J. of Korean Oil Chemists' Soc.,
Vol. 25, No. 1. March, 2008. 91~106

Recent Progress in the Development of Small Organic Molecules for White Organic Light Emitting Devices

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(Received November 30, 2007 ; Accepted February 29, 2008)

Abstract : Development of white light emitting materials has been an interesting area for scientists and scientists have developed many organic, polymer and inorganic materials for white electroluminescent devices. Among them, single component small molecules gave best results in terms of efficiency, simplicity of device fabrication, and CIE values. Therefore, this review covers detailed discussion about syntheses of small compounds used in white organic light emitting devices until 2007.

Keywords: *organic molecule, white organic light emitting compounds, WOLEDs, photoluminescence, electroluminescence.*

1. Introduction

Luminescence is the emission of light from an excited molecule. In photoluminescence, excitation occurs via the absorption of light by a molecule, while in electroluminescence electrical energy is transformed into light.

In recent years, white organic light emitting diodes (WOLEDs) have gained increasing research interest and are considered as a new generation of light sources for backlighting of liquid crystal displays and electric lamp[1]. For WOLEDs, efficiencies were significantly improved in red, green and blue emission by utilizing phosphorescent dyes[2-4], and phosphorescent WOLED were fabricated by several groups [5,6], with high efficiencies of 11 lm/W at 1000 cd/m^2.

First infrared LED was developed in 1962, but until 1993 LEDs could only produce red, green and yellow light. After then Nichia Chemical of Japan figured out how to produce blue LEDs[7]. By combining blue LEDs with red and green LEDs or adding a yellow phosphor to blue LEDs, manufacturers were able to create white light, which opened up a number of new applications. However, these LEDs tend to produce white light with a cool, bluish tinge.

Similarly OLEDs are also in the market. Electroluminescence (EL) of organic materials has been known since the 1960s[8,9]. Intense research activity, however, started in the late 1980s after the development of the multilayered cell structure by Tang and Vanslyke[10].

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White light is made up of nearly equal intensities of light from all regions (VIBGYOR) of the visible spectrum. Combining three different wavelengths of light can produce white light, provided they are widespread in the visible region. Any such sets of the three different wavelengths that on additive mixing can produce white light are called primary colors. The most common set of primary color is red, green and blue. Another dimension to this approach is complementary colors where the combination of only two colors can produce white light. The most used of such set is blue and yellow. Here, it is important to mention that organic molecules generally have very broad emission spectra. Therefore, the combination of emission from not exactly complementary colors may also give white light. The good white point in Commission Internationale de l'éclairage (CIE) coordinate system is defined as (0.33, 0.33).

Many researchers have synthesized new organic materials and tried to obtain white emission either from single molecules or by mixing two complementary colors (e.g., red/bluish green, blue/orange, or green/magenta) or the three primary colors (red, green, and blue)[11-17]. This review covers detailed discussion about syntheses of compounds used in white light emitting devices.

2. The Luminescence of WOLEDs

The most successful approaches used to generate white OLEDs described previously involve separating three different emitters (luminescent dopants) into individual layers. Three emissive centers are needed to achieve good color rendering index (CRI) values[17], as the lines are typically not broad enough to cover the entire visible spectrum with fewer than three emitters. One approach to the design of WOLEDs involves segregating the individual dopants into separate layers. The emissive zone in such a device is thus composed of distinct emissive layers. The design of such a device can be complicated, since careful control of the thickness and composition of each layer is critical for achieving good color balance. The separation of emitters into individual layers is essential to prevent energy transfer between the red, green and blue emitters. The problem is that the highest energy emitter (blue) will efficiently transfer its exciton to the green and red emitters. The efficiency of this energy transfer process is described by the Förster energy transfer equations[18,19]. If the blue emitter has good spectral overlap with the absorption spectra of the green or red emitters, and the oscillator strengths are high for all of the transitions, the energy transfer process will be efficient. Likewise the green emitter will readily transfer its exciton to the red emitter. The end result is that the red emitter dominates the spectral composition if the three are doped into the film at equal concentrations. With fluorescent dyes, the exciton migration lengths are comparatively short and the balance between the three emission colors can be controlled by varying the dopant ratios (more blue is needed than green and more green than red to achieve equal intensity at all three colors). If the dopant concentration is kept low the average distance between dopants can be kept below the Förster energy transfer distance and the affects of energy transfer can be minimized. Having all three dyes within a single layer involves a four component film, with each dopant present at <1%. The preparation of such a film is difficult to carry-out reliably. Any shift in dopant ratio will severely affect the color quality of the device.

