Recent Advances in White Organic Light-Emitting Materials and Devices (WOLEDs)

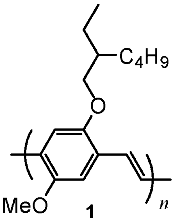
By *Kiran T. Kamtekar,* Andrew P. Monkman,** and *Martin R. Bryce**

WOLEDs offer new design opportunities in practical solid-state lighting and could play a significant role in reducing global energy consumption. Obtaining white light from organic LEDs is a considerable challenge. Alongside the development of new materials with improved color stability and balanced charge transport properties, major issues involve the fabrication of large-area devices and the development of low-cost manufacturing technology. This Review will describe the types of materials (small molecules and polymers) that have been used to fabricate WOLEDs. A range of device architectures are presented and appraised.

1. Introduction to OLEDs

Since the publication of the first low-voltage organic light-emitting diode (OLED) by Tang and VanSlyke at Eastman Kodak in 1987,[1] intense academic and industrial interest has resulted in a vast number of publications and patents in the area. Since the initial device—a combination of a hole-transporting aromatic diamine and the fluorescent green tris(8-hydroxyquinoline) (Alq$_3$)—research has concentrated on delivering suitable molecules for emission of the three primary colors: red, green, and blue. Many novel systems have been designed, synthesized, and tested as functional materials for OLED applications. The landmark discovery of electroluminescence using a conjugated polymer as the emitter in Cambridge in 1989[2] led to the development of solution-processable OLED materials (viz. polymer light emitting devices, PLEDs). Initial research concentrated on poly(phenylenevinylene) (PPV)[2] and poly(*para*-phenylene) (PPP) systems.[3] Key developments were provided by soluble polymers that enabled high quality thin films to be prepared via spin coating onto solid substrates. Two of the most widely studied families of conjugated polymers are based on (i) PPV, e.g., the orange-red emitting MEH-PPV **1**[4] and "Super-Yellow," a PPV derivative developed by Covion Semiconductors GmbH,[5] and (ii) poly(9,9-dialkylfluorene)s, e.g., the blue-emitting PFO **2**.[6,7] Over the past two decades, many reviews have been published on the development of organic/polymeric LED materials.[8–11]

WOLEDs have been studied as the backlight for flat panel displays and as full color LEDs when combined with color filters. However, their greatest promise is in the arena of solid-state lighting.



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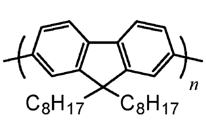

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2. Introduction to Solid-State Lighting (SSL)

The common incandescent light bulb dates from the original U.K. patent in 1860 by Joseph Swan and was commercialized shortly afterward when Swan joined forces with his American rival Thomas Edison. One and a half centuries later and the technology is still found in most homes. However, incandescent lights are very inefficient. They convert only ca. 5% of the input electrical energy into visible light; ca. 95% of their output energy is emitted in the infrared as heat, not illumination.[12] While the low power efficiencies of incandescent bulbs are well known (ca. 15 lm W^{-1}), the power efficiency of fluorescent tubes is often quoted at above 100 lm W^{-1}.

The fluorescent tube also has several drawbacks: the quoted efficiency is for a bare lamp at room temperature. When the lamp is assembled into a finished product such as a luminaire, the efficiencies are greatly reduced to the 30–40 lm W^{-1} range. Additionally, the lifetimes quoted by suppliers assume the light is operated continuously in an ideal environment (usually stated at 25 °C, whilst most lights operate far above this temperature). Switching the light on and off, as in a typical residential setting, has been shown to vastly reduce lifetimes of the lights.

The spectral coverage of the tubes is often poor leading to low color-rendering index (CRI) which is a measure of how true the color of an object appears under illumination, compared to a reference

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source of illumination—a black body source. Fluorescent tubes contain small amounts of mercury (ca. 5 mg per tube) that can create environmental hazards upon breakage or disposal of the product. At present, lighting accounts for ca. 19% of the total global energy consumption, with worldwide demand for lighting rapidly increasing, so any new solutions must help reduce this value.[12–14]

White light has three characteristics: (i) the CIE (Commission Internationale d'Eclairage) coordinates (the chromaticity); (ii) the color temperature (CT), and (iii) the color rendering index (CRI). However, to be meaningful, both the CRI and CT should be quoted as the CRI is measured relative to a reference of a given CT. A black body radiator, by definition, has a CRI of 100. Traditionally, the efficiency of OLEDs is reported as either their external quantum efficiency (EQE) or in candelas per amp (cd A^{-1}). However, lighting sources are usually quoted in terms of power efficiency (or efficacy), i.e., the efficiency taking into account the response of the human eye, which is sensitive to light only in the visible spectrum (ca. 400–700 nm) with a maximum sensitivity to green light (555 nm). Power efficiency is reported in lumens per watt which for the most ideal monochromatic green light source has a maximum value of 683 lm W^{-1}. For white light with a CRI of 90 the maximum value is 408 lm W^{-1} and for a CRI of 100 it is 240 lm W^{-1}.[15]

Whilst the white point, or equal energy point, on the CIE chart is defined as (0.33, 0.33) the desired coordinates for lighting fall along the black body curve with color temperatures between ca. 3000–10 000 K (Fig. 1). Different markets have

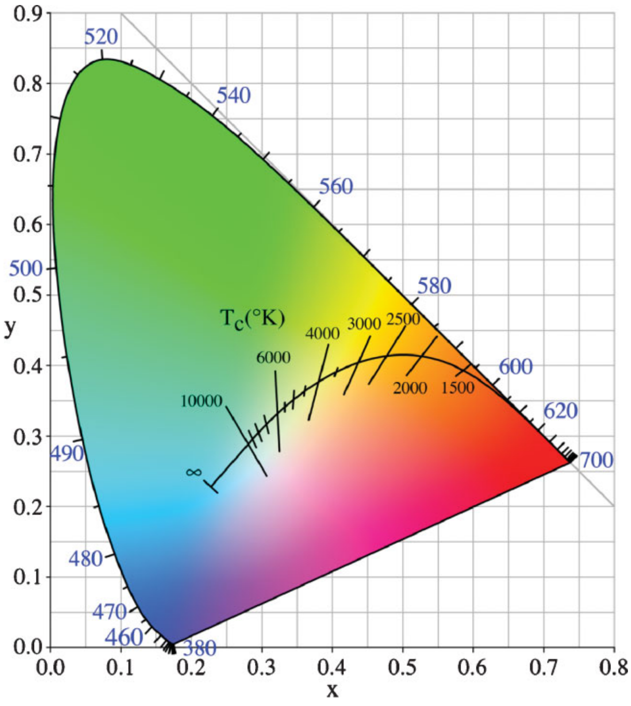

Figure 1. CIE (*x*,*y*) chromaticity diagram. All the colors in the visible spectrum lie within or on the boundary of this diagram. The internal arc is the Planckian locus, which is the plot of the coordinates of black body radiation at the temperatures shown, described as color-correlated temperatures.

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Andrew Monkman obtained his degree and PhD from the University of London. In 1988 in became lecturer in Molecular Electronics at the University of Durham. He was appointed Professor of Physics at Durham in 2002. Currently, Professor Monkman is Director of the Durham Photonic Materials Center. Current research themes range from femtosecond laser spectroscopy, triplet exciton dynamics and lasing in luminescent polymers to the development of novel phosphorescent materials and spectroscopic measurements of materials in OLED devices.

Martin Bryce obtained his PhD from the University of York. After postdoctoral positions at UBC Vancouver and Bristol University, he moved to Durham University, initially as a lecturer. He was appointed Professor of Chemistry in 1995. He is currently Director of the Durham University Center for Molecular and Nanoscale Electronics. His research includes the synthesis of new functional π–systems, fluorophores and phosphors for OLEDs, materials for single-molecule electronics and novel heterocycles.

different requirements. For example, warm countries prefer "cooler" >ca. 5000 K light, whilst colder countries prefer the "warmer" <ca. 5000 K light. Thus, the ability to simply change the color of the emitted light by altering the materials during production is greatly desired.

