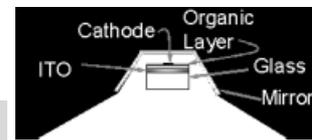**ADVANCED MATERIALS**

White Organic Light-Emitting Devices for Solid-State Lighting**

By *Brian W. D'Andrade* and *Stephen R. Forrest***

White organic light-emitting devices (WOLEDs) have advanced over the last twelve years to the extent that these devices are now being considered as efficient solid-state lighting sources. Initially, WOLEDs were targeted towards display applications for use primarily as liquid-crystal display backlights. Now, their power efficiencies have surpassed those of incandescent sources due to improvements in device architectures, synthesis of novel materials, and the incorporation of electrophosphorescent emitters. This review discusses the advantages and disadvantages of several WOLED architectures in terms of efficiency and color quality. Hindrances to their widespread acceptance as solid-state lighting sources are also noted.

1. Introduction

Solid-state organic devices are at the vanguard of a generation of electronic components that promise to be as easily manufactured as colorful magazines and newspapers. These common printed products are produced using web-processing technologies, where continuous rolls of paper measuring several feet in diameter are fed into machines to be cut, pressed, dyed, and packaged. Similar mass-production techniques for organic electronics may eventually replace traditional semiconductor batch processes, and thereby allow electronics to compete with well-established and inexpensive devices such as the incandescent bulb, and usher in new low-cost and disposable products such as electronic identification tags.

One device that is on the cusp of widespread use is the organic light-emitting diode (OLED). Research over the last decade has paved the way for the implementation of efficient blue, green, and red OLEDs in passive and active matrix displays. Low-information-content OLED displays fabricated by Philips, TDK, Nippon Seiki, Sanyo, and Pioneer have already

been commercialized, and there is significant effort under way to make laptop monitors and eventually television screens using OLED technology.

Interest in the application of white OLED (WOLED) technology for general solid-state lighting applications and flat-panel display backlights has also been steadily increasing. Indeed, the total number of refereed journal reports has more than tripled in the last two years, with numbers now exceeding twenty per year, as shown in Figure 1. Coupled to the increase in the published work in this area, the power efficiency of WOLEDs has steadily increased over the last ten years and has attained a level (see Fig. 1) requisite for WOLED acceptance into the lighting market; hence, there is a greater appre-
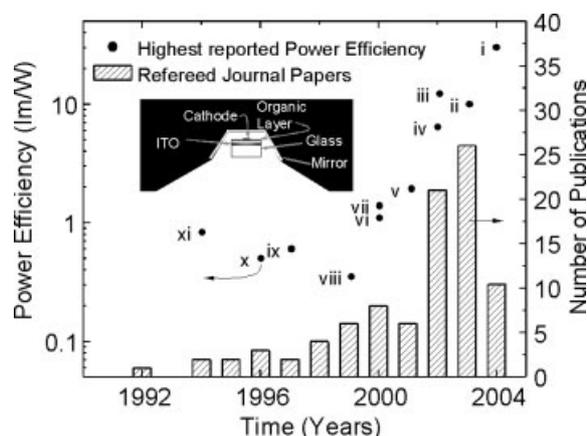

Figure 1. Time line of number of journal paper reports on white organic light-emitting devices together with the timeline of the power efficiency of WOLEDs. Inset: Example of a WOLED lighting fixture that redirects light in the forward direction.

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ciation for the potential of energy saving, thin, flexible WO-LEDs to replace traditional incandescent white light sources.

Lighting consumes ~765 TWh of electricity each year in the United States, or nearly 30 % of all electricity produced for buildings, which corresponds to 18 % of total building energy consumption. In terms of total primary energy consumption, lighting accounts for 8.3 % of all the energy used in the United States, or about 22 % of all the electricity produced. The cost for consumers to light their homes, offices, streets, and factories amounts to almost $58 billion a year.[1]

Given these figures, it is clear that increasing the efficiency of lighting by a small amount has the potential to generate tremendous savings in both cost and energy use. Incandescent lamps were developed over one hundred years ago, and still account for 42 % of the electrical energy consumed.[2] The total power efficiency (η_t) of a typical incandescent light bulb is 12–17 lm W^{-1}, whereas OLED laboratory demonstrations already have achieved $\eta_t = 30$–60 lm W^{-1}, suggesting that there are considerable advantages to be gained by using OLEDs in this application. However, questions of whether WOLED brightness, cost, and reliability can all meet the targets of the lighting industry are by no means assured. Nevertheless, the U.S. Department of Energy is anticipating that solid-state lighting in the form of WOLEDs and white inorganic LEDs will decrease national energy consumption by 29 % by 2025.[3] Provided solid-state lighting devices achieve their projected price and performance targets, the United States could accumulate energy savings of $125 billion from 2005 to 2025, defer the construction of forty 1000 MW power plants, and create solid-state lighting market revenues of $10 billion per year nationally.[3] To meet these goals over the next ten years, the necessary government and private sector investment is estimated at $1 billion.

Inorganic white LEDs appear to be capable of fulfilling some solid-state lighting needs given that such devices have achieved power efficiencies of > 44 lm W^{-1},[4] and that their operational lifetime is > 9000 h.[4] Products such as white LED flashlights are commercially available; however, some of the major challenges facing this technology for general illumination purposes are reducing the cost per lumen, obtaining an adequate green LED quantum efficiency, and finding efficient, long-life phosphorescent materials for wavelength down conversion.[3] A comparison of various approaches to white lighting sources is provided in Table 1.

Table 1. Chromaticity coordinates, color correlated temperature (CCT), and color rendering indices (CRI) for common light sources.