3. Meaning of CIE Chromaticity and CRI

The quality of white illumination sources can be fully described by a simple set of parameters. The color of the light source is given by its CIE chromaticity coordinates x and y [20]. The CIE coordinates are typically represented on a two dimensional plot. Monochromatic colors fall on the perimeter of the horseshoe shaped curve starting with blue in the lower left, running through the colors of the spectrum in a clockwise direction to red in the lower right. The CIE coordinates of a light source of given energy and spectral shape will fall within the area of the curve. Summing light at all wavelengths uniformly gives the white or neutral point, found at the center of the diagram (CIE x,y-coordinates, 0.33, 0.33). Mixing light from two or more sources gives light whose color is represented by the intensity weighted average of the CIE coordinates of the independent sources. Thus, mixing light from two or more sources can be used to generate white light. While the two component and three component white light sources will appear identical to an observer (CIE x,y-coordinates, 0.32, 0.32), they will not be equivalent illumination sources. When considering the use of these white light sources for illumination, the CIE color rendering index (CRI) needs to be considered in addition to the CIE coordinates of the source. The CRI gives an indication of how well the light source will render colors of objects it illuminates. A perfect match of a given source to the standard illuminant gives a CRI of 100. Though a CRI value of at least 70 may be acceptable for certain applications, a preferred white light source will have a CRI of about 80 or higher[17].

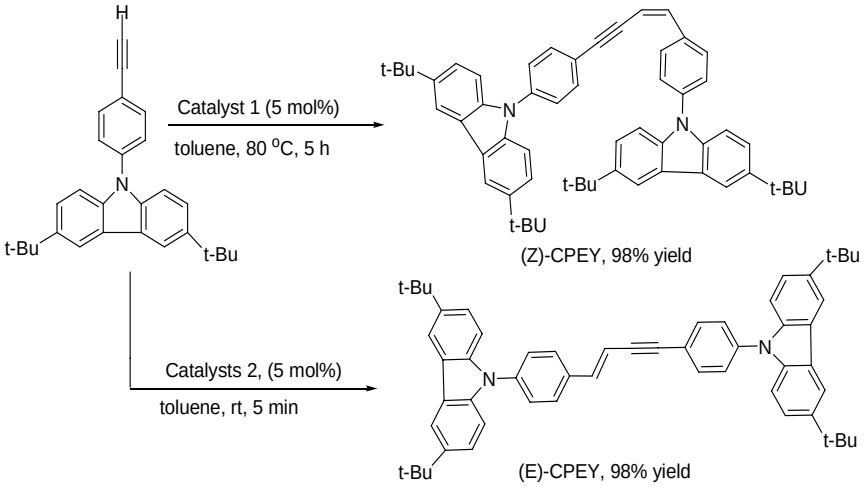
4. Small Organic Molecules for White Light Luminescence

White organic light-emitting devices (WOLEDs) in which emitting layer is pure small organic molecules or small molecules doped with some other molecules have gained much interest for their use in such applications as flat panel displays and lighting sources. To date, WOLEDs with various configurations combined with suitable materials have been reported to generate the desired white light either by using single undoped molecule or by doping one or more emissive layer with fluorescent or phosphorescent materials[21-23]. Undoped single molecules emitting white light have rarely been reported in literature.

