanism of the DC layer, which contrasts to mixed EML WOLEDs in which energy transfer from the blue to orange emitters is achieved through Förster or Dexter energy transfer (on much shorter distance scales). These processes instead deplete the blue emission band uniformly [27–29]. This filtering effect is also not significantly observed in inorganic DC WOLEDs, where the much narrowed blue emission spectrum of the inorganic OLED means that it is more uniformly absorbed by the DC layer [6–9].



**Figure 5:** EL spectrum of OLED with and without 70 layers PDI2 (left), and CIE coordinates of filtered and unfiltered EL spectra between 380 and 547 nm (right).

Based on these results, we identified that complementary green emission from PTE could provide ‘course correction’ in our path through CIE space, and move the device emission towards balanced white. On a new OLED, we first deposited 60 layers of PDI1 in zeonex to roughly approach white coordinates. Directly upon the orange perylene layers, additional layers of green PTE (0.1% in zeonex) were deposited and the overall emission colour checked to determine when a suitable white balance was achieved. The green DC layer was deliberately spun on top of the orange one, so that the forwards green emission of the PTE would not be strongly attenuated by absorption from the PDI layers (Figure 3a). Additionally, any emission from the PTE back towards the OLED would be reabsorbed and reemitted with high PLQY by the PDI1 layer, boosting the otherwise small absorption cross section of the PDI1 relative to the original blue EL. Different combinations of Orange (O) and Green (G) layer thicknesses were investigated, with the CIE coordinates plotted in Figure 6. Representative EL spectra are shown in Figure S12. Pure white emission was achieved by coating with 60 layers of PDI1 and 30 layers of PTE. The Colour Rendering Index (CRI) and Correlated Colour Temperature (CTT) of the hybrid white light produced were 80 and 5477, respectively.



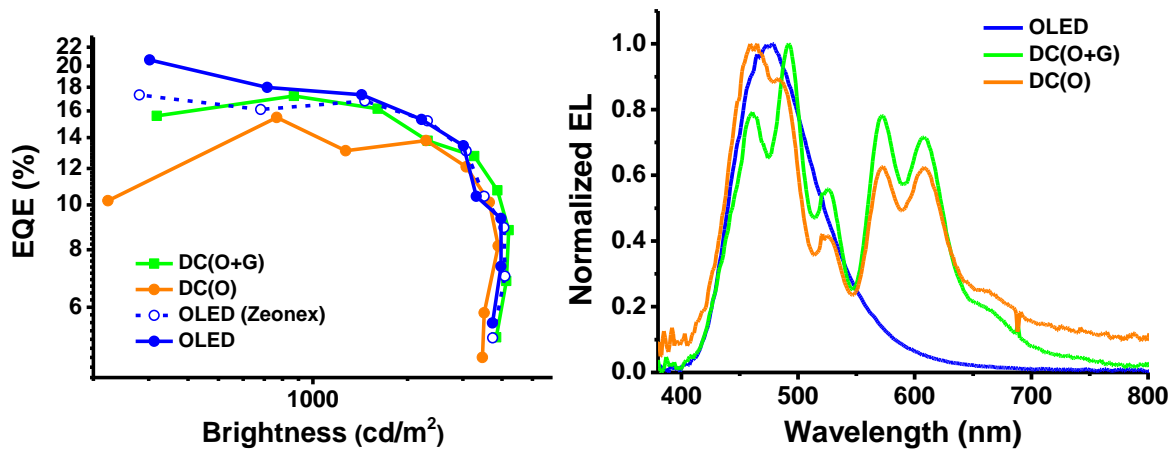
**Figure 6:** Top: photographs of uncoated blue TADF OLED and equivalent DC WOLED. Bottom: CIE coordinated of DC WOLED emission resulting from different combinations of Blue EL, PTE (Green, G) and PDI1 (Orange, O).