Lamp	x	y	CCT	CRI
High pressure sodium	0.519	0.417	2100 K	24
Xenon	0.324	0.324	5920 K	94
Tungsten Halogen (CIE Standard Illuminant A)	0.448	0.407	2856 K	100
Daylight (CIE Standard Illuminant D$_{65}$)	0.313	0.329	6500K	90
Fluorescent, cool white	0.375	0.367	4080 K	89
Fluorescent, warm white	0.440	0.403	2940 K	72
Incandescent bulb	0.448	0.408	2854 K	100

Brian D'Andrade was born and raised in Trinidad. He completed his B.Sc. in electrical engineering under the tutelage of Prof. Thomas Jackson at The Pennsylvania State University in 1999, and he received his M.A. and Ph.D. from Princeton University in 2001 and 2004, respectively. His Ph.D. thesis was on phosphorescent white organic light-emitting devices, and he is the author of over ten papers based on phosphorescent devices. Presently, he is a senior scientist at Universal Display Corporation working on the operational stability of organic devices, and on using white organic light-emitting devices as solid-state lighting sources.
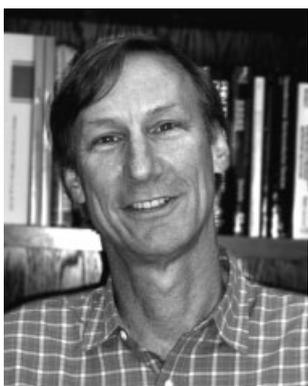
Stephen Forrest received his B.A. in Physics, in 1972, from the University of California, and an M.Sc. and Ph.D. in Physics in 1974 and 1979 from the University of Michigan. First at Bell Labs, then at the Electrical Engineering and Materials Science Departments at USC, in 1992, Prof. Forrest became the James S. McDonnell Distinguished University Professor of Electrical Engineering at Princeton University. A Fellow of the IEEE and OSA and a member of the National Academy of Engineering, he received the IEEE/LEOS Distinguished Lecturer Award in 1996–97, and in 1998 he was co-recipient of the IPO National Distinguished Inventor Award as well as the Thomas Alva Edison Award for innovations in organic LEDs. In 1999, Prof. Forrest received the MRS Medal for work on organic thin films. In 2001, he was awarded the IEEE/LEOS William Streifer Scientific Achievement Award for advances made on photodetectors for optical communications systems.

Among methods for achieving practical WOLEDs, electrophosphorescence is most effective due to its demonstrated potential for achieving 100 % internal emission efficiency.[5] Electrophosphorescence, typically achieved by doping an organometallic phosphor into a conductive host, has been successfully used to generate the primary colors necessary for display applications,[6–8] and efficient generation of the broad spectral emission required of a white light source has been reported to exceed that of incandescent bulbs.[9]

The challenges facing WOLED technology are due, in large part, to the fact that fluorescence or phosphorescence emission from typical organic materials only spans about one third of the visible spectrum. Color tuning molecules to emit in the blue, green, or red portion of the visible spectrum can be accomplished with a variety of molecular structures and their derivatives; however, a single molecule has not been designed that efficiently emits over the entire visible spectrum, such that high-quality white light is produced. Given the limited spectral bandwidth of single dopants, there are numerous WOLED architectures that combine the emission from multiple dopants.

The purpose of this review is to compare several WOLED architectures and to consider the prospects for the use of WOLEDs as practical white lighting sources. This paper is organized as follows: Section 2 begins with a short discussion on appropriate measurement and characterization of WOLED efficiency for display and lighting purposes. Section 3.1 provides a description of solution-processed WOLEDs, and Section 3.2 discusses vacuum-deposited WOLEDs with separate red, green, and blue emissive bands. WOLEDs with a single, multiply doped emissive layer are discussed in Section 3.3, and Section 3.4 elaborates on the use of broad spectral emission from excimers and exciplexes. Down-conversion phosphor systems, multiresonant cavities, and stacked WOLEDs are grouped into Section 3.5. In Section 4, we examine the theoretical limits to the power efficiency (η_p) of current WOLEDs, and suggest improvements in outcoupling efficiency needed to exceed those limits. We conclude by examining some of the challenges facing this emerging technology in Section 5.

2. Efficiency Characterization of WOLEDs

There are two critical parameters that define the color quality of a white light source: the color rendering index[10] (CRI), and the Commission Internationale de L'Éclairage[11] (CIE) chromaticity coordinates. For blackbody and non-blackbody radiators, the chromaticity can also be specified by their color temperature and correlated color temperature (CCT), respectively.[12] The CCT is the temperature of a blackbody radiator that has a color that most closely matches the emission from a non-blackbody radiator.

The color of two white-light sources may appear identical, or *metameric*, when viewed directly, and will therefore have the same chromaticity; however, the color of the reflected light from an object illuminated by these two sources may be significantly different, and therefore the light sources will have a different CRI. For high-quality white-light illumination, sources with CIE coordinates similar to that of a blackbody radiator with a CCT between 2500 K and 6500 K, and a CRI above 80 are required.[12]

Only photons emitted in the viewing direction are relevant for measuring the forward viewing power efficiency, η_p, of WOLEDs for display applications (e.g., for use as backlights in liquid-crystal displays). In contrast, standard techniques for measuring illumination-quality white-light sources must account for all emitted photons. Therefore, in the characterization of WOLEDs for these applications, one must also account for light waveguided in both the glass substrate and in the device and contact layers.[13,14] The waveguided light can exit from the sides of the substrate, or from the (back) metal cathode surface, and is therefore lost for display applications, although efficient lamp fixtures are designed to redirect light into the space being illuminated. For example, the fixture in Figure 1, inset, redirects light emitted from the sides and back of a small area WOLED into the forward direction. Hence, it is standard practice in the lighting industry[12] to state the *total power efficiency* (η_t) based on the total number of photons emitted, requiring measurement with an integrating sphere.[15]

3. WOLED Architectures

A time-line of the increase in η_p of WOLEDs listed in Table 2 is given in Figure 1. The largest increase in the efficiency of WOLEDs came when electrophosphorescent dop-

Table 2. Selected WOLED architectures with their corresponding performance characteristics. Where the color rendering index (CRI) is not reported, a maximum value is estimated from published spectral data.