4.1. Single Component Emitting Compounds

Most of the WOLEDs reported so far have relied on the use of a combination of several organic components that emit different colors of light to fully span the entire visible spectrum. Compared to the multi-emitting-component WOLEDs, a single-emitting-component WOLED could show many advantages, such as better stability, better reproducibility, and a simpler fabrication process. However, few materials are known to show white-light emission as a single-emitting component. Therefore, the research for new organic light-emitting materials with new structures to be used in single-emitting-component WOLEDs is of obvious interest and importance.

Hou *et al.* reported the synthesis and photo- and electroluminescence properties of carbazole-substituted aromatic (E)- and (Z)-enynes[24]. These new π-conjugated compounds, in particular, the (E)-enyne



Catalyst 1 : Me_2 $Si(C_5Me_4)(N(C_6H_3Me_2$-2,6))$Lu(CH_2SiMe_3)(THF)$,
Catalyst 2 : $(C_5Me_5)_2LaCH(SiMe_3)_2$

Scheme 1. Ragio- and stereoselective synthesis of conjugated enynes.

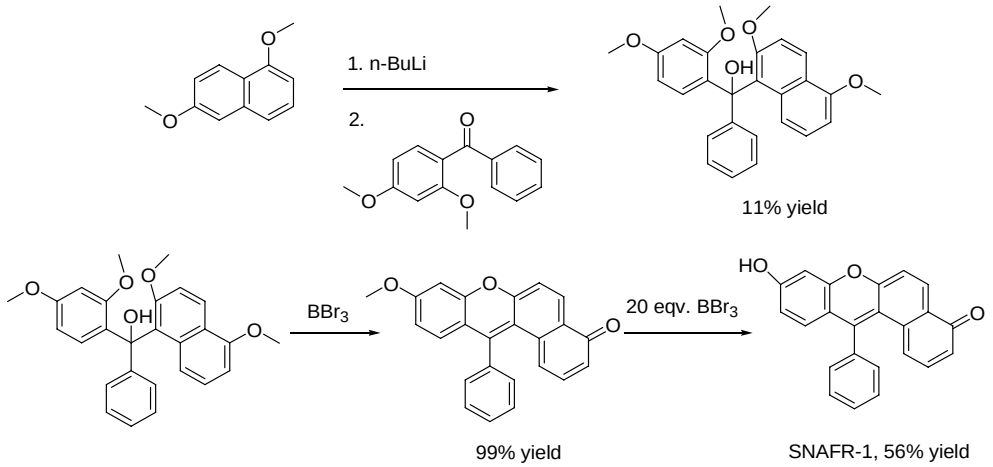
isomer, can serve as an excellent single-emitting component for WOLEDs, emitting almost "pure" white light with stable CIE (Commission Internationale de l'éclairage) coordinates under different driving voltages. Conjugated arynes were synthesized according to following Scheme 1.

These conjugated arynes can act as an excellent single-emitting component for WOLEDs as a result of combination of the blue emission from an isolated molecule with the longer wavelength emissions (green and orange-red) from excimers. The (E)-CPEY-based double-layer device emitted almost pure white light with CIE coordinates of (0.32, 0.33), maximum brightness of 1395 cd/m^2, and maximum current efficiency of 2.07 cd/A. To the date, this is the purest white emission with high luminescence and high efficiency ever reported for a single-emitting component WOLED.

