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Nathan Bakken
Zixing Wang
Jian Li

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Nathan Bakken, Zixing Wang, and Jian Li
Arizona State University, Materials Engineering, Tempe, Arizona 85287
jian.li.1@asu.edu

Abstract. White organic light-emitting devices (WOLEDs) can be fabricated using a simple, low-cost device structure with a single uniformly doped emissive layer. The Pt-17 emitter used in these devices obtains excellent color rendering ($\mathrm{CRI} = 80$) as well as bright white electrophosphoresence (CIE $x = 0.37$, $y = 0.40$) by combining efficient monomer and efficient excimer emission as demonstrated by excellent external quantum efficiency ($\eta_{\mathrm{ext}} = 15.9\%$). The Pt-17 based WOLED is also compatible with state-of-the-art charge injection and blocking materials as well as high out-coupling device structures. Application of these existing technologies is expected to extend luminance efficiencies of Pt-17 devices to world-class values ($46\ \mathrm{lm}/W$ and $100\ \mathrm{lm}/W$ respectively). In addition to avoiding the difficulty and cost of fabricating more complex device structures, the color of a single-doped device also is uniquely independent of voltage, current density, and age. Molecules like fluorine-free Pt-17 are uniquely positioned to utilize excimer emissions in order to reduce manufacturing costs and provide solutions to satisfy many of the requirements for the next generation of organic solid-state lighting. *© 2012 Society of Photo-Optical Instrumentation Engineers (SPIE).* [DOI: 10.1117/1.JPE.2.021203]

Keywords: white organic light-emitting devices; excimer; solid-state lighting; platinum complex.

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White organic light-emitting diodes (WOLEDs) with high power efficiency are strong candidates for next-generation illumination devices.[1,2] To date, however, the electroluminescent (EL) spectrum of WOLEDs is generated using multiple emitters embedded in a complex device structure.[3–7] Here we report that efficient WOLEDs with high quality white light can be fabricated using a single emitter. The ideal broad EL spectrum was realized by combining monomer and excimer emission. In a four-layer WOLED, a forward power efficiency of $\eta_p = 37.8\ \mathrm{lm/W}$ (corresponding to external quantum efficiency $\eta_{\mathrm{ext}} = 15.1\%$) was achieved with an EL spectrum yielding Commission Internationale de L'Éclairage (CIE) chromaticity coordinates of (0.37, 0.40) and a color rendering index (CRI) value of 80. The demonstration of efficient single-doped WOLEDs with high illumination quality presents a unique opportunity to simplify the device architecture and eliminate the problems of color aging and color instability for WOLEDs using multiple emitters. All of these improvements will help to enable future commercialization of WOLEDs for lighting applications.

Unlike rapidly maturing monochromatic OLEDs, one of the greatest challenges for WOLEDs is the absence of a single organic emitter (fluorescent or phosphorescent material) that covers the entire visible spectrum.[8–12] The broad spectrum required for white light in organic devices has previously been obtained from the combined emission of multiple emissive dopants shown in the relatively complex structures illustrated in Fig. 1. These structures include a multiple-doped emissive layer (EML),[4,13–15] a multilayer of emitters (doped films or neat films),[6,7,16] multiple emissions from monomer and excimers or exciplexes,[9,17–19] hybrid polymers/inorganics,[20] etc. The development of WOLEDs has progressed rapidly during the last decade. The efficacy has reached a record high of 100 lm/W with enhanced out-coupling effect.[1,2] The limited operational lifetime that has previously handicapped OLED technology

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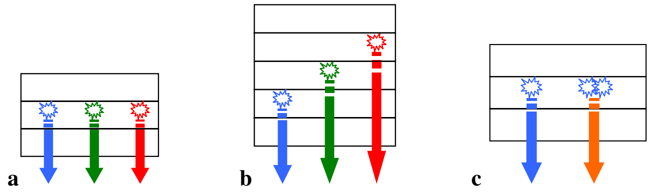