Architecture	η_{ext} [a] [%]	η_p [b] [lm W^{-1}]	CIE [c]	CRI	Label	Ref.
Phosphorescent triple-doped emissive layer	12	26, 42	(0.43,0.45)	80	i	[9]
Multilayer phosphorescent.	12	10, 17	(0.35,0.36)	≤60	ii	[38]
Phosphorescent excimer.	6.4	12.2, 21	(0.36,0.44)	67	iii	[62]
Multi-emissive phosphor doped layers.	5.2	6.4, 11	(0.37,0.40)	83	iv	[36]
Two doped and one neat emissive layers.	–	1.93, 3.3	(0.35,0.34)	≤80	v	[91]
Doped blocking layers.	–	1.39, 2.4	–	–	vi	[92]
Multiple quantum wells.	–	1.1, 1.9	(0.32,0.38)	≤80	vii	[93]
Interlayer sequential energy transfer.	0.5	0.35, 0.6	(0.33,0.33)	≤70	viii	[42]
Hybrid polymer/inorganic.	1.9	0.63, 1.1	(0.34,0.29)	≤70	ix	[94]
Three neat emissive layers.	0.7	0.5, 0.9	(0.31,0.41)	≤80	x	[35]
Triple doped polymer with vacuum deposited transport layers.	–	0.83, 1.4	–	–	xi	[32]

[a] Maximum reported forward viewing external quantum efficiency. [b] Maximum reported forward viewing external power efficiency is the first number. The second is the total external power efficiency. Where not reported, it is the maximum forward viewing external power efficiency multiplied by 1.7 that assumes a small area contact (see text). [c] Commission Internationale de L'Eclairage coordinates at 100 cd m^{-2}.

ants were successfully employed. The introduction of electrophosphorescence moved the upper limit of the η_p of large-area WOLEDs from ~20 lm W^{-1} for fluorescent materials to ~80 lm W^{-1}. The maximum possible η_p of WOLEDs achievable with CRI > 80 is ~80 lm W^{-1}, provided that they have an internal quantum efficiency of 100 %, operate at the thermodynamic limit,[16] and have an outcoupling efficiency of ~20 %.[17] Assuming random exciton formation statistics, fluorescent WOLEDs are limited to internal quantum efficiencies of ~25 %,[18] and hence are impractical for illumination purposes, since this quantum efficiency limits the maximum η_p to 20 lm W^{-1}, which is comparable to the highest efficiency incandescent bulb. Unless there is significant improvement in outcoupling efficiency, fluorescent WOLEDs do not offer a significant advantage over highly inexpensive but inefficient incandescent light sources. For this reason, we will focus the following discussion on electrophosphorescent OLEDs.

For WOLEDs to be practical illumination sources, η_t at high brightness must exceed that of incandescent lamps, which produce > 1000 lm at ~15 lm W^{-1}. Current red, blue, and green OLEDs have internal quantum efficiencies of > 60 %[5,7,19] and η_p > 60 lm W^{-1} at low luminance. However, large-area WOLEDs[9] (> 600 cm^2), will be necessary to attain illumination levels > 1000 lm, since WOLEDs are generally operated at low power densities (< 1 W cm^{-2}), in contrast to inorganic LEDs that operate at > 150 W cm^{-2}. WOLED research has primarily focused on improving η_p and not on power density; however, several reports[20–22] on vertically stacked OLEDs demonstrate that high power densities are nevertheless achievable. Ideally, the area of the WOLED necessary to produce 1000 lm is directly proportional to the number of WOLEDs that are stacked, so a factor of two reduction in substrate costs is achieved by stacking only two WOLEDs.

3.1. Solution-Processed WOLEDs

For WOLEDs to gain wide acceptance in the general illumination market, the manufacturing costs must be < $3 per 1000 lm, and this target cost is difficult to realize with vacuum-deposited device architectures. Hence, consideration must be given to manufacturing electrophosphorescent WOLEDs using solution processes, where costs are significantly lower than those associated with vacuum technologies.

The simplest WOLED architectures that incorporate polymers have several emissive chromophores, which are polymers and/or small-molecules, blended[23–26] into a solution that is coated onto the transparent anode by techniques such as spin-casting,[27] or inkjet printing.[28,29] After the organic materials are printed or coated onto the substrate, metal cathodes are vacuum deposited through a shadow mask.

The ratio of holes to electrons (charge balance) in the emissive region of devices with such simple structures tend to be unfavorable for obtaining high quantum efficiency, so additional components are blended into the emissive region, or novel conjugated polymers are developed to overcome the

limitations of poor charge injection and balance.[24,30] Alternatively, additional vacuum-deposited electron-transporting and/or hole-blocking layers may be incorporated into the device structure, but this extra vacuum deposition step eliminates advantages gained by the inherent simplicity of solution processing.[31,32]

Given the problem of charge balance and other challenges such as polymer purity, the maximum power efficiency of solution processed WOLED architectures remains below values required for solid-state lighting, i.e., below 15 lm W^{-1}. An interesting approach for solution processed WOLEDs incorporates dendrimers, which have been demonstrated to have a power efficiency of 27 lm W^{-1} in green OLEDs,[33] but to date, there are no WOLEDs reported that incorporate these materials.

3.2. Multiple Emissive Layers

Small-molecular-weight WOLEDs typically consist of several stacked organic layers, each optimized to perform a particular function, such as hole or electron transport, charge confinement, and exciton recombination. The recombination current in any one of the layers can be controlled by introducing charge-blocking layers,[34] by varying layer thicknesses,[35] and by adjusting dopant concentrations.[36] By controlling the recombination current within individual organic layers, emission from red-, green-, and blue-light-emitting layers is balanced to obtain white light of the desired color purity. This method of producing white-light emission has been reported by several authors,[35,37–44] and it is frequently applied to vacuum-deposited small molecules, since the thickness of stacked layers can be closely controlled to achieve the desired color balance and efficiency.

The WOLED shown in Figure 2 (inset), employs triplet diffusion across the several separate phosphorescent emissive layers to produce bright white devices with a high η_p, and external quantum efficiency[36] (η_{ext}). Triplets have lifetimes that are several orders of magnitude longer than singlets, hence they have longer diffusion lengths, allowing emissive layers to be > 10 nm thick.[45] To achieve a desired emission color, the thickness of each layer doped with a different phosphor is adjusted to serve as a recombination zone of the appropriate fraction of excitons initially formed at the hole-transport layer/emissive region interface.