## Device Performance

Once the desired colour balance had been achieved by repeatedly testing a single pixel on a device, an unstressed pixel on the same device (also covered by the DC layer) was then characterised. The device performance and electroluminescence spectrum for the balanced WOLED is shown in Figure 7 (DC O+G), along with that of the WOLED with only orange PDI2 DC layer (DC O). Full device performance characteristics are included for the best measured pixels in Table 2. For comparison, we also plot the best device performance recorded for an uncoated pixel (OLED). We note that device electrical stress during testing means that we cannot accurately remeasure the same pixel's performance with and without the DC layer applied. As such we cannot know *a priori* which would be the best performing device and pixel to select for DC coating. It is therefore difficult to make direct comparisons between devices, and it is possible that our DC layer could provide a higher performance WOLED than what we report here. Despite this difficulty in comparison, to exclude efficiency improvements by outcoupling enhancement due to the surface morphology zeonex top layer[30], we also characterised a blue OLED with a 'blank' DC layer of same total thickness as our best white device, but with no perylenes (60 orange and 30 green, equivalent to 90 layers zeonex, OLED Zeonex).

Again stressing the difficulty in direct comparison, we nonetheless find that the EQE of the DC WOLED is not significantly diminished by the presence of the DC layer. Balanced white emission (CIE 0.33,0.33) with a maximum EQE of 17.2% is achieved at a practical brightness of 870 cd/m<sup>2</sup>. This compares well to the maximum EQE recorded for an uncoated OLED of 20.6% (CIE 0.16,0.24) at 302 cd/m<sup>2</sup>. The optoelectronic performance and efficiency roll off is similar to previous reports for DMAC-TXO2 in all devices, as is limited by the performance of the TADF emitter.

We also find that there is no significant increase in EQE due to outcoupling enhancement from the zeonex layer (maximum EQE 17.3% at 280 cd/m<sup>2</sup>), which is as expected for a flat (spin coated) layer of this polymer with well matched refractive index to the OLED glass substrate. As is expected for its mode of operation, we also observe no difference in the I-V characteristics of the DC WOLED due to its external DC layer (Figure S13). No comparison of device brightness is attempted here, on account of the different colours of emission being weighted differently by human optical sensitivity. The DC WOLED also possesses perfect colour stability with driving voltage (Figure S13), which it simply inherits from the base TADF OLED, but is more difficult to achieve in mixed EML WOLEDs.

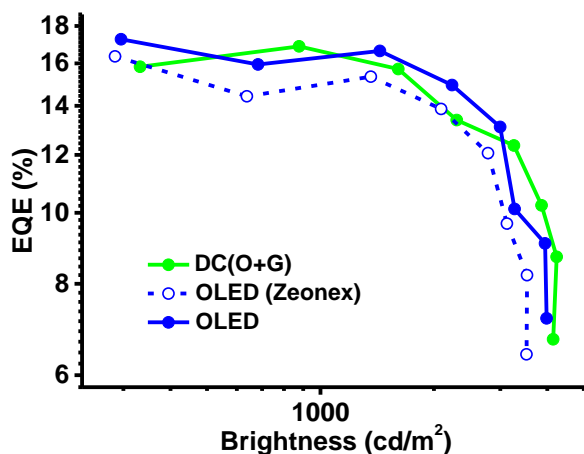


**Figure 7:** Device performance (left) and spectra (right) of blue TADF OLED and TADF DC WOLEDs with either Orange (O) or Orange and Green (O+G) DC layers. Data is also included for a bare OLED pixel, and one coated with plain zeonex.

As an additional investigation and for better comparison, we also measured the performance of different pixels on a single device with and without different DC layers. Due to the large thicknesses of the finally fabricated polymer DC layers, we found we were able to simply peel these intact directly away from the OLED substrate. The surface of a different OLED was then moistened with toluene to prevent an air gap and ensure good optical coupling, and the polymer



layer could then be pressed against its surface. This single OLED was characterised bare, with the zeonex-only polymer layer, and with the best balanced white DC layer on different unstressed pixels. The performance results (Figure 8) help to confirm that the DC layer does not significantly diminish the efficiency of the base blue OLED, but is also not assisted by improved outcoupling from the zeonex host layer.



**Figure 8:** Comparative device performance of different pixels on a single blue TADF OLED, with different coating layers.