Fig. 1 Schemes for three typical WOLED architectures. (a) Triple-doped emissive layer, (b) multiple emissive layers, and (c) emissive layer with monomer and excimer.

has now been demonstrated to exceed 10,000 h.[2] However, such high efficacy and long operational lifetime are achieved using multiple emissive dopants, which require relatively complex device structures, raising the difficulty to manufacture consistently at low cost. The use of multiple emissive dopants generates an unstable emission color for WOLEDs due to the energy transfer process between different emitters. It is common that the EL spectrum of WOLEDs varies with different driving voltages (or driving current) due to the change of ratio in blue, green, and red emission.[21] Moreover, the different degradation rates for each emissive dopant in WOLEDs could create a problem of “color aging”—that is, the color of lighting changes significantly in the degradation process, resulting in premature device failure. To prevent color aging and enhance color stability, the device structures will inevitably increase in complexity and probably manufacturing cost.

To overcome these problems, an elegant solution (one not requiring a relatively complex device architecture) will be to fabricate WOLEDs using a single emitter. This can be realized by coupling the blue-green monomer emission and orange excimer emission that has been reported for square planar Pt complexes (i.e., FPt and their analogs).[13–15] In contrast to most organic emitters, Pt complexes can form a broadly emitting excimer. The strong Pt . . . Pt interaction can destabilize the highest occupied molecular orbital (HOMO), resulting in a red-shifted emission spectrum for excimers [Fig. 2(a)]. If the excimer emission adequately complements monomer emission, the EL spectrum of excimer-based WOLEDs can include all visible wavelengths. The ratio of monomer/excimer emission can be controlled by varying the dopant concentration, the morphology of host materials, or the relative balance of hole/electron injection, providing ease of color controlling for WOLEDs.[13,14] Additionally, because an excimer does not have a bound ground state, the cascade of energy from the blue (higher energy) emitter to the excimer can be prevented, leading to a stable emission color independent of the driving voltage. Moreover, the problem of color aging can be resolved by the use of a single emitter. The combination of monomer and excimer emission provides a simple and elegant solution to achieve voltage-independent, stable, broad-spectrum WOLEDs.

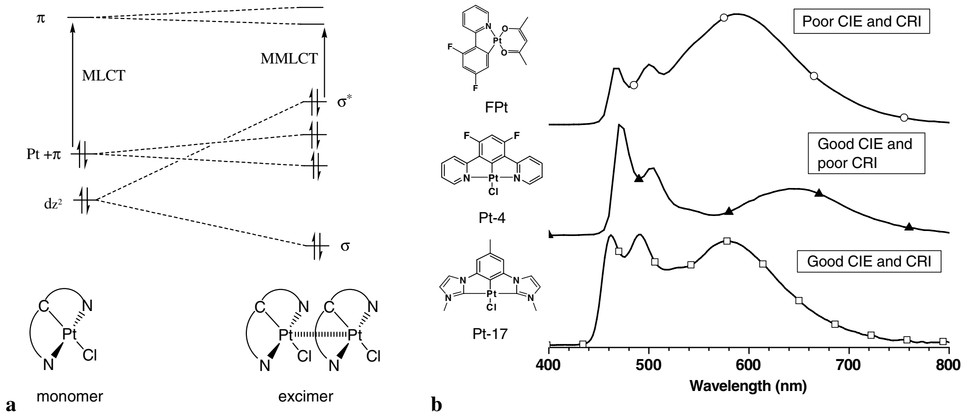

Fig. 2 The formation mechanism and EL spectra of phosphorescent excimers. (a) Excimer formation for square planar Pt complexes like Pt(N^C^N)Cl and their analogs. The newly formed Pt . . . Pt bond destabilized the highest occupied molecular orbital (HOMO), resulting in a red-shifted emission spectrum for excimer of Pt complexes. (b) Electroluminescent spectra of OLEDs using FPt, Pt-4, and Pt-17, the chemical structures of which are shown to the left of the graphs.