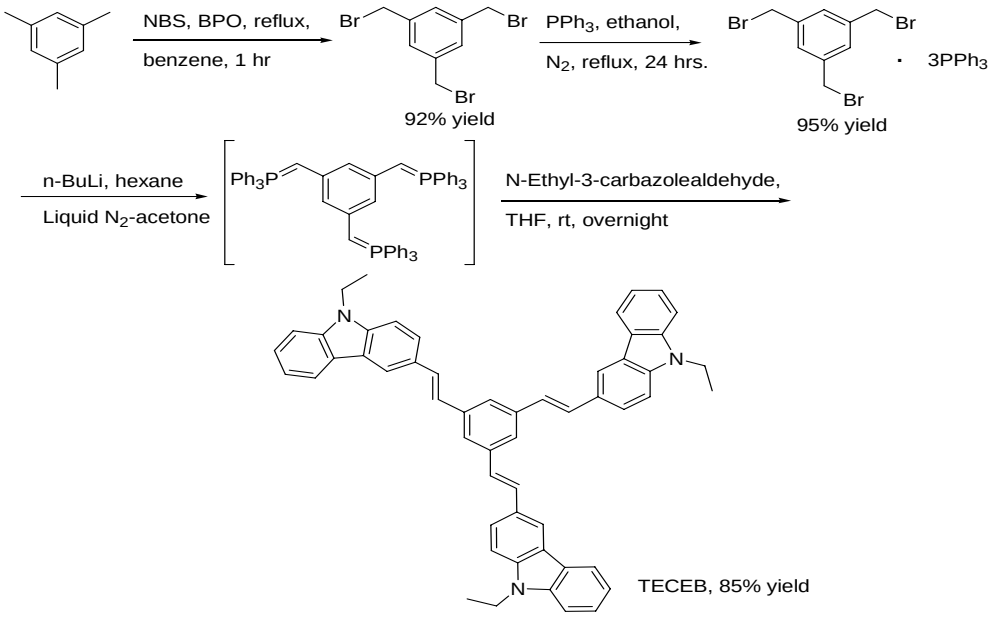
Similarly Strongin *et al*. synthesized a unique dye benzo-[a]xanthene, seminaphtho[a] fluorone (SNAFR-1), and studied it in variety of media[25]. SNAFR-1 was synthesized as shown in Scheme 2.

The optimization of solution parameters and excitation wavelengths allows SNAFR-1 to produce white light. It has been shown that excitation beyond 340 nm dramatically decreases the relative intensity of the blue emission band as excitation of the naphthyl unit becomes less efficient. When excited at 350 nm, the ratio of the two emission bands is nearly equal. The mixing of these emission bands again results in near white emission. Although two band white light has a poorer color rendering index (CRI) owing to the unbalanced red color, this result is of interest since currently two band white light is the main origin of white emission in white polymer light emitting diodes (WPLEDs).

Lee *et al*. first time reported the fabrication of WOLED using single component emitting material 1,3,5-tris(2-(9-ethylcarbazyl-3-ethylene)benzene (TECEB)[26]. Synthetic procedure and chemical structure of TECEB is below (Scheme 3).



Scheme 2. Synthesis of SNARF-1.



Scheme 3. Synthesis of TECEB

Using TECEB, single layer and three layer WOLED were prepared. It is observed that TECEB is capable of emitting white light in single emitting component EL devices whose emitting color is independent of driving voltage. The three layer devices fabricated from this component exhibited bright and white light with a maximum luminescence and current efficiency of 1200 cd/m^2 and 1.1 cd/A, respectively.

Similarly Mazzeo *et al.* synthesized and demonstrated the use of 2,6-bis (dimesitylboryl)-3,5-dimethyl-dithieno[3,2-b:2′, 3′-d]thiophene as single active molecular materials in WOLED[27]. Structure of the compound is below.

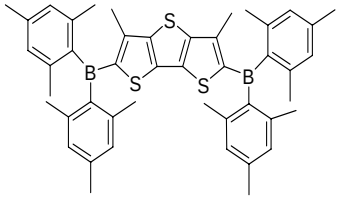


Fig. 1. 2,6-bis(dimethylboryl)-3,5-dimethyl-dithieno[3,2-b:2′.3′-d]thiophene.

In the above compound, white emission was achieved by superposition of the intrinsic blue-green light emission (BGE) of single molecule with red-shifted emission (RSE) that occurs only in the solid state. The origin of RSE peak is due to the formation of cross-like dimer between the molecules. The WOLED prepared by using this material shows bright luminance, up to 3800 cd/m^2 and a quantum efficiency of 0.35 % in air.