Inorganic LEDs are currently entering the marketplace, but suffer from a range of problems and are mainly suitable for point- source illumination rather than surface-source illumination. A review of the status of inorganic SSL has recently been published.[15] WOLEDs must offer significant benefits over fluorescent technology and inorganic LEDs in order to gain traction in the lighting market. WOLED developers envisage luminescent walls or ceilings with uniform emission from a large surface area.[16] The applicability of OLEDs to lighting applications has been touted for a long time. However, the lighting industry and its customers have very high requirements in terms of product specification. The introduction of OLEDs promises to revolutionize the way light sources are designed. WOLEDs represent a move away from bulky bulbs and tubes allowing designers and architects to plan new and more elegant ways to introduce light into buildings. Developments such as transparent WOLEDs and flexible substrates promise many new innovations in this regard. Most OLED lifetimes are quoted as the time to decay to half of the initial brightness (T_{50}), whilst lighting products are typically quoted as the time to decay to 70% of the initial brightness (T_{70}).

Since the first reports of white-emitting devices by the group of Kido,[17,18] many approaches to generating white light from organics have been described in the literature. The initial devices exhibited efficiencies of <1 lm W^{-1} but this value has grown to >50 lm W^{-1} over the last few years.[19] White emission from OLEDs can now be achieved in both small molecule and polymer systems. Reviews which focus on thermally evaporated small molecules[20] and solution processable polymers have been published.[21] This review will highlight recent progress on WOLEDs from a materials perspective, with representative examples covering the literature up to May 2009. We note that there are no universally recognized standards for reporting OLED performance. Both the scientific and commercial literature use proprietary techniques for calculating the key parameters such as efficiency, turn-on voltage, and lifetime and frequently not all the relevant data (EQE, power efficiency, CT, CIE coordinates, CRI, and device lifetimes) are reported.

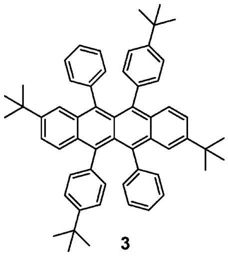
3. Energy Transfer

Excitons formed in a high band gap host material are transferred to any lower band-gap materials via either long range Förster resonance energy transfer (dipole-dipole interaction) of singlet excitons, or shorter range Dexter transfer of triplet excitons. Exciton diffusion through the host material can greatly increase the range over which these processes occur.[22] If the relative levels of high and low band gap materials are balanced, it is possible to saturate the lower energy emitter and obtain emission from both host and dopant(s). However, it is very difficult to control the energy transfer between the R, G, and B materials to attain balanced white emission under different bias conditions. Consequently, voltage dependent EL is often observed, especially if more than one dopant is used.

For polymeric systems, it is a considerable challenge to attain the low highest occupied molecular orbital–lowest unoccupied molecular orbital (HOMO–LUMO) gaps required to achieve good charge injection from both low work function metals and ITO, whilst at the same time having sufficiently high triplet energies to support green or blue phosphorescent dopants. An example from Philips used *meta*-linkages between 3,3′-biscarbazole and 2,5-diphenyl-1, 3,4-oxadiazole units to ensure a limited conjugation length in copolymer **20** and therefore raise the triplet level to ca. 2.6 eV.[23] Copolymer **20** is able to host the soluble green iridium dopant **21** as an emitter without quenching, attaining a luminous efficiency of 23 cd A^{-1}, independent of current density (20–500 A m^{-2}).[24] However, polymer systems capable of hosting blue phosphorescent dopants have not yet been reported so this strategy to white EL is limited to using orange-red phosphorescent dopants with blue and green host polymers.

4. Multilayer Devices

In this approach, two or more emissive materials are employed to emit simultaneously colors that are together perceived as white. This technique is primarily based on evaporation of small molecules. In addition to providing multiple emissive layers, this technique also uses additional charge transport layers to minimize the barriers to charge carriers. For example, Wu et al. fabricated an eight-layer device stack containing two emissive blue layers and a complementary yellow emissive layer to give a two-color white. Multiple charge-transport/blocking layers served to suppress the change in EL spectra with drive current density.[25] The device achieved efficiencies of 3.9% EQE (9.9 cd A^{-1}, 2.7 lm W^{-1}) with CIE coordinates of (0.31, 0.41). The emissive materials in this device are the fluorescent molecules TBRb **3** (yellow) and DSA-Ph **4** (blue).



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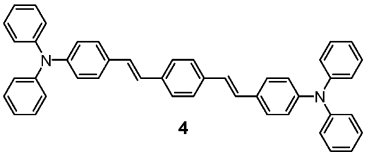

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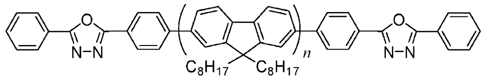
Few solution-processed multilayer devices have been reported due to the nature of their fabrication. In depositing sequential layers, the solvents can redissolve previously deposited layers. One way to circumvent this problem is to use materials that can be deposited from orthogonal solvents. A three-layer device was fabricated consisting of an emissive layer of polyfluorene host **5** and iridium dopant **6** (both soluble in organic solvents; insoluble in water) sandwiched between a water-soluble PVK derivative **7** as a hole-transport layer (HTL) and a water-soluble PBD derivative **8** as an electron-transport layer (ETL).[26] Devices turned on at ca. 6 V and emitted white light via energy transfer (see Section 3) with coordinates of (0.33, 0.33), color temperature ca. 6400 K, and efficiencies of 10.4 cd A^{-1}, 3 lm W^{-1}, and CRI of 92. The emission spectra were insensitive to the applied voltage in the range 7–20 V, thus, the brightness could be increased without compromising the quality of the white light.



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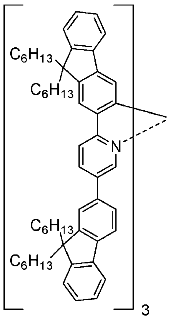

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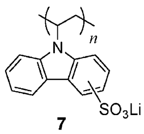

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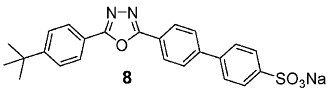

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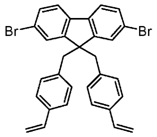
A different approach involves rendering deposited layers insoluble through post-deposition treatment. Crosslinking strategies have been put forward by several groups.[27] Miller and co-workers at IBM endcapped polyfluorenes with 4-vinylbromobenzene[28] and subsequently used crosslinkable HTLs to afford a three-layered solution-processed device.[29] Dow developed 9,9-di(4-vinylbenzyl)fluorene monomers **9** and both Dow and HP studied benzocyclobutane (BCB) groups, e.g., **10**, for crosslinking purposes.[30] Small amounts (feed ratios ~5%) are added into the polymerization reaction and after the polymers have been deposited, the substrate is heated to initiate an autopolymerization reaction which renders the polymer film insoluble allowing further layers to be deposited. An added benefit is that the crosslinked film usually has an increased glass transition temperature which imparts higher stability to the devices, which is manifested as longer device lifetimes.



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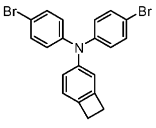

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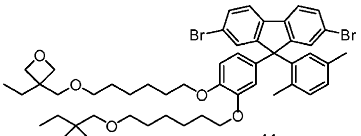
Recently, the groups of Meerholz and Merck GmbH have used pendant oxetane units as crosslinkers.[27,31–33] The functionalized monomer or stand-alone units, e.g., **11** and **12**, are incorporated into the polymers in small amounts. The crosslinking occurs by irradiating the film with UV light in the presence of a photoacid to generate H^+ leading to cationic ring-opening polymerization (CROP) of the oxetane. In addition to rendering the films insoluble, the crosslinking of oxetane units leads to minimal volume shrinkage of the film (ca. 5%) and thus delamination from the substrate is avoided.[27]



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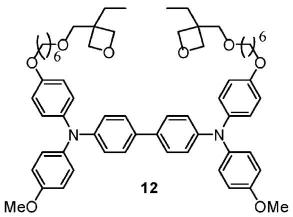

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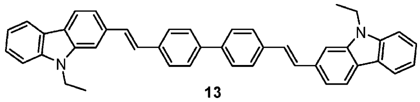
Using masks to crosslink only certain areas of a film allows for simple solution-lithography to yield white light from parallel films of red, green, and blue copolymers.[34] This approach enables the RGB colors to be varied to negate their differential aging. Optimization of the curing procedure enabled features with line widths of 2 μm to be fully resolved. This approach also overcomes the effect of charge-trapping which leads to color-shifts with different driving voltages in single copolymers.[35]

Meerholz and co-workers have shown that PSS present in the hole injection layer (HIL) can act as a photoacid which leads to crosslinking migrating from the HIL interface upward.[36] As the devices are fabricated in inert conditions, there is no formal termination step to the CROP mechanism which allows subsequent layers to be crosslinked with the same source of protons, enabling true multilayer organic solution-processed devices to be fabricated. If the UV illumination is removed before the crosslinking has reached the film surface, then the uncrosslinked polymer can be washed away to give uniform films. This removes the necessity to spin or print uniform films. This approach can also be used to allow the surface of the crosslinked polymer to mimic that of the substrate/PEDOT:PSS.