Finally, we note that this approach for achieving high efficiency WOLEDs should be widely applicable to other TADF emitters, and other types of blue OLEDs generally. Additionally, fabrication of the DC layer is not limited to perylene dyes, nor zeonex as a polymer host. As an example, we suggest that suitable BODIPY dyes in PMMA may achieve similar success, although special care must be taken to develop suitable film deposition protocols for specific material and solvent combinations. For dyes that suffer quenching at high concentrations, the multiple spin-coating procedure presented here allows very thick films (with large optical densities necessary for DC applications) to be built up sequentially while maintaining low dye concentrations. However, because this DC fabrication method relies on rapid film drying and

slow dissolution of underlying layers to prevent surface damage by solvent, it is likely only applicable for polymeric DC host materials in contrast to small molecule hosts.

**Table 2:** Best recorded devices performance of bare and zeonex coated (90times) blue TADF OLEDs, as well as Orange DC and Orange/Green DC WOLEDs

Device	$\eta_{\text{ext a}}$ (% @ $\text{cd/m}^2$ )	$\eta_{\text{L a}}$ ( $\text{cd/A}$ )	$\eta_{\text{P a}}$ ( $\text{lm/W}$ )	$\eta_{\text{ext b}}$ (%@ $\text{cd/m}^2$ )	CIE $c$ ( $x, y$ )	CCT (K)	CRI $c$
<b>TADF OLED</b>	20.6 @ 302	35.1	24.5	7.3 @ 3973	0.16, 0.24	-	-
<b>TADF OLED with Zeonex</b>	17.3 @ 280	32.3	22.5	7.0 @ 4086	0.16, 0.24	-	-
<b>DC WOLED (orange)</b>	15.4 @ 766	31.1	19.5	5.3 @ 3889	0.29, 0.26	8918	72
<b>DC WOLED (orange and green)</b>	17.2 @ 870	38.7	24.3	8.8 @ 4202	0.33, 0.33	5477	80

<sup>a</sup>Measured maximum values at specified luminance.

<sup>b</sup>Measured values at maximum luminance.

<sup>c</sup>Measured values at maximum  $\eta_{\text{ext}}$ .

## CONCLUSION

Concentration aggregation and associated PL quenching of perylene dyes a major problem for many of their applications. In this study, we used a low-concentration multi-step spin coating method to obtain polymer films with high optical density that retain high fluorescence quantum yield. These films were applied as DC layers atop a blue TADF OLED to convert their emission

to white, with tuning achieved through number of film depositions (i.e. thickness). In a single component WOLED, absorptive filtering of the base OLED emission by the orange DC layer prevented the desired colour tuning. This problem was overcome using a double (orange and green) DC strategy, which provided WOLEDs with balanced white emission and maximum EQEs of 17.2%. Comparing pixels on the same device reveals that the DC layer does not significantly diminish the efficiency of the base OLED, and is not being assisted by outcoupling enhancement due to the polymer host of the DC layer. This approach is widely applicable for generating white emission from blue OLEDs, and is compatible with other classes of OLED emitters, other DC dyes, and other polymeric DC hosts.

## **ACKNOWLEDGEMENTS**

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## **REFERENCES**

- [1] D.D.S. Pereira, P.L. Dos Santos, J.S. Ward, P. Data, M. Okazaki, Y. Takeda, et al., An optical and electrical study of full thermally activated delayed fluorescent white organic light-emitting diodes, *Sci. Rep.* 7 (2017) 1–8. doi:10.1038/s41598-017-06568-3.
- [2] X. Tang, Y. Li, Y. Qu, C. Peng, A. Khan, Z. Jiang, All-Fluorescence White Organic Light-Emitting Diodes Exceeding 20 % EQEs by Rational Manipulation of Singlet and Triplet Excitons, 1910633 (2020) 1–8. doi:10.1002/adfm.201910633.