4.2. Host-Guest System

To obtain white emission, often a higher energy-emitting host (donor) material is doped with lower energy emitting guest (dye, dopant or acceptor) materials to cause energy transfer from the host to the guests. The dopant site can be excited directly by capturing the charge carriers or by energy transfer from the host to guest, as a result light emission can come from the host and guests, the combined effect of which produces white light. The simplest device structure with a single emitting layer is obtained by doping primary[28-30] or complementary[31-33] color emitting dyes in small molecule host. In these devices, the concentration of the dopants was so maintained that emission from the host was small or negligible. But, an alternative to these systems is incomplete energy transfer from the host to the guest and then combining the emission of both host and guest. As an example, blue and red/orange color emitting dyes are co-deposited to form the emission layer[34-37]. It is not necessary to use dyes to take advantage of the energy transfer; blends of two polymers can also be used as host guest systems[38]. The guest molecules can be fluorescent or phosphorescent in nature. However, phosphorescent dyes based on Ir and Pt complexes have provided significantly higher efficiency of OLEDs because of their ability to emit from both singlet and triplet excitons of the host molecule, whereas a florescent dye can only utilize the singlet exciton. The devices based on phosphorescent dyes are named as electrophosphorescent devices. Important aspect of host guest systems is the choice of host and guest materials for both single and multidoped systems. The energy transfer from host to guest can be either Förster[18] or Dexter type[19]. The primary conditions for such energy transfers are overlap of the emission spectrum of the host and absorption spectrum of the guest (Fig. 2a). Therefore, the host material is always one with emission at higher energies, generally a blue-emitting material. However, there are some basic differences between the two mechanisms. Förster energy transfer results from dipole-dipole interaction between the host and guest while Dexter energy

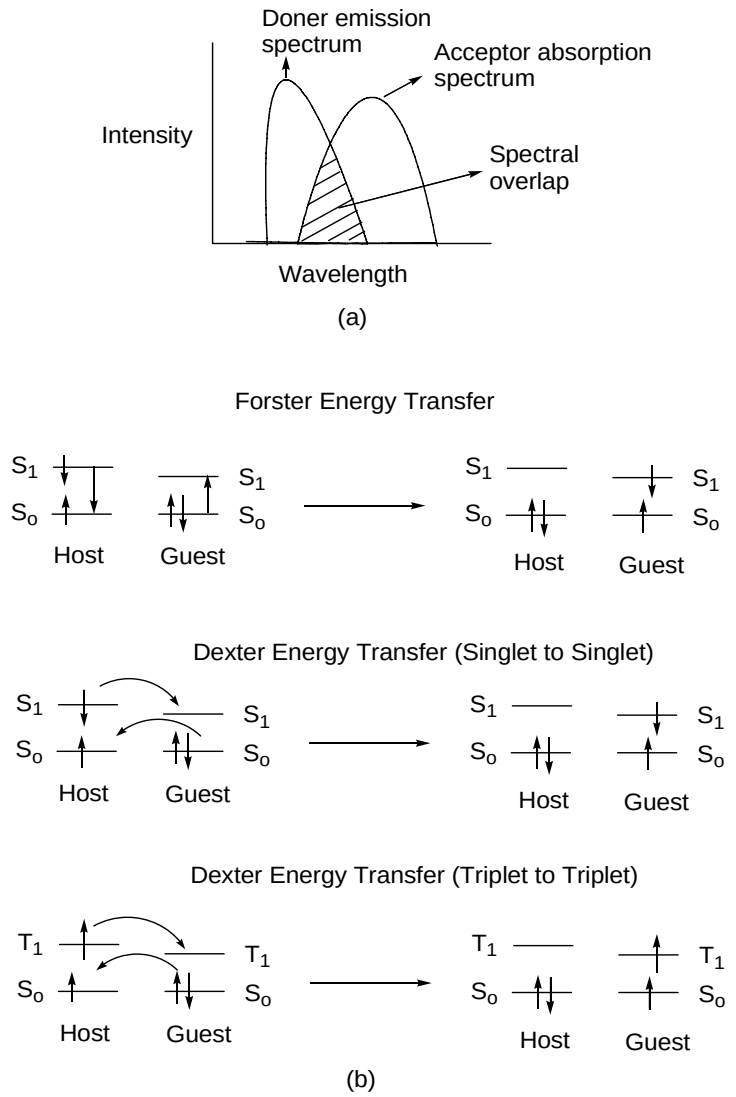


Fig. 2. (a) Spectral overlap of the emission spectrum of the donor and absorption spectrum of the acceptor. (b) Schematic diagram of the Forster and Dexter mechanism.

transfer involves electron exchange between them. Additionally, Förster energy transfer is operative for host guest separation of up to 40100 Å, whereas Dexter transfer requires host guest separation of about 10 Å. Both the processes are schematically shown in Fig. 2b. For multi-doped systems energy transfer between guest molecules can also take place.