A breakthrough in multilayer WOLED efficiency came in 2006 when the groups of Thompson and Forrest achieved 24 lm W^{-1} and 18% EQE with a CRI of 85 and high color stability with varying current density over the range 1–100 mA cm^{-2}. The eight-layer device harvested singlet excited states with the blue fluorescent singlet emitter **13**, while the triplet states were transferred to the red PQIr and green $Ir(ppy)_3$ complexes **14** and **15**, respectively.[37] All the luminophores were doped into 4,4′-bis(*N*-carbazolyl)biphenyl (CBP) host (each in a separate layer) with additional HT and ET layers. The device included (undoped) buffer layers of CBP around the triplet emitters to keep the singlet excitons in the blue layer which was sited at the recombination zone, while the triplet excitons, with their longer diffusion lengths, can reach the red and green dopants situated away from the recombination zone. The proposed energy transfer mechanism is shown in Figure 2.



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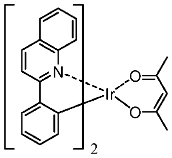

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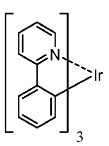

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Leo and co-workers postulated that the efficiencies of such a device stack would be limited by the use of a common host material with a triplet energy below that of the blue fluorescent

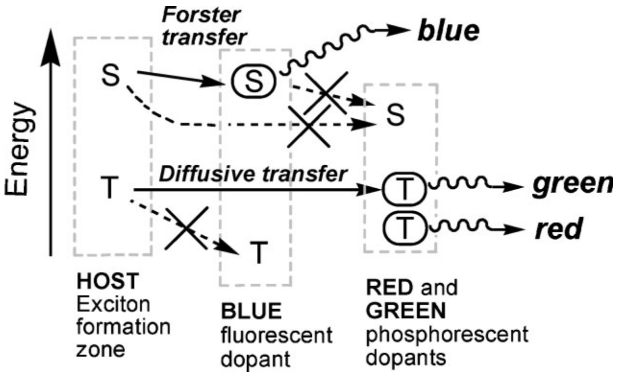
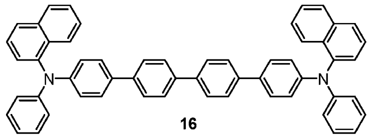


Figure 2. Proposed energy transfer mechanisms in a fluorescent/phosphorescent WOLED. This illustrates the separate channels for triplet (T) and singlet (S) formation and transfer directly onto their corresponding emissive dopants. Modified from Reference [37].

dopant which would result in some triplets being trapped on the blue dopant and therefore unable to reach the phosphorescent dopants.[38] Consequently, these workers used a fluorescent blue emitter with a smaller singlet-triplet energy gap to ensure that all the triplets generated in the recombination zone were able to migrate to the phosphors (Fig. 3). The dopant, 4P-NPD, **16**, also has a high quantum yield of 0.94 and has energy levels such that any triplets generated on it can be passed to the host material. A series of devices adjusting the location and concentration of the red **17** and green Ir(ppy)$_3$ dopants gave devices exhibiting color coordinates of (0.43, 0.43) (i.e., "warm white") at 1000 cd m^{-2} with a CRI of 82 and efficiencies of 25.4 lm W^{-1} and EQE 11.2% without enhanced outcoupling. A redder device with coordinates of (0.49, 0.42) with a CRI of 62 gave efficiencies of 31.6 lm W^{-1} and EQE 15.2%.



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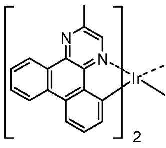

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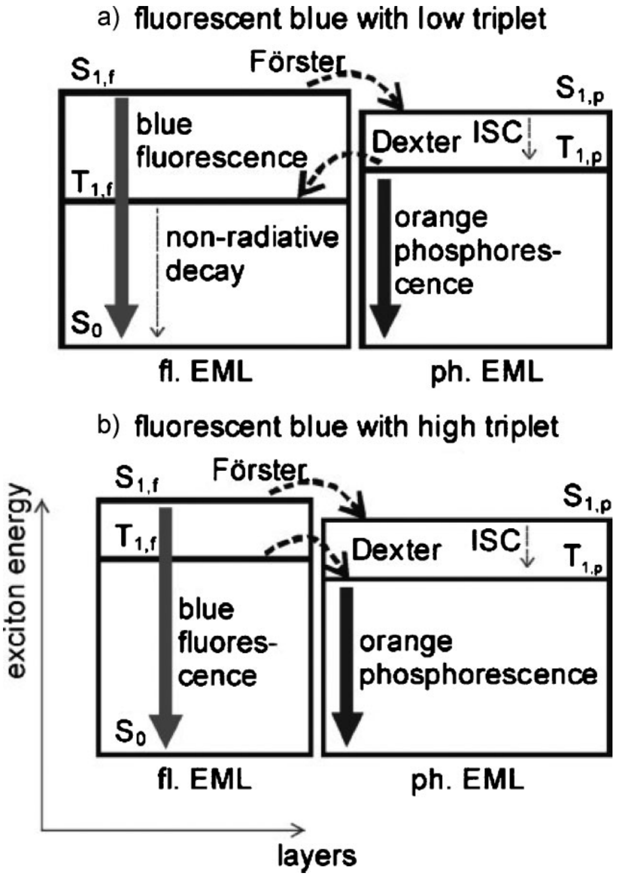


Figure 3. Exciton energy diagram for a blue fluorophor and an orange phosphor. Reproduced with permission from Reference [38].

Leo and co-workers combined the fluorescent blue arylamine 4P-NPD, **16** with green and orange iridium phosphors in a p-i-n structure to obtain outcoupled devices reaching a power efficiency of 32 lm W^{-1} and EQE 14% for CIE of (0.43, 0.43) and CRI of 82.[39] A new strategy involved directly doping the orange phosphor (0.2 wt %) into the fluorescent blue layer to facilitate the rapid transfer of triplets. This layer simultaneously emits fluorescence and phosphorescence. A reduction in current/efficiency roll-off was observed compared to devices with separate emission layers.

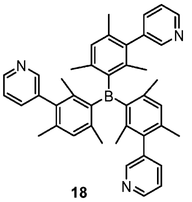
A recent modification combines a blue phosphor (FIrpic) with the emission layer placed further away from the reflective cathode and improved outcoupling. An impressive power efficiency of 90 lm W^{-1} was achieved with color coordinates (0.44, 0.46) and CRI of 80 at 1000 cd m^{-2}. The short operating lifetimes were explained by degradation of the FIrpic emission.[40]

Chemical doping of the transport layers to achieve a much simpler device stack has been reported.[41] By co-evaporating the strong electron acceptor tetrafluoro-tetracyano-*p*-quinodimethane (F$_4$TCNQ) with the hole transport layer and, in later work, Cs with the electron transporting layer,[42] the barrier to charge injection from the electrodes was significantly reduced resulting in OLEDs that achieve high brightness at very low driving voltages and thus extremely long projected lifetimes. This approach mirrors the p-i-n structure seen in inorganic systems comprising a hole conductor-insulator-electron conductor device architecture. The doping of the HT and ET layers increases injection and charge conduction into the organic layers from the inorganic electrodes, greatly lowering the required drive voltage and increasing the current flowing through the device leading to more efficient light generation.

Novaled AG has subsequently developed less toxic dopants and demonstrated full RGBW devices using the p-i-n technology. Most of the recent literature on high-performance WOLEDs adopts the p-i-n structure with the common configuration anode/HIL/HTL(doped) /EBL/EML/HBL/ETL(doped)/cathode, with the blocking layers (BLs) ensuring that the excitons formed in the emissive layer cannot defuse away from this layer. To date, using this technology in conjunction with Philips, emitters at 50.4 lm W^{-1} have been reported, with color coordinates of (0.43, 0.44) via the OLLA project.[43]

Ma and co-workers co-doped blue and orange Ir complexes into a host [viz. 4,4′,4″-tris(*N*-carbazolyl)triphenylamine (TCTA)] that has a higher triplet energy than the complexes to achieve increased triplet confinement on the complexes.[44] WOLEDs with coordinates of (0.38, 0.43), CRI 65 and limited current/efficiency roll-off were demonstrated. The maximum current and power efficiencies reached 37.3 cd A^{-1} and 33.0 lm W^{-1}, with efficiency and EQE changing from 36.7 cd A^{-1} and 13.3% at ca. 100 cd m^{-2}, to 31.0 cd A^{-1} and 11.3% at 1000 cd m^{-2}.