To meet the requirement for solid state lighting, several factors must be considered for excimer-based WOLEDs: the emission efficiency of monomer/excimer, the emission colors of monomer/excimer, and the optimization of charge injection and recombination inside emissive layers. Particularly, the perceived quality of illumination is the key to success in lighting applications. Two important parameters are used to define the color quality of a white light source: the CIE chromaticity coordinates and the CRI. The highest quality white illumination requires sources with CIE coordinates close to (0.33, 0.33) and a CRI value over 80.[22] The phenomenon of excimer formation has been observed for a long time,[23] but the photophysical properties of excimers remain poorly understood. For this reason, progress on phosphorescent excimers has depended almost entirely on the synthesis and characterization of new materials. As illustrated in Fig. 2(b), previously reported FPt and Pt-4 cannot produce a satisfactory white EL spectra due to either inefficient monomer emission (FPt)[14] or unsuitable excimer emission color (Pt-4).[15] Here, we demonstrate the first reported efficient single-doped WOLEDs with high-quality white light with a novel Pt complex, that is, Pt-17. Compared with FPt, both Pt-17 and Pt-4 are more efficient blue phosphorescent emitters due to the specific choice of metal complex system, that is, Pt (N^C^N)Cl, where N^C^N are dipyridinyl benzene and their analogs.[24] Replacing pyridinyl group with methyl-imidazolyl group could potentially weaken the intermolecular interaction, resulting in a blue-shifted excimer emission for Pt-17. Thus, an ideal white EL spectrum can be accomplished by combining monomer and excimer emission of Pt-17 as shown in Fig. 2(b).

To evaluate the electroluminescent properties of Pt-17, a series of WOLEDs were fabricated in a four-layer device structure: NPD/TAPC/Pt17:26mCPy/PO15, with the dopant concentration ranging from 2% to 26%.[25] Forward-viewing external quantum efficiency (EQE) is plotted versus current density for these devices in Fig. 3(a), with the corresponding EL spectra and CIE coordinates displayed in Fig. 3(b). A summary of device performance is presented in Table 1. Similar to the previously reported phosphorescent excimers, the gradual increase of dopant concentration raises the contribution from Pt-17 excimers, tuning the color of OLEDs from blue (exclusive monomer emission for a 2% Pt-17 device) to broad and whiter emission. In contrast to both FPt and Pt-4, all of the Pt-17 devices have demonstrated reasonably high device efficiencies with peak values ranging from 8 to 13%, indicating that both monomer and excimer of Pt-17 are efficient phosphorescent emitters. This presents a great opportunity to fabricate an efficient WOLED in addition to having the ideal color combination of monomer and excimer emission. Figure 3(b) illustrates how a simple variation in the dopant concentration allows tuning the color of WOLEDs from cool white (CIE $x = 0.31$, $y = 0.38$ for 14% Pt-17 device) to warm white (CIE $x = 0.42$, $y = 0.43$ for 26% Pt-17 device) while maintaining a high CRI value (>75).