The tri-layer WOLED consisted of the three phosphorescent dopants: iridium(III) bis(4,6-di-fluorophenyl)-pyridinato-N,C^{2})picolinate (FIrpic) for blue, bis(2-phenyl benzothiozolato-$N,C^{2'}$) iridium (acetylacetonate) [Bt$_2$Ir(acac)] for yellow, and bis(2-(2'-benzo[4,5-a]thienyl)pyridinato-N,C^3) iridium (acetylacetonate) [Btp$_2$Ir(acac)] for red, doped into separate layers, each employing a common 4,4'-N,N'-dicarbazole-biphenyl (CBP) host. The color of the WOLED was tuned by varying the thicknesses of the doped layers, and their dopant concentrations, and by introducing an exciton-blocking layer (consisting of bathocuproine, or BCP) between each emissive
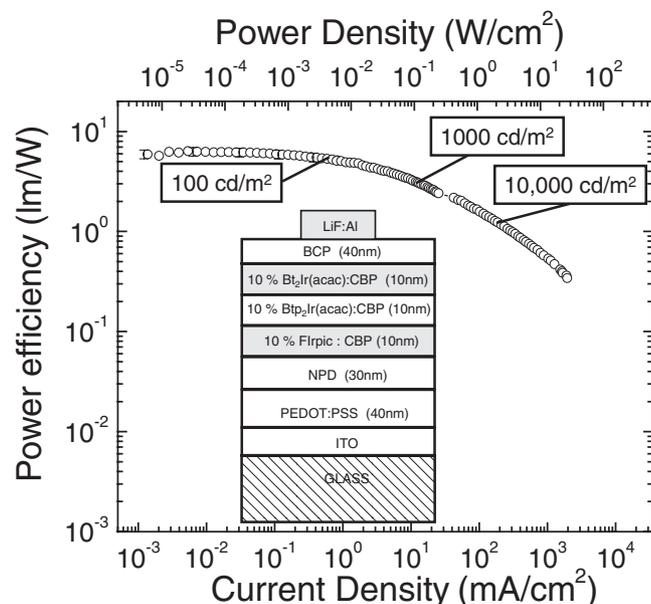
Figure 2. Power efficiency versus current density and power density of WOLED shown in inset. Inset: Schematic cross-section of the multiemissive layer WOLED consisting of 4,4′-bis[*N*-(1-napthyl)-*N*-phenyl-amino]biphenyl (NPD), CBP, 2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline (BCP) hole- and exciton-blocking layer, and the phosphor dopants FIrpic (greenish blue), Bt₂Ir(acac) (orange), and Btp₂Ir(acac) (red). Reproduced from [36].

3.3. Multiply Doped Emissive Layer

Given that organic materials have very low conductivities, organic layer thicknesses in WOLEDs must be thin to ensure low voltage (< 5 V) operation. Alternatively, the layers can be doped to improve their conductivities.[16,53] Unfortunately dopants that are used to improve conductivities (e.g., Li, Cs, and F_4-TCNQ) are generally also exciton quenching sites. The voltage across the emissive region of electrophosphorescent WOLEDs has been shown to linearly decrease and the quantum efficiency to exponentially decrease with thickness.[46] Inserting electron- and hole-blocking layers improves the quantum efficiency of devices with very thin (< 10 nm thick) emissive layers, with optimized monochromatic OLEDs recently reported with emissive layer thicknesses of only 5 nm.[16]

To ensure that all emission originates from a single thin layer, several dopants required to produce white-light emission can be mixed into a single host layer. Alternatively, a single emissive layer can be achieved using dopants that produce spectrally broad excimer or exciplex emission (see Sec. 3.4). Polymer fluorescent and phosphorescent WOLEDs with three emissive dopants mixed in appropriate proportions to produce white light have been reported.[25,26,32,54,55] Double-doped emissive layers in vacuum-deposited small-molecule WOLEDs have also been reported,[56,57] however, we are unaware of any reports of white devices with emission only generated in a double-doped host.

The WOLED shown in Figure 3 has only a single emissive layer containing three electrophosphorescent dopants: 2 wt.-%

layer. The WOLEDs had CRI > 80, CIE coordinates of (0.37, 0.40), and $\eta_p = 6.0$ lm W^{-1} at 100 cd m^{-2}, falling to 3 lm W^{-1} at 1000 cd m^{-2}, as shown in Figure 2. Analysis has shown that in the small device approximation, $\eta_t/\eta_p = 1.7$,[9] corresponding to a maximum total efficiency of $\eta_t = 10.2$ lm W^{-1} for this device.
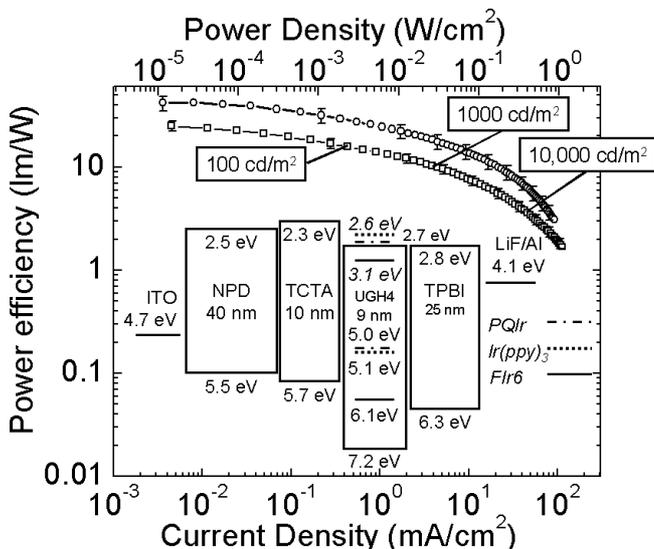
The most significant disadvantage of this structure is its relatively high operating voltage due to the combined thicknesses of the many layers used in the emission region. The average electric field[46] across the emissive and electron-transport regions of an electrophosphorescent WOLED is > 10^6 V cm^{-1}, hence the total thickness of the emissive and electron-transport layers should be kept thin (< 10 nm). Doping transport layers with Li,[16] FeCl, or 2,3,5,6-tetrafluoro-7,7,8,8-tetracyanoquinodimethane (F_4-TCNQ)[47–51] may assist in reducing voltage drops across conductive but non-emissive layers, although this further complicates the device structure and can lead to a significant reduction in operational lifetime.[52] A high concentration of both electrons and holes existing in the blocking layers can lead to undesired recombination, reducing η_{ext}, and hence additional loss in efficiency. Furthermore, using multiple emissive dopants can lead to differential aging of the various chromophores, hence causing an undesired shift in color coordinates as the device ages. Finally, multi-emissive layer devices are complicated to fabricate, and hence potentially costly, due to the large number of materials and interfaces compared with monochromatic OLEDs.