Eom et al. recently reported a p-i-n structure comprising two adjacent emissive layers each co-doped with three phosphorescent iridium emitters (red, green, and blue). Efficiency improvements were achieved by using the high-triplet level transport materials 1,1-bis(di-4-tolylaminophenyl)cyclohexane (TAPC) as the HTL and tris[3-(3-pyridyl)mesityl]borane (3TPYMB) **18** as the ETL which led to enhanced charge and exciton confinement in the emissive layers. The R, G, and B emitters were PQIr (an analog of **14**), $Ir(ppy)_3$ and FIr6 **19**, respectively.[45] The devices showed low turn on voltages (ca. 3 V), a peak efficiency of 40 lm W^{-1} at 100 cd m^{-2} which rolled off to 26 lm W^{-1} at 1000 cd m^{-2}. The color coordinates were (0.39, 0.41) at 10 cd m^{-2} and (0.35, 0.40) at 1000 cd m^{-2} with a CRI of 79 at both luminances. The maximum EQE was 19%.



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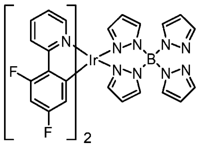

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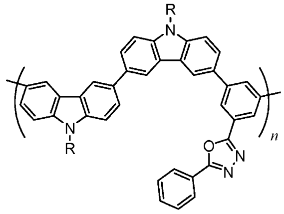

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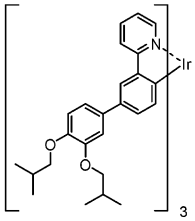

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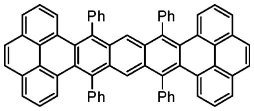
5. Polymer Blends

In polymer blend systems, one or more polymers are blended with other optional dopants and solution-deposited onto device substrates. Whilst this approach has the benefit of simplifying the synthesis to arrive at the multiple components, problems can arise if crystallization and film morphology changes occur during device operation.

An example of stable white EL with CIE coordinates that are independent of current density over the range 10–120 mA cm^{-2} is provided by the combination of the fluorescent heptacene derivative **22** (ca. 1 wt %) doped into a polyfluorene host, in a spin-coated, two-component, single-(emissive)layer structure.[46] The device exploits a combination of energy transfer from blue host to dopant **22** plus host emission: the maximum luminance was ca. 20 000 cd m^{-2}, with maximum luminous efficiency of 3.55 cd A^{-1} at 4228 cd m^{-2}, and maximum power efficiency of 1.6 lm W^{-1} at 310 cd m^{-2}.



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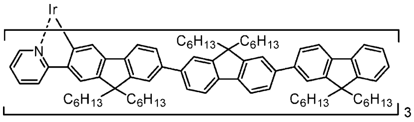
Huang et al. spin-coated a blend of MEH-PPV (0.25–2.0 wt %) and PFO onto the PEDOT:PSS layer and capped it with a layer of caesium carbonate spin-coated from a water solution to give a device that emitted from both polymers to give white light with an efficiency of 12.6 lm W^{-1}.[47] Without the Cs_2CO_3 layer the efficiency dropped from >10 cd A^{-1} to ca. 3 cd A^{-1}.

An attractive feature of cyclometalated iridium complexes is that the emission color can be finely tuned over a large range of wavelengths by varying the substituents on the ligands. Our group in Durham has carefully balanced the amount of the yellow-orange emitter $Ir(PyFl_3)_3$ **23** doped into the polyfluorene host (PF2/6-am4) **24** yielding white light with color coordinates of (0.35, 0.37) (Fig. 4).[48] A peak EQE of 2.8% and luminance of 16 000 cd m^{-2} at applied voltage of 5 V (i.e., 4.57 cd A^{-1}) was achieved.



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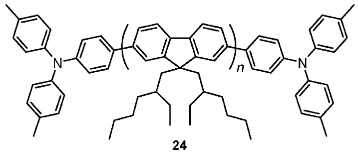
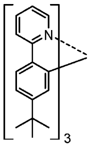


24

The group of Cao doped the green and red-emitting complexes $Ir(Bu\text{-}ppy)_3$ **25** and $(Piq)_2Ir(acaF)$ **26**, respectively, into films of silsequioxane-terminated PFO (PFO-poss) in devices with the structure ITO/PEDOT:PSS/PVK/blend/Ba.[49] Doping 0.14% of both **25** and **26** resulted in white light with a peak efficiency of 5.5 lm W^{-1}/9 cd A^{-1} and CIE coordinates of (0.33, 0.33). The WOLEDs retained high efficiency of >7 cd A^{-1} at a brightness of 1000 cd m^{-2}.



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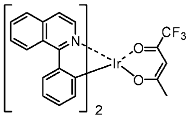

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A combination of phosphorescent R, G, and B iridium complexes were doped into a high-bandgap PVK host.[50] The electron-transporting OXD-7 was also added to the mixture. By varying the dopant concentrations emissions with color temperatures from 2000 to 9000 K were achieved. Efficiencies peaked with an RGB ratio of 1:1:50 at 10 lm W^{-1}/ EQE 13.8% with CIE coordinates of (0.301, 0.467) and for a ratio of 1:1:20 at 9.5 lm W^{-1}/14% with CIE coordinates of (0.343, 0.468). All the devices exhibited a low turn-on voltage of <4 V and low color-drift with varying bias with Δ CIE_x 0.02 and Δ CIE_y 0.01, between 1.2 and 12 mA cm^{-2}.

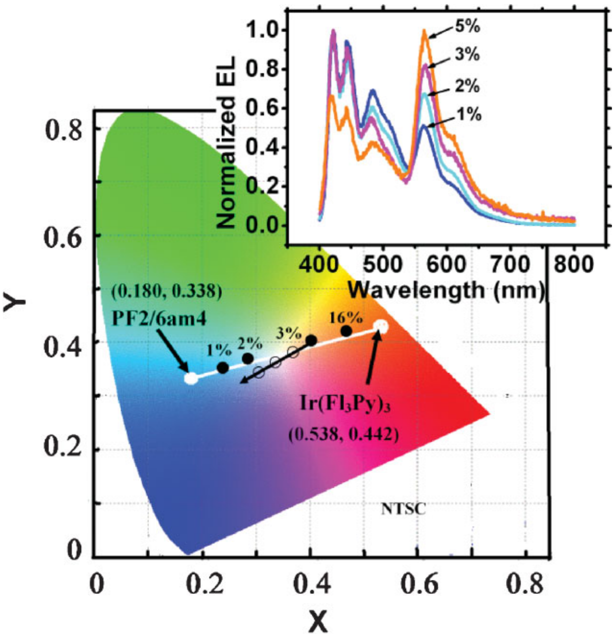
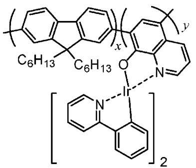


Figure 4. CIE chromaticity diagram for different dopant concentrations of **23** blended into host **24**. Inset: The normalized EL spectra for different dopant concentrations. All data obtained at a bias of 5 V. As the bias voltage increases the overall emission increases and more excitation to the higher energy fluorescent states takes place and the CIE coordinates shift to the blue as indicated by the central black arrow. Device architecture: ITO/PEDOT:PSS/**23–24** blend/Ca/Al. Reproduced with permission from Reference [48]. Copyright 2005, American Institute of Physics.