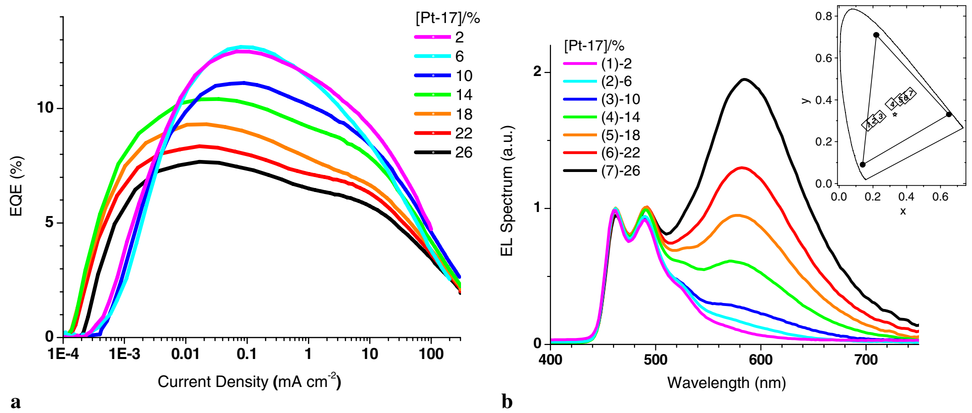

Fig. 3 Performance characteristics of Pt-17 based WOLEDs using 26mCPy as a host material. (a) Forward-viewing external quantum efficiency versus current density of the WOLEDs. The device structure is ITO/PEDOT/NPD (30 nm)/TAPC (10 nm)/x% Pt-17:26mCPy (25 nm)/PO15 (20 nm)/LiF/Al, where x ranges from 2 to 26. (b) Normalized electroluminescent spectra of the devices and the CIE coordinates (inset) of the WOLEDs operated at a current density of 1 mA/cm^2.

Table 1 A summary of device characteristics. Current density is 1 mA/cm^2. The device structure is ITO/PEDOT/NPD (30 nm)/TAPC (10 nm)/x% Pt-17:26mCPy (25 nm)/PO15 (20 nm)/LiF/Al unless otherwise noted.

[Pt-17]	Bias (V)	Luminance (cd/m^2)	Forward-viewing EQE	CIE coordinates		CRI	Power efficiency (lm/W)
				x	y		
2%	4.5	198	11.5	0.17	0.28		14.1
6%	4.6	214	11.4	0.20	0.30		14.8
10%	4.4	213	10.1	0.24	0.32	65	16.0
14%	4.0	212	9.2	0.31	0.38	75	16.2
18%	3.9	175	7.9	0.36	0.40	77	14.5
22%	4.0	172	7.1	0.39	0.41	77	12.9
26%	3.8	154	6.5	0.42	0.43	75	12.3
18%[20][a]	3.9	219	10.3	0.30	0.36	75	17.7
18%[40][a]	4.0	350	15.7	0.37	0.40	80	27.3

[a] 26mCPy is replaced by a co-host of TAPC:PO15 (1:1) where the thickness of the electron transporting layer PO15 is presented in brackets.

Increasing the dopant concentration slightly lowers the driving voltage; however, it also decreases the device efficiency. The relative inefficiency of the excimer as compared to monomer emission as well as the effect of charge carrier imbalance internal to the emissive layer could account for this behavior. However, an observed independence of efficiency with changing concentration in other host materials suggests that the charge carrier imbalance better explains the decrease in efficiency. Employing a co-host system with more balanced hole/electron transporting capabilities can then be expected to improve efficiency, especially for higher dopant concentrations capable of achieving white CIE coordinates of interest. A mixed layer of TAPC:PO15 is documented as one example, which can replace 26mCPy as host materials in a similar device setting.[22] Electrical and optical performance characteristics for device structure NPD/TAPC/18% Pt-17:TAPC:PO15/PO15 are shown in Figs. 4(a)–4(c). Forward-viewing EQE and power efficiency are plotted versus luminance in Fig. 5(a). The thickness of electron transporting layer PO15 was optimized to 40 nm to maximize the device efficiencies (Table 1 and Fig. 4). By maintaining the assumption of Lambertian emission, it is plausible to account for the increase in efficiency as a result of ideal charge carrier balance. A maximum forward-viewing EQE of $\eta_{\mathrm{ext}} = 15.9\%$ was achieved at a current density of $J = 0.3\ \mathrm{mA/cm^2}$ and only decreases slightly to $\eta_{\mathrm{ext}} = 15.6\%$ at a high forward-viewing luminance of 500 cd/m^2. This device also gives a maximum forward power efficiency of $\eta_p = 37.8\ \mathrm{lm/W}$ and remains at a high $\eta_p = 25.2\ \mathrm{lm/W}$ at a practical luminance (500 cd/m^2). The electroluminescent spectrum of the device has a slight change due to the variation in a host system, yielding highly desirable CIE coordinates of (0.37, 0.40) and CRI of 80, which also demonstrates an independence of current density. The illumination quality of this WOLED is comparable, or even superior, to WOLEDs in the literature that require significantly more complex structures.