Figure 3. Total power efficiency, η_t (squares) and η_p (circles) versus current and power densities of the WOLED shown in the inset. Inset: Proposed energy level diagram of the multi-doped WOLED showing the HOMO and LUMO energies of NPD, CBP, FIr6, PQIr, Ir(ppy)₃, UGH2, TCTA = 4,4′,4″-tri(*N*-carbazolyl)triphenylamine, [99] and TPBI = 1,3,5-tris(*N*-phenylbenzimidazol-2-yl)benzene [100], and the Fermi energies of the electrodes (indium-tin-oxide = ITO and Al). Reproduced from [9].

iridium(III) bis(2-phenylquinolyl-*N*,*C*$^{2'}$) acetylacetonate[58] (PQIr) providing red-light emission, 0.5 wt.-% *fac*-tris(2-phenylpyridine) iridium [Ir(ppy)$_3$] for green-light emission, and 20 wt.-% bis(4',6'-difluorophenylpyridinato)tetrakis(1-pyrazolyl)borate[19] (FIr6) for blue-light emission, simultaneously co-doped into an "ultrawide" energy gap *p*-bis(triphenylsilyly)benzene[19] (UGH2) host. Blue OLEDs employing FIr6 in the inert host, UGH2, were shown to have direct (or resonant) charge injection and triplet exciton formation on FIr6.[19] The process of direct triplet exciton formation avoids exchange energy losses (0.5–1.0 eV) common to red, green, and blue electrophosphorescent OLEDs,[6] where host-to-guest energy transfer is employed for excitation of the emissive species. This leads to a reduction in operating voltage, and hence an increase in η_p. Additionally, device characteristics that are essential for achieving high external power efficiency in triple-doped WOLEDs is achieved using thin layers for low voltage operation, and confining charge and excitons within the emissive layer.

Confinement of charge and excitons reduces the loss of either species from the emissive layer, which results in a reduction in internal quantum efficiency. The schematic energy level diagram of the layers in the triple-doped WOLED is shown in Figure 3, inset. The effectiveness of charge confinement relies on differences between the lowest unoccupied molecular orbital (LUMO) and highest occupied molecular orbital (HOMO) energy levels of the emissive and adjacent layers. As in Figure 3, the LUMO of 4,4',4''-tri(*N*-carbazolyl)-triphenylamine (TCTA) is ≥0.3 eV than that of the dopants and host, thus preventing electron leakage from the emissive layer. In addition, the 0.2 eV energy barrier between the HOMOs of FIr6 at 6.1 eV, and 1,3,5-tris(*N*-phenylbenzimidazol-2-yl)benzene (TPBI) at 6.3 eV, acts as an efficient barrier to hole transport across the interface. Exciton diffusion from the emissive layer towards the electron- and hole-transport layers is also prevented by employing materials with energy gaps greater than that of the blue dopant. Here, TCTA and TPBI have energy gaps of 3.4 and 3.5 eV, respectively. These barriers improve charge balance within the 9 nm thick emission layer, and hence the recombination efficiency, leading to improvement in η_{ext}.

The triple-doped WOLEDs have a peak $\eta_t = 42$ lm W^{-1} at low intensities, and $\eta_p > 10$ lm W^{-1} at 1000 cd m^{-2}. The dependence of η_t (circles) and η_p (squares) with current and power density are shown in Figure 3, and the ratio $\eta_t/\eta_p = 1.7$ is obtained from these data. Now, $\eta_t/\eta_p > 1$ for small-area devices since η_p is found by measuring only light emitted from the substrate surface; however η_t is measured in an integrating sphere and hence also includes light emitted from the edges and top substrate surface.

An advantage of the triple-doped WOLED structure is the possibility that the white color can remain unchanged throughout the operational lifetime of the device (i.e., there is no differential color aging as in the multiple-layer emitter), given that only the blue dopant conducts the charge, and is the sole site for direct exciton formation. Once the exciton is generated, energy transfer to the green and red phosphors ensues to produce balanced emission. Hence, as the blue-light emission decreases with age, the green-light and red-light emission should decrease proportionally, since their relative intensities are directly linked to that of the blue phosphor.

3.4. WOLEDs Based on Excimer and Exciplex Emission

One promising approach to reducing the number of dopants and structural heterogeneities inherent in the multiple layer architecture (see Sec. 3.2) is to employ a lumophore that forms a broadly emitting exciplex (i.e., an excited state whose wavefunction overlaps a neighboring, dissimilar molecule). Fluorescent exciplex OLEDs have been demonstrated with CIE coordinates of (0.33,0.33), close to that of the ideal white light source. Some of the best fluorescent exciplex results include an external quantum efficiency[59] of $\eta_{ext} = 0.3$ %, a luminance efficiency[60] of $\eta_p = 0.58$ lm W^{-1} and a maximum luminance[60] of 2000 cd m^{-2}. These performance values lie well below those needed in practical lighting applications.

In contrast, phosphorescent excimer emission from platinum(II)(2-(4',6'-difluorophenyl)pyridinato-*N*,*C*$^{2'}$)(2,4-pentanedionato) (FPt1), coupled with blue-light emission from monomer FIrpic have been shown to result in broad white-light emission from efficient electrophosphorescent WOLEDs.[61] In contrast to an exciplex, an excimer is an emissive excited state whose wavefunction overlaps two adjacent molecules of like composition.