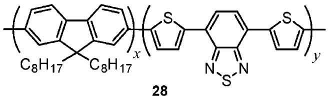
Slugovc and co-workers used a blend of polyfluorene and polyfluorene derivative **27** with an iridium complex directly conjugated into the backbone. Although white light with coordinates of (0.30, 0.35) was obtained, the device gave very poor efficiencies.[51] The reason for this is the long range interaction of the spin orbit coupling from the heavy atom, mediated via the π conjugation, known as the remote intermolecular heavy atom effect (RHAE).[52]



27

6. Single Polymer Devices

Single (co)polymers for WOLEDs offer the advantage of avoiding phase behavior in blended layers; however, they are synthetically more challenging. Research at Dow in the 1990s established that the covalent incorporation of a smaller band gap "dopant" into the polyfluorene backbone results in exciton diffusion to, and emission from, the dopant rather than the fluorene moieties.[53] Cao and co-workers showed that incorporation of as little as 0.5 mol % of the red dopant 4,7-bis(thienyl)-2,1,3-benzothiadiazole (BTB) conjugated into a polyfluorene backbone, **28**, results in purely red emission.[54,55] By reducing the feed ratio of BTB to ca. 0.01 mol % the red emitters could be saturated leading to incomplete energy transfer and emission from both the blue "host" and red "guest". This approach was extended to random copolymer **29** incorporating green benzothiadiazole (BT) and red (BTB) units in precisely controlled ratios. Simultaneous emission from the R, G, and B units gave three-color white light. The CIE coordinates showed good stability at driving voltages between 9 (0.34, 0.32) and 16 V (0.31, 0.31) with brightnesses of 416 and 6994 cd m^{-2}, respectively (Fig. 5). EQE of 3.8% and luminance efficiency of 6.2 cd A^{-1} were obtained after annealing.[56]



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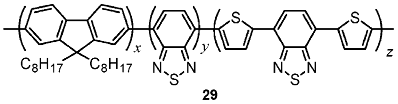
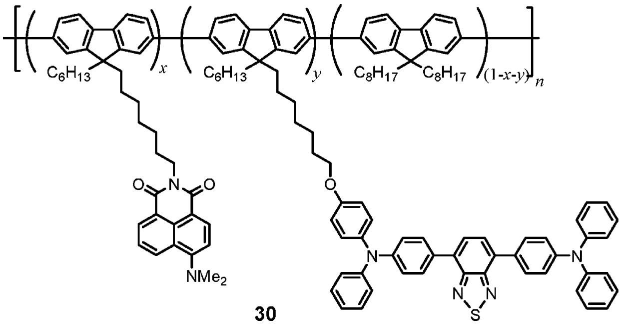


29

The group of Shu studied a copolymer similar to **29** that also incorporated charge-transporting diaryloxadiazole and triarylamine units as pendant substituents on some of the fluorene monomers.[57] ITO/PEDOT:PSS/copolymer/TBPI/Mg:Al devices gave emission with coordinates of (0.37, 0.36) and an efficiency of ca. 4 cd A^{-1}. This approach exploited similar building blocks to those used previously to attain efficient blue light-emitting materials.[58–60]

Liu et al. attached lower-energy chromophores as non-conjugated pendant groups on polyfluorene derivative **30**. The DMAN and bis(triphenylamine)BT units give cyan (ca. 470 nm) and orange emission, respectively, via energy transfer from the blue fluorene backbone. Simple, single-layer devices gave efficiencies of 12.8 cd A^{-1} and power efficiencies of 8.5 lm W^{-1} from a two-color white with coordinates of (0.31, 0.36).[61]



30

We have developed a new strategy for white emission from a single polymer by exploiting dual fluorescence from random (9,9-dioctylfluorene) (dibenzothiophene-*S*,*S*-dioxide) (pF-S)copolymers. For **31** emission arises from both locally excited (LE) and intramolecular charge transfer (ICT) states; the latter is broad and red-shifted.[62] As the incorporation of the S unit increases from 2–30 mol %, the EL color changes from a PFO-like blue through to greenish-white (0.24, 0.41) with increased efficiencies and brightness of 3700 cd m^{-2} at 10 V for **31**.[63] Excellent stability in EL efficiency is maintained for **31** over a wide range of current

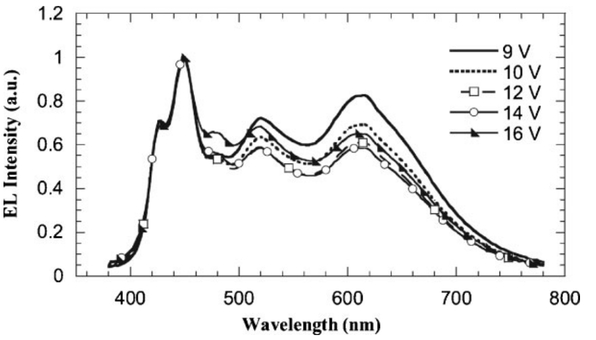
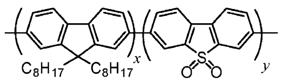


Figure 5. EL spectra of devices made from copolymer **29** at different voltages. Device architecture: ITO/PEDOT:PPS/PVK/**29**/Ba/Al. Reproduced with permission from Reference [56].

densities (2–200 mA cm^{-2}). An extension of this work to copolymer **32** has shown that the dual emission process is unaffected by the additional red chromophore ($z \leq 0.1$ mol %) which provides enhanced spectral coverage (see Table of contents graphic) with EL tuned to (0.35, 0.29) which is closer to the desired pure white region.[64] Utilizing dual emission could reduce problems associated with Förster energy transfer from high-energy to low-energy excited states which requires that the low energy emitters are saturated before the higher energy blue emission can contribute.



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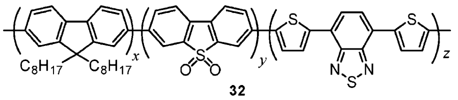
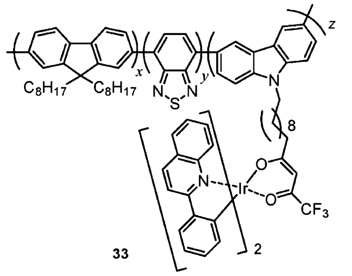


32

Several groups, e.g., Chen,[65] Cao,[66] and Holmes,[67,68] have used polyfluorene-based backbones with a pendant triplet iridium emitter. As with the fluorescent copolymers, high-energy emission from the backbones was completely quenched with as little as 0.5% of the iridium complex,[66] again due to the RHAE. At present, these types of systems are limited to red-orange triplet emitters as the majority of polymer backbones do not have a sufficiently high triplet energy (see Section 4) to prevent quenching of green-yellow iridium emitters. For example, polyfluorenes have a triplet energy of ca. 2.2 eV whilst green triplet emitters, such as those based on $Ir(ppy)_3$, have triplet energies of ca. 2.4 eV.

Cao and co-workers combined a red-emitting iridium unit as a pendant group on carbazole with green-emitting benzothiadiazole groups and by balancing the incorporation rates white light was achieved by simultaneous fluorescence and phosphorescence from polymer **33**.[69,70] Devices with architectures ITO/PEDOT:PSS/PVK/**33**/CsF:Al showed CIE coordinates of (0.32, 0.44) and efficiencies of 6.1 cd A^{-1}, or (0.44, 0.38) at 5.6 cd A^{-1}.

For copolymer **34**, fluorenone is the green emitter and the red iridium complex is incorporated via the acac unit into the polymer backbone.[71] ITO/PEDOT:PSS/**34**/CsF:Al devices were tuned to



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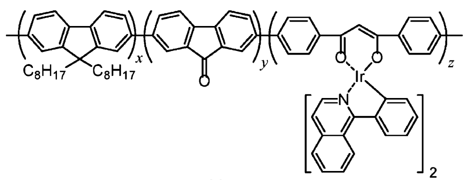


34

emit white light with coordinates of (0.32, 0.45) and 5.5 cd A^{-1} efficiency, or (0.28, 0.32), 3.25 cd A^{-1} and a maximum luminance of 1015 cd m^{-2}. However, it should be noted that fluorenone units have a long fluorescence lifetime (6 μs) and concomitant low quantum yield so are poor emitters. They also generate charged species from excitons and have a high intersystem crossing yield leading to greater triplet production.[72] The lifetimes of these devices were not stated and it should be noted that Holmes and co-workers reported that the use of acac in an iridium complex can result in poor device lifetimes as PSS (from the PEDOT layer) can lead to dissociation of the iridium center from the acac ligand.[68]

Several companies are investigating white light-emitting polymers. Devices using Sumation's range of polymers, developed by Cambridge Display Technology, give efficiencies of 13–14 lm W^{-1} at 1000 cd m^{-2} at coordinates of (0.4, 0.4) and a lifetime of 6000 h from an initial brightness of 1000 cd m^{-2}. The device configuration is ITO/HIL/IL/polymer/cathode (IL = interlayer).[73]

7. Stacked Devices

As with multilayer devices, stacked devices are assembled by vacuum sublimation. The devices comprise two or more complete stacks layered on top of each other with charge generation layers (CGLs) between them. Stacked devices can achieve white emission either through multiple white layers to improve device lifetime, or by individual color stacks which can be independently driven from different voltage sources.[74] The color mix can thereby be tuned and this architecture can reduce problems arising from differential rates of ageing of the several emitting layers.