The efficiency of the present co-host device can be further improved by using state-of-the-art charge-injection materials and charge blockers, resulting in a higher forward-viewing EQE (20% or higher) and a higher forward power efficiency (46 lm/W or higher).[26] The use of a higher-refractive-index substrate could extract more photons from the device, leading to a twofold increase in power efficiency according to demonstrated methods.[1,27] Thus, a single-doped WOLED with η_p of 100 lm/W can be realized by integrating Pt-17 monomer plus excimer emission with established out-coupling enhancement technologies. Moreover, Pt-17 is a fluorine-free blue phosphorescent emitter, indicating that its design is aligned with molecules that have demonstrated stability for a long operational lifetime.[28] Continued characterization and development should provide a viable route to develop stable and efficient phosphorescent excimers for lighting applications. Demonstration of single-doped WOLEDs with high efficiency and high illumination quality presents a unique opportunity to significantly simplify the device architecture and eliminate the problems of color aging and color instability for WOLEDs using multiple emitters. This will help to expedite the commercialization of WOLEDs for lighting applications.

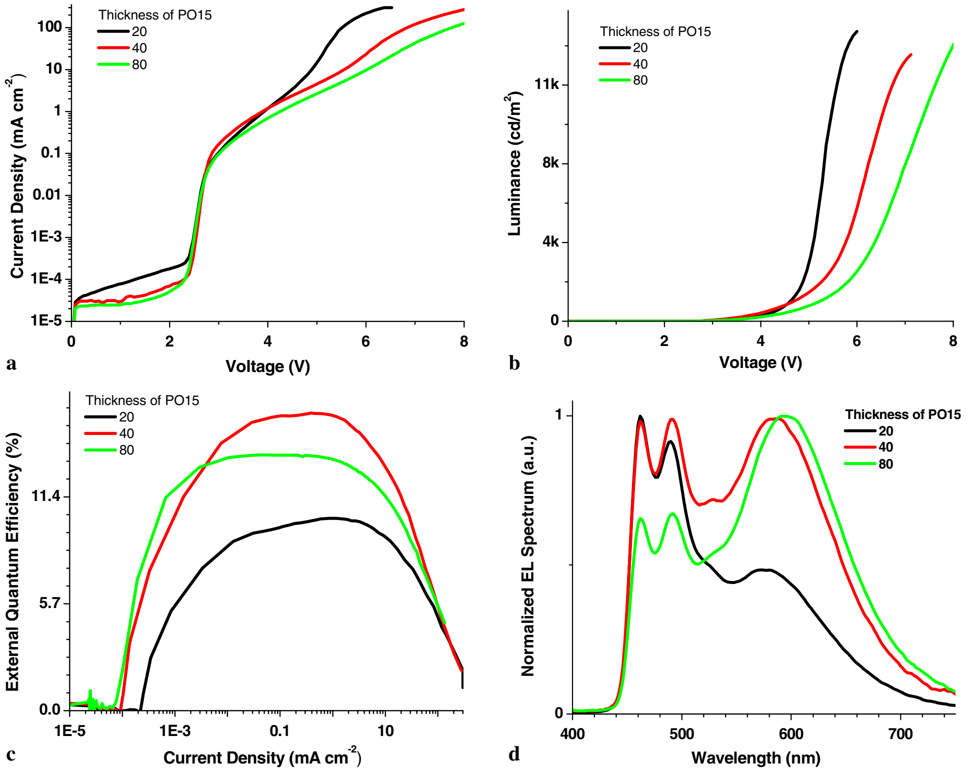

Fig. 4 Performance characteristics of Pt-17 based WOLEDs using TAPC:PO15 as co-host materials. (a) Current density versus voltage, (b) luminance versus voltage, (c) forward-viewing external quantum efficiency versus current density, (d) electroluminescent spectra of WOLEDs. The normalized spectra were indistinguishable when collected at 1 mA/cm^2 and 10 mA/cm^2. The device structure is ITO/PEDOT (45 nm)/NPD (30 nm)/TAPC (10 nm)/18% Pt-17:TAPC:PO15 (25 nm)/PO15 (*x* nm)/LiF/Al. PO15 thickness ranges from 20 to 80 nm.