Both excimers and exciplexes lack a bound ground state, and hence provide a unique solution to achieving efficient energy transfer from the host to the light-emitting centers. Due to the absence of a bound ground state, for example, an excimer prevents the cascade of energy from the host, and from higher-energy (blue) dopants to the lower-energy (excimer-orange) dopant. Complicated intermolecular interactions, which make color balancing using multiple dopants problematic, are thus eliminated. Hence, by using only one,[62] or at most two dopants,[61] coverage of the entire visible spectrum has been achieved with an electrophosphorescent excimer WOLED. Figure 4 shows the power efficiency characteristics of a single-dopant excimer WOLED, whose schematic cross-section is shown in Figure 4, inset. Here, $\eta_p = (4.1 \pm 0.4)$ lm W^{-1} and decreases to (1.2 ± 0.1) lm W^{-1} at 1000 cd m^{-2} and 10000 cd m^{-2}, respectively. A maximum $\eta_p = (11 \pm 1)$ lm W^{-1} is obtained at low luminance.

Two key features of the device in Figure 4 are the electron-blocking layer,[62] consisting of *fac*-tris(1-phenylpyrazolato-*N*,*C*$^{2'}$)iridium(III) [Ir[ppz]$_3$], and the thin emissive region. The Ir(ppz)$_3$ blocking layer increases the recombination efficiency of electrons and holes in the emissive region by preventing escape of electrons into the hole-transporting material, 4,4'-bis[*N*-(1-napthyl)-*N*-phenyl-amino]biphenyl (NPD). The emissive region thickness, consisting of *N*,*N*'-dicarbazolyl-3,5-benzene (mCP) and FPt1, is optimized for high power efficiency by keeping the voltage drop to a minimum while maximizing the recombination efficiency.[46]
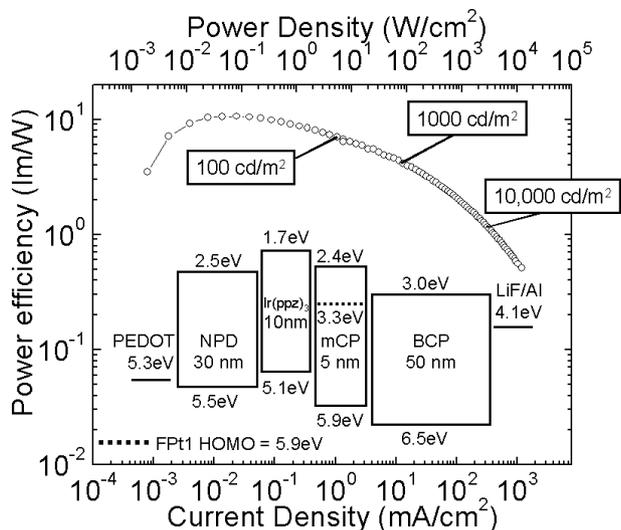
Figure 4. Power efficiency, η_P (circles), versus current and power densities of the excimer-based WOLED shown in inset. Inset: Proposed energy level diagram of the excimer WOLED showing the HOMO and LUMO energies of NPD, Irppz, mCP, BCP, and FPt1, and the Fermi energies of the electrodes (PEDOT:PSS and Al) (data from [62]).

known to form excimers when positioned in close proximity in concentrated solutions and thin films.[63–67] For example, FPt1 shows monomer emission in a 10^{-6} M solution of dichloromethane, and excimer emission in a concentrated 10^{-3} M solution. Neat films of FPt1 show no additional absorption peaks characteristic of dimer emission. Furthermore, the photoluminescence excitation spectra of the monomer and excimer peaks are similar, suggesting they have a common excitation pathway, which would not be the case for a different (e.g., dimer) species.

Beyond the inherent simplicity of this particular approach is the possibility for reduced differential color aging in single dopant excimer devices. Given that only a single species is responsible for both blue-light (monomer) and orange–red-light (excimer) emission, it is anticipated that the color coordinates for emission will not shift with device age. However, the square Pt complex operational lifetime remains inadequate for most lighting applications.

To achieve efficient excimer emission, control of the dopant concentration is crucial. Square-planar Pt complexes are

3.5. Alternative WOLED Architectures

An alternative white-light emitter comprised of separately energized red, green, and blue regions is shown in Figure 5a.[22] The device separates each of the three dyes into blue, green, and red OLEDs stacked one on another with four
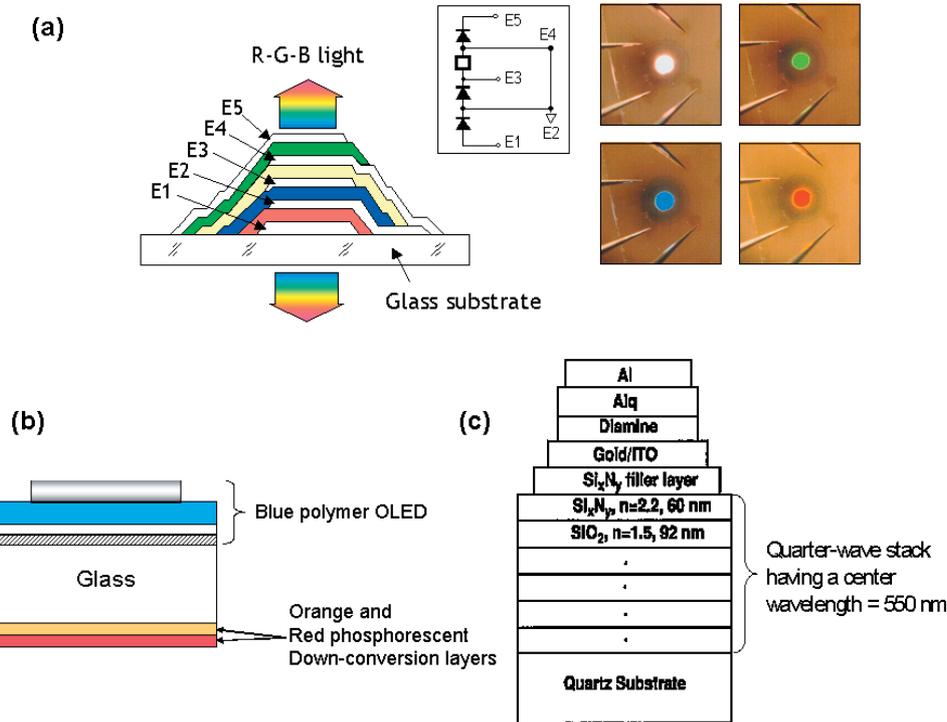
Figure 5. Schematic structures of several different WOLED designs. a) Stacked OLED (SOLED). Red, green, and blue OLEDs are stacked such that they can be independently driven to produce green, red, blue, or white light as shown. Also shown is the equivalent circuit and biasing schematic for this device. Reproduced from [22], with permission: copyright 1997, Elsevier. b) A blue polymer OLED with orange and red down-conversion layers deposited onto the opposite substrate surface. See [69]. c) Structure of a multimode microcavity OLED. Reproduced from [72], with permission: copyright 1994, American Institute of Physics.