<Single-doped materials>

The use of only a single dopant in WOLEDs significantly simplifies the fabrication of WOLEDs relative to other approaches to achieve white organic electroluminescence. It may also solve the problems associated with differential dopant aging. The lifetimes of monochromatic

OLEDs, prepared with different dopants, vary over a wide range, due to different chemical and electrochemical stabilities of the various dopants and host materials that are used. While there have been no reports of the lifetimes or color stabilities of WOLEDs, it is expected that WOLEDs utilizing multiple dopants will show different characteristic aging times for each of the dopants. Differential aging of the dopants would change the color of the WOLED over time, as the dopant emission ratio changes. It is expected that a single dopant WOLED will not suffer from these limitations, since the two emission bands (monomer and aggregate) come from the same dopant.

Forrest and Thompson *et al.* reported efficient white electrophosphorescence with a single emissive dopant[39]. The dopant in these white organic light emitting diodes (WOLEDs) emits simultaneously from monomer and aggregate states, leading to a broad spectrum and high quality white emission. The dopant molecules are based on a series of [2-(4′,6′-difluorophenyl)-pyridinato-N,$C^{2'}$] platinum (II) (Compound a-d in Figure 3).

These complexes were prepared according to the procedure described elsewhere[40]. The dopant complexes described herein have identical photophysics in dilute solution with structured blue monomer emission (λ_{max} = 468, 500, 540 nm). A broad orange aggregate emission (λ_{max} = 580 nm) is also observed when doped into OLED host materials. The intensity of the orange band increases relative to the blue monomer emission as the doping level was increased. The ratio of monomer to aggregate emission can be controlled by the doping concentration, the degree of steric bulk on the dopant and by the choice of the host material. A doping concentration for which the monomer and excimer bands are approximately equal gives an emission spectrum closest to standard white illumination sources. WOLEDs have been fabricated with doped CBP (N,N′-dicarbazolyl-4,4′-bipheny) and mCP (N,N′-dicarbazolyl-3,5-benzene) luminescent layer. The synthesis of mCP was based on a known literature procedure using palladium-catalyzed cross coupling of aryl halides and arylamines[41].

An analysis of color mixing in the Commission Internationale l'éclairage (CIE)

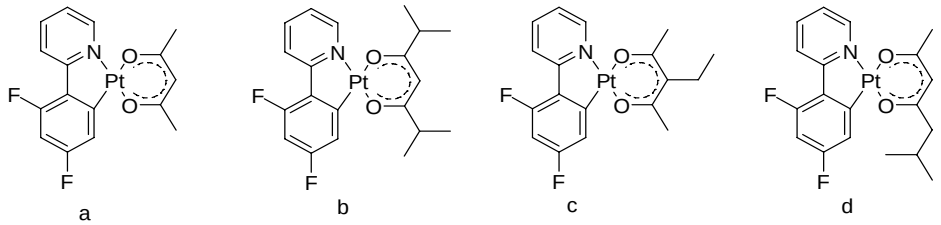


Fig. 3. The structure of platinium dopant.