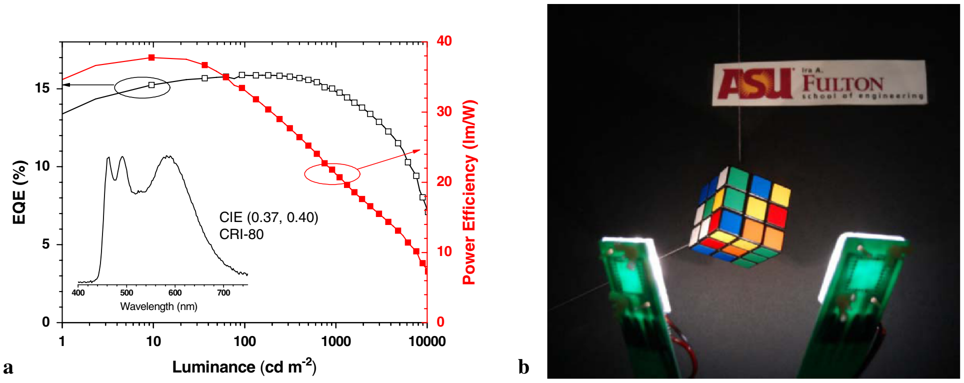

Fig. 5 Performance characteristics of Pt-17 based WOLEDs using TAPC:PO15 as co-host materials. (a) Forward-viewing external quantum efficiency (open squares) and power efficiency (filled squares) versus luminance of the WOLED. The device structure is ITO/PEDOT/NPD (30 nm)/TAPC (10 nm)/18% Pt-17:TAPC:PO15 (25 nm)/PO15 (40 nm)/LiF/Al. (b) An image of a Rubik's cube illuminated by the WOLED showing its white color and high color-rendering-index value.

2 Methods

Devices were fabricated on indium tin oxide (ITO) coated glass substrates. PEDOT:PSS was filtered through a 0.2 μm filter and spin-coated on the precleaned substrates, giving a 40- to 50 nm-thick film. All other materials were deposited in a glovebox-hosted vacuum deposition system at a pressure of 10^{-7} torr. Spectra were measured with an Ocean Optics HR-4000 spectrometer, and current-voltage-luminance characteristics were taken with a Keithley 2400 Source Meter and a Newport 818 Si photodiode. All device operation and measurement were carried out inside a nitrogen-filled glovebox. CRI and CIE calculations were obtained from device EL spectra using methods consistent with established literature.[29] Individual devices had areas of 0.04 cm^2. Agreement between luminance, optical power, and EL spectra was verified with a calibrated Photo Research PR-670 spectroradiometer with all devices assumed to be Lambertian emitters.

The chemical names of the materials abbreviated thus far are as follows: Pt-17, platinum(II) bis(methyl-imidazolyl)toluene chloride; FPt, platinum(II) [2-(4′,6′-difluorophenyl) pyridinato-*N*, $C^{2\prime}$)](2,4-pentanedionato); Pt-4, platinum(II) 1,3-difluoro-4,6-di(2-pyridinyl)benzene chloride; NPD, *N*,*N*′-diphenyl-*N*,*N*′-bis(1-naphthyl)-1,1′-biphenyl-4,4′′-diamine; TAPC, di-(4-*N*,*N*-ditolyl-amino-phenyl) cyclohexane; PO15, 2,8-bis(diphenylphosphoryl)-dibenzothiophene; and 26mCPy, 2,6-bis(*N*-carbazolyl) pyridine.

The layer sequence for WOLEDs on top of the ITO substrate is 45 nm PEDOT:PSS as a hole injection material/30 nm NPD as a hole-transporting layer/10 nm TAPC as an electron-blocking layer/25 nm emissive layer/20 to 80 nm PO15 as an electron-transporting layer and a hole-blocking layer/1 nm LiF/90 nm Al cathode. The emissive layer consists of either 26mCPy as a host or TAPC:PO15 (1∶1) as co-host materials with Pt-17 as a phosphorescent emitter.