Kido et al. have recently demonstrated eight-stack devices that show long device lifetimes brought about by possessing multiple backup stacks in case a stack fails.[75] The recently launched company Lumiotec aims to commercially exploit these systems. The group of Chen demonstrated devices comprising two white stacks separated by a tungsten oxide CGL which delivered an efficiency of 22 cd A^{-1} which was three times that of a single-stack device and gave color coordinates of (0.29, 0.44).[76] Qi et al. demonstrated a stacked device with three layers, each emitting one of the primary colors using Fir6, Ir(ppy)$_3$ and PQIr as blue, green, and red dopants. Using molybdenum trioxide as the CGL in a complex device, a power efficiency of 21 lm W^{-1} and an EQE of 32% were reached with color coordinates of (0.45, 0.36). The efficiency at 1000 cd m^{-2} was 13 lm W^{-1}.[77] Using a stacked WOLED Novaled has obtained 38 lm W^{-1} with coordinates of (0.43, 0.44) and a CRI of 90 at a brightness of 1000 cd m^{-2}. A lifetime of >100 000 h is reported for this device.[78]

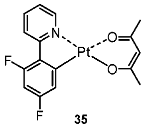
8. External Phosphors

A further method of energy transfer involves the use of external phosphors. A UV or blue emitter is contained in the device and red, green (and blue in the case of a UV emitter) phosphors are added to the outside of the display to convert some of the light allowing any shade of white to be obtained. A further benefit of this approach is that all the colors age at the same rate as they are all derived from the same emitter. A General Electric paper from 2002 describes a blue-emitting polymer supplied by Cambridge Display Technology and red and green external high efficiency (>98%) phosphors leading to a tuned white light with a CRI of 93 and efficiencies of 3.8 lm W^{-1} and 7 cd A^{-1}.[79] However, for marketing purposes, the research was discontinued as the devices have a yellowish appearance which was undesirable compared to the mirror-like off-state effect of standard devices.

9. Excimer/Exciplex Emission

Another approach to broad emission from a single component is through excimer emission. While fluorescent excimers display much lower efficiency than that the corresponding monomer, phosphorescent excimers show good efficiencies. Devices with an emission layer of square planar platinum complexes can stack at high doping levels leading to emission from both the isolated complex molecules and red-shifted emission from the excimer.

A device with 16% of complex **35** in 1,3-bis(*N*-carbazolyl)-benzene (mCP) host **36a** gave white light emission from both the turquoise complex emission and red-orange excimer with a device efficiency of 8.1 lm W^{-1}.[80] Jabbour et al. subsequently used the pyridine analog **36b** as host, obtaining devices with 16% EQE (12 lm W^{-1}, 38 cd A^{-1}) at a rather high drive voltage, i.e., 9.4 V for 500 cd m^{-2}.[81] The color coordinates of (0.46, 0.47) demonstrated poor CRI of 69. The same group subsequently added a second platinum complex **37** in a vacuum-deposited device structure ITO/PEDOT:PSS/TCTA/**36b**:**35**(8%):**37**(2%)/BCP/CsF/Al which showed an EQE of 14.5% (17 lm W^{-1}) at 500 cd m^{-2} with CIE of (0.38, 0.40) and an improved CRI of 81 which was almost independent of drive voltage in the range 2–7 V.[82]



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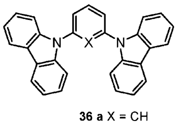
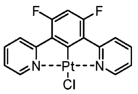


36 a X = CH
b X = N



37

In a separate study, efficient white emission has been achieved from a single (vacuum deposited) emissive layer comprised of an electron donor (D: an arylamine derivative) and a phosphorescent electron acceptor (A: an N^C^N Pt complex, related to **37**) which are able to create exciplexes displaying a broad emission spectrum located between monomer and excimer spectra of the phosphor. Additional ETHB and EB layers were present.[83] Three different emissive states are produced efficiently at the same time: (i) molecular excitons (^{3}A*) producing monomer phosphorescence, (ii) triplet excimers ^{3}(AA)*, and (iii) an exciplex ^{3}(DA)*. A maximum EQE of 6.5% was reported at very low luminance (0.1 cd m^{-2}) changing to 3% at 500 cd m^{-2}. Devices displayed CIE coordinates (0.46, 0.46) and CRI of 88 at 16 V. At this high voltage low device lifetimes would be expected.

10. Future Opportunities and Challenges

While much of the pioneering work into electroluminescent materials focused on the three primary colors, research on white light has grown rapidly in recent years. This review has sought to highlight some of the key advances in this topic. Chemists, physicists, and engineers have developed several different approaches to WOLEDs and in recent months companies such as Philips, Osram, NEC, and Mitsubishi have demonstrated WOLED lighting prototypes. However, whilst the performance of commercial materials is undoubtedly higher than the data reported in the literature, several key problems remain to be overcome. Many of these relate to scaling small prototype laboratory devices to large-scale processing technologies as most high performance materials are based on small-molecule strategies. A drawback is the need to vacuum deposit these materials which leads to both a high wastage rate of material and requires the use of expensive/unavailable machinery. The cost of evaporators is proportional to the size of the substrate cubed. Current emphasis is, therefore, moving toward solution-processable materials with the prospect of generating the economy of scale needed to bring down the price of WOLED lighting to compete in the mainstream markets. This may take the form of inkjet, screen or roll-to-roll printing such as that recently demonstrated by General Electric (Fig. 6).[84] Osram's novel "wave" lighting structure, shown in Figure 7,[85] comprises individual panels produced using solution processing of Sumation polymers. Key challenges with large-area devices include avoiding increased operating voltages, resistive heating, and differential degradation across the active area, while retaining stable high-quality emission with a long lifespan.

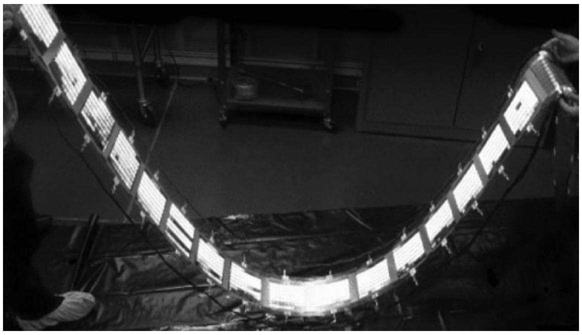


Figure 6. GE demonstration of a roll-to-roll printed OLED. Reproduced with permission from Reference [84]. Copyright 2009, General Electric Company.

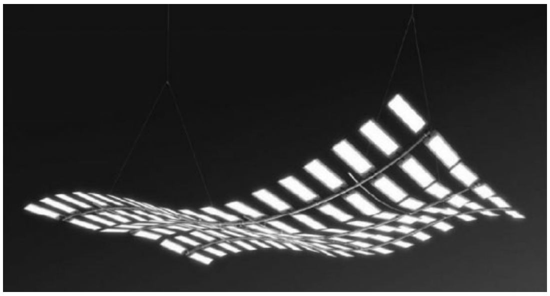


Figure 7. "Wave" light manufactured by Osram using Sumation polymers. Reproduced with peremission from Reference [85]. Copyright 2008, OSRAM GmbH.

On the materials side, there is a need for systems which maintain charge balance at high currents to avoid quenching effects and quantum efficiency roll-off.[86] Higher efficiencies and CRIs may be facilitated by improved outcoupling[87] to reduce "wastage" of light generated, and by moving from two or three color white to four or more color white. Even a small increase in the efficiency of lighting will lead to considerable savings in terms of cost and energy. WOLED research is vibrant and although the commercial introduction of WOLEDs as general lighting sources would seem to be several years away, the ongoing improvements in this technology will continue to inspire scientists and designers to explore the potential of prototype products, initially for the more specialized niche markets.

Glossary

Brightness: The brightness specifies the amount of light emitted per unit area of the device, usually given in candelas per meter squared (cd m^{-2}). A typical laptop display has a brightness of ca. 100 cd m^{-2}.