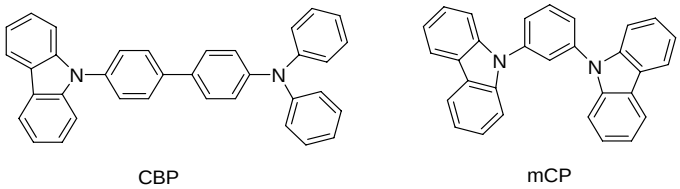
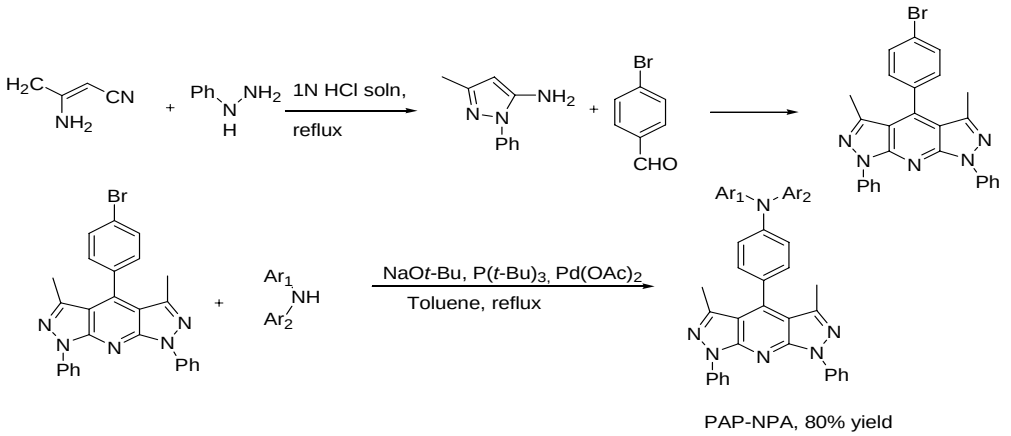


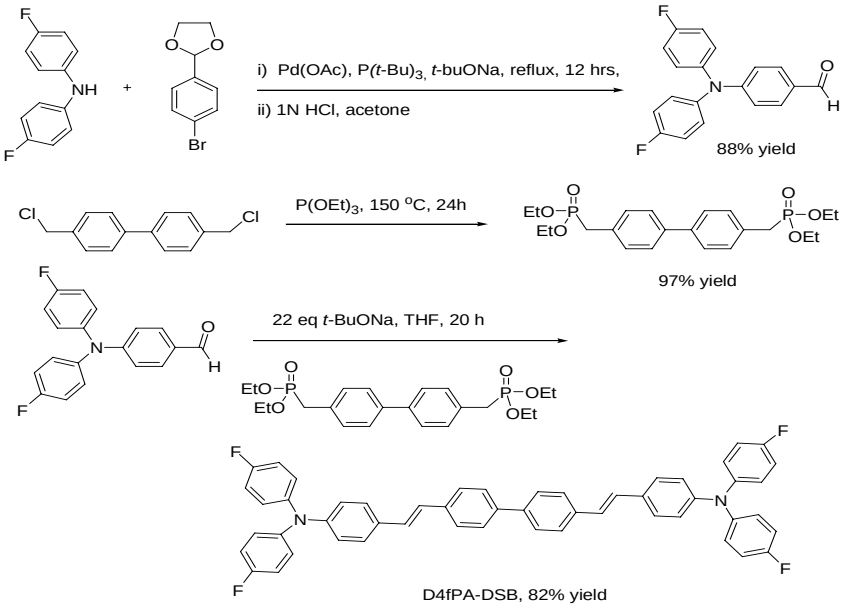
Fig. 4. Structures of mCP and CBP.



Scheme 4. Synthesis of PAP-NPA where Ar_1=phenyl and Ar_2 is naphthyl.

chromaticity diagram shows that mixing of two colors of appropriate CIE coordinates should yield a white color and if a low energy component is present as a dopant dispersed uniformly in a higher energy host and accepts only partially energy from the host, a single white emission layer can be generated. Color variation due to a shift in recombination zone should be minimized in such a configuration. Using this idea, Tao *et al.* reported very bright white organic LED[35]. The emission layer composes of a layer of blue-emitting material 4-{4-[N-(1-naphthyl)-N-phenylaminophenyl]}-1,7-diphenyl-3,5-dimethyl-1,7-dihydro-dipyrazolo[3,4-b;4′3′-e]pyridine (PAP-NPA) uniformly doped with orange-emitting rubrene. The PAP-NPA is a new blue-emitting material with high Tg of 102 °C synthesized by Tao *et al.* according to the following Scheme 4[42].