To synthesize Pt-17, a mixture of bis(methyl-imidazolyl)toluene diiodide (1 mmol) and 0.5 mmol silver oxide was stirred in a solution of 100 mL acetonitrile for 5 h at room temperature before 1 equiv. platinum chloride and 1 equiv. potassium carbonate were added. The reaction mixture was heated to reflux for an additional 24 h. Then the mixture was cooled to room temperature before 100 mL water was added. The resulting yellow precipitate was filtered off and washed with excessive methanol, water, and ether and dried under vacuum. The light yellowish product (in 20% yield) was obtained after thermal evaporation under high vacuum. NMR (500 MHz, DMSO): δ 9.88 (s, 2H), 8.36 (d, 2H), 8.02 (d, 2H), 3.99 (s, 6H), 2.53 (s, 3H).

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N. B. fabricated the devices, analyzed the data, and wrote the manuscript; Z. W. designed and synthesized the emissive materials; and J. L. directed the project and co-wrote the manuscript.

The authors declare that they have no competing financial interests or other interests that might be perceived to influence the results and/or discussion reported in this article.

References

1. S. Reineke et al., “White organic light-emitting diodes with fluorescent tube efficiency,” *Nature* **459**, 234–238 (2009), http://dx.doi.org/10.1038/nature08003.
2. B. W. D’Andrade et al., “Realizing white phosphorescent 100 lm/W OLED efficacy,” *Proc. SPIE* **7051**, 70510Q (2008), http://dx.doi.org/10.1117/12.793758.
3. B. W. D’Andrade and S. R. Forrest, “White organic light-emitting devices for solid-state lighting,” *Adv. Mater.* **16**(18), 1585–1595 (2004), http://dx.doi.org/10.1002/adma.200400684.
4. M. C. Gather, A. Kohnen, and K. Meerholz, “White organic light-emitting diodes,” *Adv. Mater.* **23**(2), 233–248 (2011), http://dx.doi.org/10.1002/adma.v23.2.
5. B. W. D’Andrade, R. J. Holmes, and S. R. Forrest, “Efficient organic electrophosphorescent white light-emitting device with a triple doped emissive layer, ”*Adv. Mater.* **16**(7), 624–628 (2004).http://dx.doi.org/10.1002/adma.200306670
6. J. Kido, M. Kimura, and K. Nagai, “Multilayer white light-emitting organic electroluminescent device,” *Science* **267**(5202), 1332–1334 (1995), http://dx.doi.org/10.1126/science.267.5202.1332.
7. Y. Sun et al., “Management of singlet and triplet excitons for efficient white organic light-emitting devices,” *Nature* **440**, 908–912 (2006), http://dx.doi.org/10.1038/nature04645.