Candela: The unit of luminous intensity emitted by a light source.

Color/Chromaticity: The color of light is identified by the CIE (Commission Internationale d'Eclairage 1931) colorimetry system. Any color can be described by the chromaticity (x,y) coordinates on the CIE diagram (Fig. 1).

Color Rendering Index (CRI): A numerical measure of how true a color appears to the human eye when illuminated with a light source. The scale is 0–100, where 100 is a true color representation.

Color Temperature (CT): The CT of a light source is determined by comparing its chromaticity with that of an ideal black body radiator. The temperature at which the heated black body radiator matches the color of the light source is the CT of this source. High CTs ($>$ ca. 5000 K) are cool (green–blue); lower CTs ($<$ ca. 5000 K) are warm (yellow–orange). (See Fig. 1).

Lumen: The lumen is the unit of light intensity perceived by the human eye, taking into account the varying sensitivity of the human eye as a function of wavelength.

OLED: Organic light emitting diode (or device).

Outcoupling: The process by which photons are extracted from the OLED. A large percentage of light is lost by internal absorption and internal reflections due to the high refractive indices of the materials in the device. Enhanced outcoupling efficiency, which increases the EQE of the device, has been achieved by embedding a grid of low index material in the organic layers to redirect the light toward the substrate normal.

PLED: Polymer light-emitting diode (or device).

Power Efficiency: The power efficiency is the output light power from a device per electrical power input. It is measured as the luminous efficiency in candela per ampere (cd A^{-1}), or the luminous power efficiency (also termed the efficacy) in lumens per watt (lm W^{-1}). The power efficiency of a typical incandescent light bulb is ca. 15 lm W^{-1}.

Quantum Efficiency: EQE: External electroluminescence quantum efficiency (η_{ext}). The ratio of the number of photons emitted by the device into the viewing direction to the number of electrons injected. *IQE*: Internal electroluminescence quantum efficiency (η_{int}). The ratio of the number of photons generated within the device to the number of electrons injected. The IQE is related to the EQE by the outcoupling efficiency. Note that a device with a high efficiency value at a low luminance of only a few cd m^{-2} is not suitable for practical applications, although it may be valuable for the fundamental understanding of the materials and the device characteristics.

Solid-State Lighting (SSL): The type of lighting which uses LEDs, OLEDs, and PLEDs as the source of illumination, rather than electrical filaments, plasma, or gas.

WOLED: An organic light-emitting diode that emits white light.

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[1] C. W. Tang, S. A. VanSlyke, *Appl. Phys. Lett.* **1987**, *51*, 913.
[2] J. M. Burroughes, D. D. C. Bradley, A. R. Brown, A. N. Marks, K. Mackay, R. H. Friend, P. L. Burn, A. B. Holmes, *Nature* **1990**, *347*, 539.
[3] G. Leising, B. Ullrich, G. Grem, G. Leditzky, *Adv. Mater.* **1992**, *4*, 36.
[4] D. Braun, A. J. Heeger, *Appl. Phys. Lett.* **1991**, *58*, 1982.
[5] H. Spreitzer, H. Becker, E. Kluge, W. Kreuder, H. Schenk, R. Demandt, H. Schoo, *Adv. Mater.* **1998**, *10*, 1340.
[6] Q. Pei, Y. Yang, *J. Am. Chem. Soc.* **1996**, *118*, 7416.
[7] *Polyfluorenes*, (Eds: U. Scherf, D. Neher), Springer, Heidelberg **2009**.
[8] J. L. Segura, *Acta Polym.* **1998**, *49*, 319.
[9] A. Kraft, A. C. Grimsdale, A. B. Holmes, *Angew. Chem., Int. Ed.* **1998**, *37*, 402.
[10] U. Mitschke, P. Bäuerle, *J. Mater. Chem.* **2000**, *10*, 1471.
[11] A. C. Grimsdale, K. Leok Chan, R. E. Martin, P. G. Jokisz, A. B. Holmes, *Chem. Rev.* **2009**, *109*, 897.
[12] J. Johnson, *Chem. Eng. News* **2007**, *85*, 46.
[13] A. S. Kydes, in *Handbook of Energy Efficiency and Renewable Energy*, (Eds: F. Kreith, D. Y. Goswani), CRC Press, New York **2007**, Ch. 7.
[14] Report "Basic Research Needs for Solid State Lighting", http://www.er.doe.gov/bes/reports/files/SSL_rpt.pdf (last accessed: May 2009).
[15] C. J. Humphreys, *MRS Bull.* **2008**, *33*, 459.
[16] A. H. Tullo, *Chem. Eng. News* **2008**, *86*, 20.
[17] J. Kido, K. Hongawa, K. Okuyama, K. Nagei, *Appl. Phys. Lett.* **1994**, *64*, 815.
[18] J. Kido, M. Kimura, K. Nagai, *Science* **1995**, *267*, 1332.
[19] R. F. Service, *Science* **2005**, *310*, 1762.
[20] B. W. D'Andrade, S. R. Forrest, *Adv. Mater.* **2004**, *16*, 1585.
[21] I. ul Haq Raja, J. Y. Lee, I. T. Kim, S. H. Lee, *Monatsh. Chem.* **2008**, *139*, 725.
[22] B. P. Lyons, A. P. Monkman, *Phys. Rev. B: Condens. Matter* **2005**, *71*, 235201.
[23] K. Brunner, A. van Dijken, H. Börner, J. J. A. M. Bastiaansen, N. M. M. Kiggen, B. M. W. Langeveld, *J. Am. Chem. Soc.* **2004**, *126*, 6035.
[24] A. van Dijken, J. J. A. M. Bastiaansen, N. M. M. Kiggen, B. M. W. Langeveld, C. Rothe, A. Monkman, I. Bach, P. Stössel, K. Brunner, *J. Am. Chem. Soc.* **2004**, *126*, 7718.
[25] Y.-S. Wu, S.-W. Hwang, H.-H. Chen, M.-T. Lee, W.-J. Shen, C. H. Chen, *Thin Solid Films* **2005**, *488*, 265.
[26] X. Gong, S. Wang, D. Moses, G. C. Bazan, A. J. Heeger, *Adv. Mater.* **2005**, *17*, 2053.
[27] K. Meerholz, C.-D. Müller, O. Nuyken, in: *Organic Light-Emitting Devices, Synthesis, Properties and Applications*, (Eds.: K. Müllen, U. Scherf), Wiley-VCH, Weinheim **2006**, p. 293.

[28] G. Klärner, J. I. Lee, E. Chan, J. P. Chen, A. Nelson, D. Markiewicz, R. Siemens, J. C. Scott, R. D. Miller, *Chem. Mater.* **1999**, *11*, 1800.

[29] J. P. Chen, G. Klaerner, J. I. Lee, D. Markiewicz, V. Y. Lee, R. D. Miller, J. C. Scott, *Synth. Met.* **1999**, *107*, 129.

[30] M. Inbasekaran, W. Yu, WIPO, WO 2005049689, **2005**.

[31] C. D. Müller, A. Falcou, N. Reckefuss, M. Rojahn, V. Wiederhirn, P. Rudati, H. Frohne, O. Nuyken, H. Becker, K. Meerholz, *Nature* **2003**, *421*, 829.

[32] E. Bacher, M. Bayerl, P. Rudati, N. Reckefuss, C. D. Muller, K. Meerholz, O. Nuyken, *Macromolecules* **2005**, *38*, 1640.

[33] S. Jungermann, N. Riegel, D. Muller, K. Meerholz, O. Nuyken, *Macromolecules* **2006**, *39*, 8911.

[34] M. C. Gather, A. Köhnen, A. Falcou, H. Becker, K. Meerholz, *Adv. Funct. Mater.* **2007**, *17*, 191.

[35] M. C. Gather, R. Alle, H. Becker, K. Meerholz, *Adv. Mater.* **2007**, *19*, 4460.

[36] A. Köhnen, N. Riegel, J. H. W. M. Kremer, H. Lademann, D. C. Müller, K. Meerholz, *Adv. Mater.* **2009**, *21*, 879.

[37] Y. Sun, N. C. Giebink, H. Kanno, B. Ma, M. E. Thompson, S. R. Forrest, *Nature* **2006**, *440*, 908.

[38] G. Schwartz, S. Reineke, T. C. Rosenow, K. Walzer, K. Leo, *Adv. Funct. Mater.* **2009**, *19*, 1319.