electrodes and three voltage sources independently controlling the emission from each of the devices. This so-called stacked OLED, or SOLED, is a versatile WOLED that allows for independent tuning of the color mix, thereby providing for tuning of the CRI and chromaticity over a wide range. Furthermore, this tuning strategy can delay the onset of differential aging of the several emitting layers. It has been shown that by layering several devices in this manner, a high total brightness can be achieved without driving any particular element in the stack at such a high intensity that its operational lifetime is reduced.[68]

A variant of the SOLED that allows the contacts between intermediate OLEDs in the stack to electrically "float" has also been demonstrated by Liao et al., and Kido et al.[20,21] These simplified stacked structures perform as a series of independent OLEDs, with a single electron exciting the multiple OLEDs as it passes though the circuit. Hence, very high luminance without a significant loss in power efficiency is achieved, thereby once again reducing the aging in high intensity lighting sources. In the device of Liao et al.,[20] multiple green electrophosphorescent devices were stacked with intervening charge recombination zones consisting of NPD doped with $FeCl_3$, with results shown in Figure 6. It is apparent that the efficiency (and brightness at a particularly current density)
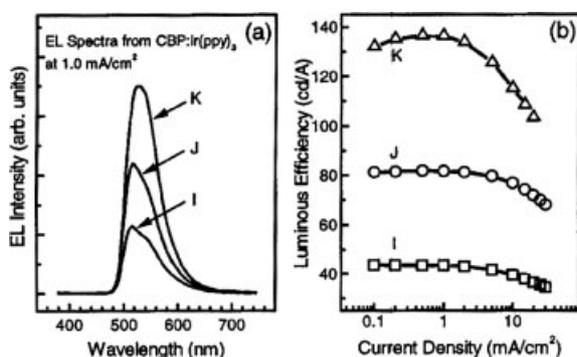
Figure 6. a) Electrophosphorescence spectra of one- (I), two- (J), and three-layer (K) stacked, green-light-emitting devices. b) Luminous efficiency versus current of those same devices, showing the linear dependence of efficiency on the number of elements in the stack. Reproduced from [20], with permission: copyright 2004, American Institute of Physics.

scales linearly with the number of elements in the stack, reaching a peak efficiency of 130 cd A^{-1}. To date, only monochrome multielement (or tandem) OLEDs have been produced, although it should be straightforward to place a different color emitter in each position in the stack to get the desired white-light emission. However, unlike the independently contacted SOLED, these simplified devices do not have the ability to tune or compensate for changes in color coordinates as a particular element in the stack ages.

A nearly similar concept to the stacked, tunable emitter is to place red-, green-, and blue-light emitters side by side, per-

haps in strips. If spaced sufficiently closely, the colors will merge, as in a full color display, once again producing white light. This device can be equally as bright and efficient as a SOLED emitter, with the possibility that the side-by-side geometry is somewhat less complex to manufacture. Indeed, the ultimate acceptance of such devices will depend on cost and yield issues.

Another approach employs a blue OLED fabricated on one side of a glass substrate, and green and red phosphorescent down-conversion layers deposited onto the opposite side of the substrate, see Figure 5b.[69] The use of one emitting device that is optically coupled to green and red down-conversion layers may result in improved, long-term color stability. These down-conversion WOLEDs demonstrated the highest CRI value, 93, yet reported, and modest $\eta_p = 3.8$ lm W^{-1}. Light scattering[70] in the down-conversion layers improves the outcoupling efficiency. The disadvantage of the approach is the loss of power efficiency incurred by generating a blue photon, which then pumps a green- or red-light-emitting phosphor. This power efficiency loss is similar to that incurred in the triple-doped WOLED since excitation of the blue-light-emitting phosphor in that case also leads to further low energy (and hence lossy) excitation of the red and green dopants in the emissive layer.

Finally, WOLED architectures employing multimode resonant cavities have also received some attention.[71–73] In this architecture, several modes of an optical cavity formed between the reflecting metallic cathode and a dielectric mirror under the transparent anode overlap the free-space emission spectrum of an organic material, such that two or three modes are selected to result in apparent white-light emission. For example, fluorescent emission from tris(8-hydroxyquinolinato) (Alq$_3$) coupled into two modes with full-width at half maximum 25 nm and centered at 480 nm and 650 nm has been shown to produce white light with CIE coordinates (0.34, 0.39) (see Fig. 5c). For this device, $\eta_{ext} < 1$ % and CRI < 70, although further optimization is possible. One disadvantage of this approach is that the color coordinates change with viewing angle, making the use of microcavities unacceptable in many lighting applications.[74]

4. Improving Outcoupling Efficiency

For practical WOLEDs with an area > 600 cm^2, a negligibly small fraction of the light exits from the side of the substrate, in which case $\eta_p \approx \eta_t$. Additionally, a large-area metal cathode results in significant absorption losses (approximately 15 % upon each reflection at the organic–metal interface). Therefore, efficient light outcoupling in the forward or backward direction, or utilizing structures with highly reflective (> 95 %) metal, or thin (< 150 nm) indium tin oxide (ITO) cathodes, is essential.
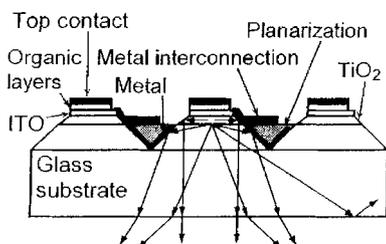
Table 3 compares the area required to produce a total brightness of 800 lm (approximately the brightness of a 50 W incandescent light bulb) for triple-doped WOLEDs operating

Table 3. Selected outcoupling schemes for a triple-doped WOLED emitting a total of 800 lm and operating at 12 lm W^{-1}, which is comparable to an approximately 50 W incandescent bulb.