- [3] L. Popp, R. Scholz, P. Kleine, R. Lygaitis, S. Lenk, S. Reineke, High performance two-color hybrid TADF-phosphorescent WOLEDs with bimodal Förster and Dexter-type exciton distribution, *Org. Electron.* 75 (2019) 105365. doi:10.1016/j.orgel.2019.07.023.
- [4] X.K. Liu, Z. Chen, J. Qing, W.J. Zhang, B. Wu, H.L. Tam, et al., Remanagement of Singlet and Triplet Excitons in Single-Emissive-Layer Hybrid White Organic Light-Emitting Devices Using Thermally Activated Delayed Fluorescent Blue Exciplex, *Adv. Mater.* 27 (2015) 7079–7085. doi:10.1002/adma.201502897.
- [5] K. Wang, Y.Z. Shi, C.J. Zheng, W. Liu, K. Liang, X. Li, et al., Control of Dual Conformations: Developing Thermally Activated Delayed Fluorescence Emitters for Highly Efficient Single-Emitter White Organic Light-Emitting Diodes, *ACS Appl. Mater. Interfaces.* 10 (2018) 31515–31525. doi:10.1021/acsami.8b08083.
- [6] A. a. Wiles, J. Bruckbauer, N. Mohammed, M. Cariello, J. Cameron, N.J. Findlay, et al., A poly(urethane)-encapsulated benzo[2,3- d :6,7- d ' ]diimidazole organic down-converter for green hybrid LEDs , *Mater. Chem. Front.* (2020). doi:10.1039/c9qm00771g.
- [7] E. Aksoy, N. Demir, C. Varlikli, White LED light production using dibromoperylene derivatives in down conversion of energy, in: *Can. J. Phys.*, 2018: pp. 734–739. doi:10.1139/cjp-2017-0752.
- [8] T. Guner, E. Aksoy, M.M. Demir, C. Varlikli, Perylene-embedded electrospun PS fibers for white light generation, *Dye. Pigment.* 160 (2019) 501–508. doi:10.1016/j.dyepig.2018.08.040.
- [9] J.W. Han, C.W. Joo, J. Lee, D.J. Lee, J. Kang, S. Yu, et al., Enhanced outcoupling in down-conversion white organic light-emitting diodes using imprinted microlens array films with breath figure patterns, *Sci. Technol. Adv. Mater.* 20 (2019) 35–41. doi:10.1080/14686996.2018.1551040.
- [10] A.R. Duggal, J.J. Shiang, C.M. Heller, D.F. Foust, Organic light-emitting devices for illumination quality white light, *Appl. Phys. Lett.* 80 (2002) 3470–3472. doi:10.1063/1.1478786.
- [11] S. Chen, H.S. Kwok, Top-emitting white organic light-emitting diodes with a color conversion cap layer, *Org. Electron.* 12 (2011) 677–681. doi:10.1016/j.orgel.2011.01.005.
- [12] V. Gohri, S. Hofmann, S. Reineke, T. Rosenow, M. Thomschke, M. Levichkova, et al., White top-emitting organic light-emitting diodes employing a heterostructure of down-conversion layers, *Org. Electron.* 12 (2011) 2126–2130. doi:10.1016/j.orgel.2011.09.002.
- [13] N.J.L.K. Davis, R.W. Macqueen, D. a. Roberts, A. Danos, S. Dehn, S. Perrier, et al., Energy transfer in pendant perylene diimide copolymers, *J. Mater. Chem. C.* 4 (2016) 8270–8275. doi:10.1039/c6tc02555b.