[39] G. Schwartz, S. Reineke, K. Walzer, K. Leo, *Appl. Phys. Lett.* **2008**, *92*, 053311.

[40] S. Reineke, F. Lidner, G. Schwartz, N. Seidler, K. Walzer, B. Lüssem, K. Leo, *Nature* **2009**, *459*, 234.

[41] J. Blochwitz, M. Pfeiffer, T. Fritz, K. Leo, *Appl. Phys. Lett.* **1998**, *73*, 729.

[42] J. Huang, M. Pfeiffer, A. Werner, J. Blochwitz, K. Leo, S. Liu, *Appl. Phys. Lett.* **2002**, *80*, 139.

[43] OLLA Project final report, http://www.hitech-projects.com/euprojects/olla/external%20deliverables/PUBLIC%20OLLA_FINAL_ACTIVITY_REPORT_V3.0.pdf (last accessed: May 2009).

[44] Q. Wang, J. Ding, D. Ma, Y. Cheng, L. Wang, *Appl. Phys. Lett.* **2009**, *94*, 103503.

[45] S.-H. Eom, Y. Zheng, E. Wrzesniewski, J. I. Lee, N. Chopra, F. So, J. Xue, *Appl. Phys. Lett.* **2009**, *94*, 153303.

[46] Q. Xu, H. M. Duong, F. Wudl, Y. Yang, *Appl. Phys. Lett.* **2004**, *85*, 3357.

[47] J. Huang, G. Li, E. Wu, Q. Xu, Y. Yang, *Adv. Mater.* **2006**, *18*, 114.

[48] H. A. Al Attar, A. P. Monkman, M. Tavasli, S. Bettington, M. R. Bryce, *Appl. Phys. Lett.* **2005**, *86*, 121101.

[49] Y. Xu, J. Peng, J. Jiang, W. Xu, W. Yang, Y. Cao, *Appl. Phys. Lett.* **2005**, *87*, 193502.

[50] H. B. Wu, J. H. Zou, F. Liu, L. Wang, A. Mikhailovsky, G. C. Bazan, W. Yang, Y. Cao, *Adv. Mater.* **2008**, *20*, 696.

[51] S. Kappaun, S. Eder, S. Sax, R. Saf, K. Mereiter, E. J. W. List, C. Slugovc, *J. Mater. Chem.* **2006**, *16*, 4389.

[52] C. Rothe, S. King, A. Monkman, *Nat. Mater.* **2006**, *5*, 463.

[53] M. Inbasekaran, W. Wu, E. P. Woo, M. T. Bernius, WIPO, WO 0046321 (A1), **2000**.

[54] Q. Hou, Y. Xu, W. Yang, M. Yuan, J. Peng, Y. Cao, *J. Mater. Chem.* **2002**, *12*, 2887.

[55] J. Luo, J. Peng, Y. Cao, Q. Hou, *Appl. Phys. Lett.* **2005**, *87*, 261103.

[56] J. Luo, X. Li, Q. Hou, J. B. Peng, W. Wang, Y. Cao, *Adv. Mater.* **2007**, *19*, 1113.

[57] C.-Y. Chuang, P.-I. Shih, C.-H. Chien, F.-I. Wu, C.-F. Shu, *Macromolecules* **2007**, *40*, 247.

[58] F. Wu, S. Reddy, C. Shu, *Chem. Mater.* **2003**, *15*, 269.

[59] C. Shu, R. Dodda, F. Wu, M. S. Liu, A. K. Jen, *Macromolecules* **2003**, *36*, 6698.

[60] F.-I. Wu, P.-I. Shih, C.-F. Shu, Y.-T. Tung, Y. Chi, *Macromolecules* **2005**, *38*, 9028.

[61] J. Liu, S. Y. Shao, L. Chen, Z. Y. Xie, Y. X. Cheng, Y. H. Geng, L. X. Wang, X. B. Jing, F. S. Wang, *Adv. Mater.* **2007**, *19*, 1859.

[62] F. B. Dias, S. King, A. P. Monkman, I. I. Perepichka, M. A. Kryuchkov, I. F. Perepichka, M. R. Bryce, *J. Phys. Chem. B* **2008**, *112*, 6557.

[63] S. M. King, I. I. Perepichka, I. F. Perepichka, F. B. Dias, M. R. Bryce, A. P. Monkman, *Adv. Funct. Mater.* **2009**, *19*, 586.

[64] F. B. Dias, K. T. Kamtekar, T. Cazati, G. Williams, M. R. Bryce, A. P. Monkman, *ChemPhysChem.* **2009**, *10*, 2096.

[65] X. Chen, J.-L. Liao, Y. Liang, M. O. Ahmed, H.-E. Tseng, S.-A. Chen, *J. Am. Chem. Soc.* **2003**, *125*, 636.

[66] J. Jiang, C. Jiang, W. Yang, H. Zhen, F. Huang, Y. Cao, *Macromolecules* **2005**, *38*, 4072.

[67] A. J. Sandee, C. K. Williams, N. R. Evans, J. E. Davies, C. E. Boothby, A. Köhler, R. H. Friend, A. B. Holmes, *J. Am. Chem. Soc.* **2004**, *126*, 7041.

[68] N. R. Evans, L. Sudha Devi, C. S. K. Mak, S. E. Watkins, S. I. Pascu, A. Kohler, R. H. Friend, C. K. Williams, A. B. Holmes, *J. Am. Chem. Soc.* **2006**, *128*, 6647.

[69] J. X. Jiang, Y. H. Xu, W. Yang, R. Guan, Z. Q. Liu, H. Y. Zhen, Y. Cao, *Adv. Mater.* **2006**, *18*, 1769.

[70] Y. Xu, R. Guan, J. Jiang, W. Yang, H. Zhen, J. Peng, Y. Cao, *J. Polym. Sci. Part A: Polym. Chem.* **2008**, *46*, 453.

[71] K. Zhang, Z. Chen, C. Yang, Y. Tao, Y. Zou, J. Qin, Y. Cao, *J. Mater. Chem.* **2008**, *18*, 291.

[72] S. I. Hintschich, C. Rothe, S. Sinha, A. P. Monkman, P. Scandiucci de Freitas, U. Scherf, *J. Chem. Phys.* **2003**, *119*, 12017.

[73] R. Wilson, 'Cambridge Display Technology', Lecture given at the *4th Global Plastic Electronics Conference and Showcase*, Berlin, October **2008**.

[74] G. Parthasarathy, G. Gu, S. R. Forrest, *Adv. Mater.* **1999**, *11*, 907.

[75] J. Kido, Int. Symp. Soc. Information Display, Los Angeles, **2008**, *39*, 931.

[76] C.-C. Chang, J.-F. Chen, S.-W. Hwang, C. H. Chen, *Appl. Phys. Lett.* **2005**, *87*, 253501.

[77] X. Qi, M. Slootsky, S. R. Forrest, *Appl. Phys. Lett.* **2008**, *93*, 193306.

[78] Novaled website, http://www.novaled.com/oledcompetence/advantage_-whiteoleds.html (last accessed: May 2009).

[79] A. R. Duggal, J. J. Shiang, C. M. Heller, D. F. Foust, *Appl. Phys. Lett.* **2002**, *80*, 3470.

[80] B. W. D'Andrade, J. Brooks, V. Adamovich, M. E. Thompson, S. R. Forrest, *Adv. Mater.* **2002**, *14*, 1032.

[81] E. L. Williams, K. Haavisto, J. Li, G. E. Jabbour, *Adv. Mater.* **2007**, *19*, 197.

[82] X. Yang, Z. Wang, S. Madakuni, J. Li, G. E. Jabbour, *Appl. Phys. Lett.* **2008**, *93*, 193305.

[83] J. Kalinowski, M. Cocchi, D. Virgili, V. Fattori, J. A. G. Williams, *Adv. Mater.* **2007**, *19*, 4000.

[84] Global Research Blog website, http://www.grcblog.com/?p = 247 (last accessed: May 2009).

[85] OSRAM homepage, http://www.osram-os.com/appsos/showroom/projekte.php?p_id= 142&lan = eng (last accessed, May 2009).

[86] N. C. Giebink, S. R. Forrest, *Phys. Rev. B: Condens. Matter* **2008**, *77*, 235215.

[87] Y. Sun, S. R. Forrest, *Nat. Photonics* **2008**, *2*, 483.