Outcoupling scheme	F [a]	Area [b] [cm^2]	Ref.
Conventional WOLED	1.0	7100	[9]
Top emitting	1.2	3200	[95]
Nanopatterning of glass substrate	1.5	1400	[96]
Microlens array	1.5	1400	[97]
Silica aerogels	1.8	750	[98]
Shaped substrates	1.9	650	[17]

[a] Factor of outcoupling efficiency improvement over conventional WOLED. [b] Active area of WOLED.

at 12 lm W^{-1} (see Sec. 3.3) using five different outcoupling schemes. The most effective outcoupling method employs shaped substrates (see Fig. 7) that emit all locally generated light into the forward viewing direction.[17] In this case, producing 800 lm requires a surface area of only 650 cm^2, which

Figure 8. Scanning electron micrographs of microlens arrays that have demonstrated an increase in external efficiency by a factor of 1.5. Inset: Detail of the 10 mm diameter lens arrays fabricated by pouring an elastomeric polymer used for the lenses into a silicon mold. Reproduced from [97], with permission: copyright 2002, American Institute of Physics.
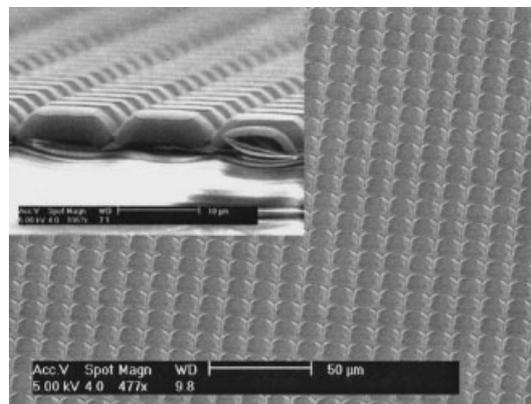

Figure 7. Schematic diagram of shaped glass substrates that improve outcoupling efficiency by a factor of 1.9. Reproduced from [17], with permission: copyright 1997, Optical Society of America.

may be further reduced by employing stacked devices[21] as mentioned in Section 3.5. Microlens arrays as shown in Figure 8 provide a potentially lower cost alternative to shaped substrates, although in this case the area needed to produce 800 lm is more than doubled to 1400 cm^2. Hence, in most cases, a tradeoff ultimately exists between cost (which is a function of device area) and outcoupling efficiency.

5. Conclusion

Research on white-light-emitting OLEDs can benefit from previous work on inorganic semiconductors, since many aspects of growth and processing that presented hurdles to the realization of inorganic illumination sources are the same as are currently being faced by organic thin film devices. Among the most important challenges are achieving high material purity, low cost, high brightness, and long operational lifetimes.

For emissive devices, impurities may result in exciton quenching, and hence decrease the emission efficiency, increase device series resistance, and decrease operational stability. Low material purity also can impact device manufac-

turing yields, since the WOLED performance characteristics will not be reproducible if the materials properties vary from run to run. Materials with very high purities (~ 99.9999 %) are used for inorganic semiconductor devices, and are commonly commercially available. Typical organic materials, on the other hand, have purities of ~ 99.95 % after two or three recrystallization and gradient sublimation[75] steps, and the cost, time, and effort needed to obtain ultrapure organic materials[76–78] are significant. Furthermore, the measurement of purity of an organic source is generally done by spectroscopic techniques whose accuracy typically does not allow for detection of impurities with concentrations < 0.1 %.[75]

Ideally, the design and optimization of a WOLED should be based on analytical models, but lacking these, engineers continue to rely on combinatorial processes that are slow and costly. Accurate theories of the operation of a WOLED, including current–voltage characteristics, exciton diffusion and formation, and emission outcoupling do not currently exist.

In addition to these basic problems, the challenges preventing the widespread manufacturing and use of the WOLED for solid-state illumination are four-fold: cost, efficiency, low outcoupling efficiency (see Sec. 4), and low operational stability. Effective competition for a share of the highly competitive lighting market is based on the potential of WOLEDs for yielding extremely inexpensive yet efficient sources. Typically, incandescent light sources have lifetimes of ~ 1000 h, with fluorescent sources perhaps ten times that number. Indeed, at a minimum, the total cost of ownership of the WOLED throughout its operational life must present a significant improvement over incandescence.

To date, the most efficient WOLEDs are grown in high-vacuum systems using thermal evaporation sources. New roll-to-roll manufacturing techniques offer the best opportunity for mass production of low-cost WOLEDs, and these processes may eventually make the cost of producing WOLEDs compe-

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titive, but are not yet available. Particularly promising for low-cost manufacturing is liquid-based processing adaptable to polymer WOLEDs, such as spray-on or spin-on deposition.[79] However, until polymeric WOLEDs can approach the efficiency of small molecular weight devices, they will not be practical in most solid-state illumination applications. Thus, work with solution processable molecular systems, and in particular dendrimers, has potential in the near term.[23,24,80–82]

Another means for deposition of small molecules that can lead to very low cost, web and roll-to-roll processing is low-pressure organic vapor phase deposition (OVPD).[83–86] This recently commercialized technique[87] has improved control over doping compared with vacuum thermal evaporation, and is adaptable to rapid, particle free, uniform deposition of organic films on large-area substrates.

Another challenge is to achieve WOLEDs with efficiencies ~ 30 lm W^{-1} at 1000 cd m^{-2}. New materials are being developed and optimized that are pushing the fundamental efficiency limits. In developing new materials, we note that ultimately the largest hurdle to overcome prior to WOLED commercial acceptance is operational stability. To be useful for lighting applications, the lifetime of a WOLED operating at 800 cd m^{-2} must exceed 1000 h. The lifetimes of the best green and red electrophosphorescent devices exceed 15 000 hrs at 100 cd m^{-2},[88–90] however, blue device lifetimes are significantly shorter and therefore currently present the most severe limits to WOLED lifetime.

Although the challenges facing WOLEDs are significant, we are optimistic that this technology will meet practical goals and expectations in the near future. Indeed, research in organic electronics is vibrant and expanding, and the future of WOLEDs is bright.

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