8. A. Köhler, J. S. Wilson, and R. H. Friend, "Fluorescence and phosphorescence in organic materials," *Adv. Mater.* **14**, 701–707 (2002), http://dx.doi.org/10.1002/1521-4095(20020517)14:10<701::AID-ADMA701>3.0.CO;2-4.
9. V. Adamovich et al., "High efficiency single dopant white electrophosphorescent light emitting diodes," *New J. Chem.* **26**, 1171–1178 (2002), http://dx.doi.org/10.1039/b204301g.
10. S. Li et al., "Dendritic europium complex as a single dopant for white-light electroluminescent devices," *J. Mater. Chem.* **15**(31), 3221–3228 (2005), http://dx.doi.org/10.1039/b504738b.
11. M. Mazzeo et al., "Bright white organic light-emitting devices from a single active molecular material," *Adv. Mater.* **17**(1), 34–39 (2005), http://dx.doi.org/10.1002/(ISSN)1521-4095.
12. L. Chen et al., "White electroluminescence from star-like single polymer systems: 2,1, 3-benzothiadiazole derivatives dopant as orange cores and polyfluorene host as six blue arms," *Adv. Mater.* **23**(26), 2986–2990 (2011), http://dx.doi.org/10.1002/adma.v23.26.
13. S.-H. Eom et al., "White phosphorescent organic light-emitting devices with dual triple-doped emissive layers," *Appl. Phys. Lett.* **94**, 153303 (2009).http://dx.doi.org/10.1063/1.3120276
14. F. I. Wu et al., "Efficient white-electrophosphorescent devices based on a single polyfluorene copolymer," *Adv. Func. Mater.* **17**, 1085–1092 (2007), http://dx.doi.org/10.1002/adfm.200600697.
15. J. S. Huang et al., "Achieving high-efficiency polymer white-light-emitting devices," *Adv. Mater.* **18**(1), 114–117 (2006).http://dx.doi.org/10.1002/adma.200501105
16. G. Schwartz et al., "Harvesting triplet excitons from fluorescent blue emitters in white organic light-emitting diodes," *Adv. Mater.* **19**, 3672–3676 (2007), http://dx.doi.org/10.1002/(ISSN)1521-4095.
17. E. L. Williams et al., "Excimer-based white phosphorescent organic light emitting diodes with nearly 100% internal quantum efficiency," *Adv. Mater.* **19**, 197–202 (2007), http://dx.doi.org/10.1002/adma.200602174.
18. X. Yang et al., "Efficient blue and white-emitting electrophosphorescent devices based on platinum(II) 1,3-difluoro-4,6-di(2-pyridyl)benzene chloride," *Adv. Mater.* **20**, 2405–2409 (2008), http://dx.doi.org/10.1002/adma.200702940.
19. J. Feng et al., "White light emission from exciplex using tris-(8-hydroxyquinoline)aluminum as chromaticity-tuning layer," *Appl. Phys. Lett.* **78**, 3947–3949 (2001), http://dx.doi.org/10.1063/1.1379788.
20. F. Hide et al., "White light from InGaN/conjugated polymer hybrid light-emitting diodes," *Appl. Phys. Lett.* **70**, 2664–2666 (1997), http://dx.doi.org/10.1063/1.118989.
21. M. C. Gather et al., "On the origin of the color shift in white-emitting OLEDs," *Adv. Mater.* **19**, 4460–4465 (2007), http://dx.doi.org/10.1002/(ISSN)1521-4095.
22. M. S. Rea, *Lighting Handbook*, 8th ed., IESNA, New York (1993).
23. N. J. Turo, *Modern Molecular Photochemistry*, University Science Books, Herndon, VA (1991).
24. Z. X. Wang et al., "Facile synthesis and characterization of phosphorescent Pt(N^C^N)X complexes," *Inorg. Chem.* **49**, 11276–11286 (2010), http://dx.doi.org/10.1021/ic100740e.
25. N. Chopra et al., "High efficiency and low roll-off blue phosphorescent organic light-emitting devices using mixed host architecture," *Appl. Phys. Lett.* **97**, 033304 (2010), http://dx.doi.org/10.1063/1.3464969.
26. H. Sasabe et al., "High-efficiency blue and white organic light-emitting devices incorporating a blue iridium carbene complex," *Adv. Mater.* **22**, 5003–5007 (2010), http://dx.doi.org/10.1002/adma.v22.44.
27. Y. Sun and S. R. Forrest, "Enhanced light out-coupling of organic light-emitting devices using embedded low-index grids," *Nat. Photon.* **2**, 483–487 (2008), http://dx.doi.org/10.1038/nphoton.2008.132.
28. N. C. Giebink et al., "Intrinsic luminance loss in phosphorescent small-molecule organic light emitting devices due to bimolecular annihilation reactions," *J. Appl. Phys.* **103**, 044501–044509 (2008), http://dx.doi.org/10.1063/1.2884530.
29. S. Yiru and S. R. Forrest, "High-efficiency white organic light emitting devices with three separate phosphorescent emission layers," *Appl. Phys. Lett.* **91**, 263503-1–263503-2 (2007), http://dx.doi.org/10.1063/1.2827178.

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