- [14] M. Kus, Ö. Hakli, C. Zafer, C. Varlikli, S. Demic, S. Özçelik, et al., Optical and electrochemical properties of polyether derivatives of perylenediimides adsorbed on nanocrystalline metal oxide films, *Org. Electron.* 9 (2008) 757–766. doi:10.1016/j.orgel.2008.05.009.
- [15] C. Karapire, C. Timur, S. İçli, A comparative study of the photophysical properties of perylenediimides in liquid phase, PVC and sol-gel host matrices, *Dye. Pigment.* 56 (2003) 135–143. doi:10.1016/S0143-7208(02)00128-6.
- [16] K. Ertekin, C. Karapire, S. Alp, B. Yenigu, B. Yenigül, S. İçli, Photophysical and photochemical characteristics of an azlactone dye in sol-gel matrix; a new fluorescent pH indicator, *Dye. Pigment.* 56 (2003) 125–133. doi:10.1016/S0143-7208(02)00125-0.
- [17] C. Karapire, M. Kus, G. Turkmen, C.C. Trevithick-Sutton, C.S. Foote, S. Icli, Photooxidation studies with perylenediimides in solution, PVC and sol-gel thin films under concentrated sun light, *Sol. Energy.* 78 (2005) 5–17. doi:10.1016/j.solener.2004.07.003.
- [18] C. Karapire, C. Zafer, S. İçli, S. İçli, Studies on photophysical and electrochemical properties of synthesized hydroxy perylenediimides in nanostructured titania thin films, *Synth. Met.* 145 (2004) 51–60. doi:10.1016/j.synthmet.2004.04.016.
- [19] Y. Huang, W. Zhang, H. Zhai, C. Li, Alkylsilane-functionalized perylenediimide derivatives with differential gas sensing properties, *J. Mater. Chem. C.* 3 (2015) 466–472. doi:10.1039/c4tc02059f.
- [20] S.A. El-daly, T.A. Fayed, Diimide in Chloromethane Solvents, *J. Photochem. Photobiol. A.* 137 (2000) 15–19. doi:10.1016/S1010-6030(00)00333-6.
- [21] P. Stachelek, J.S. Ward, P.L. dos Santos, A. Danos, M. Colella, N. Haase, et al., Molecular Design Strategies for Color Tuning of Blue TADF Emitters, *ACS Appl. Mater. Interfaces.* 11 (2019) 27125–27133. doi:10.1021/acsami.9b06364.
- [22] R. Huang, N. a. Kukhta, J.S. Ward, A. Danos, A.S. Batsanov, M.R. Bryce, et al., Balancing charge-transfer strength and triplet states for deep-blue thermally activated delayed fluorescence with an unconventional electron rich dibenzothiophene acceptor, *J. Mater. Chem. C.* 7 (2019) 13224–13234. doi:10.1039/c9tc02175b.
- [23] N. a. Kukhta, H.F. Higginbotham, T. Matulaitis, A. Danos, A.N. Bismillah, N. Haase, et al., Revealing resonance effects and intramolecular dipole interactions in the positional isomers of benzonitrile-core thermally activated delayed fluorescence materials, *J. Mater. Chem. C.* 7 (2019) 9184–9194. doi:10.1039/c9tc02742d.
- [24] M. Colella, A. Danos, A.P. Monkman, Less is more: Dilution enhances optical and electrical performance of a TADF exciplex, *J. Phys. Chem. Lett.* 10 (2019) 793–798. doi:10.1021/acs.jpcllett.8b03646.

- [25] M. Colella, A. Danos, A.P. Monkman, Identifying the Factors That Lead to PLQY Enhancement in Diluted TADF Exciplexes Based on Carbazole Donors, *J. Phys. Chem. C.* 123 (2019) 17318–17324. doi:10.1021/acs.jpcc.9b03538.
- [26] H. Langhals, J. Karolin, L.B.-Å. Johansson, Spectroscopic properties of new and convenient standards for measuring fluorescence quantum yields, *J. Chem. Soc., Faraday Trans. 94* (1998) 2919–2922. doi:10.1039/a804973d.
- [27] R.K. Gupta, D. Das, P.K. Iyer, A.S. Achalkumar, First Example of White Organic Electroluminescence Utilizing Perylene Ester Imides, *ChemistrySelect.* 3 (2018) 5123–5129. doi:10.1002/slct.201801258.
- [28] I. Oner, C. Varlikli, S. Icli, The use of a perylenediimide derivative as a dopant in hole transport layer of an organic light emitting device, Elsevier B.V., 2011. doi:10.1016/j.apsusc.2011.02.002.
- [29] L.C. Meng, Z.D. Lou, S.Y. Yang, Y.B. Hou, F. Teng, X.J. Liu, et al., White organic light-emitting diodes based on a combined electromer and monomer emission in doubly-doped polymers, *Chinese Phys. B.* 21 (2012). doi:10.1088/1674-1056/21/8/088504.
- [30] J.-H. Yen, Y.-J. Wang, C.-A. Hsieh, Y.-C. Chen, L.-Y. Chen, Enhanced light extraction from organic light-emitting devices through non-covalent or covalent polyimide-silica light scattered hybrid films, *J. Mater. Chem. C.* (2020). doi:10.1039/c9tc06449d.

## TOC GRAPHIC