With this blue-emitting PAP-NPA layer lightly doped with rubrene as the source of emission and TPBI as the electron-transporting layer, the color stability as a function of bias was greatly improved for the white-emitting device. Very high performance was achieved with the simple three-layer structure of ITO/NPB/PAP-NPA : 0.5% rubrene/ TPBI/Mg : Ag or a two-layer device of ITO/PAP-NPA : 0.4% rubrene/ TPBI/Mg : Ag. The emitting layer can be made of phosphorescent and/ or fluorescent materials. The reported power efficiency of fluorescent white OLED at 100 cd/m^2 is 5.0 lm/W of two-wavelength or 7.2 lm/W of three-wavelength[43,44]. Fluorescent white OLEDs with improved power efficiency are thus being pursued. To effectively raise power efficiency, the device must be thin, have low carriers injection barriers, and possess effective carriers and excitons confining function[45,46]. More importantly, in the incomplete energy transfer guest-host system[47], the employed light-emitting host must have high electroluminescent (EL) efficiency and exhibit good energy transfer efficiency to the guest. Keeping this in view, Jou *et al.* fabricated a high efficiency fluorescent white OLED of two-wavelength using a newly synthesized high electroluminescent blue-green host material of di(4-fluorophenyl)aminodi(styryl)biphenyl (DSB) doped with a red dye, 4-(dicyano-methylene)-2-methyl-6-(julolidin-4-yl-vinyl)-4H-pyran (DCM2)[48]. The resulting white OLED has a maximum external quantum efficiency of 4.8% and a high power



Scheme 5. Synthetic protocol of di(4 fluorophenyl)amino-di(styryl)biphenyl(DSB).

efficiency of 14.8 lm/W at 100 cd/m^2. DSB was synthesized according to the following protocol.

The device fabricated by using this material is thin and has a hole-transporting/ emission/electron-transporting tri-layered structure with relatively low carrier-injection barriers and efficient carriers and excitons confining function.

<Multi-doped materials>

To achieve white emission from WOLED among the other many approaches, one is to dope an active host emitting material with several dyes[49,50]. A new type of white organic light-emitting device has been fabricated on incorporating a single light-emitting layer of bis(2-methyl-8-quinolinolato) (para-phenylphenolato) aluminum (III) (BAlq) doped with 2,5,8,11-tetra-*tert*-butylperylene (TBPe) and 5,6,11,12-tetraphenyl-naphthacene (rubrene)[50]. Structure of host material and dopants are given below :

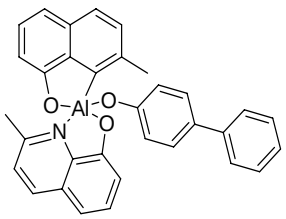


Fig. 5. Structure of BAlq.

The device showed a slight increase in the relative intensity of blue to yellow light as the drive voltage was increased. A white OLED with CIE coordinates (0.34, 0.39) and a maximum external quantum efficiency of 0.68% was achieved at 8 V. It is expected that the CIE coordinates of the white OLED can be optimized further by adjusting the doping concentrations of TBPe and rubrene in BAlq, or using a red dopant instead of rubrene.

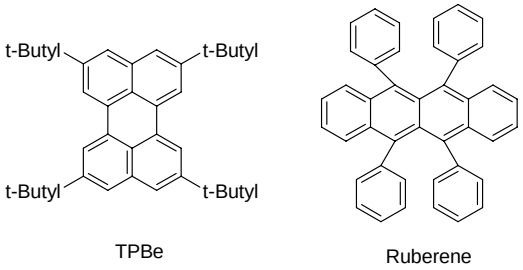


Fig. 6. Structures of dopant TBPe and Rubrene

Similarly, to improve the efficiency and stability of WOLED, Jiang *et al.* proposed triply doped white organic light emitting diode with red and blue dyes in the light emitting layer and a green dye in another layer[51]. Structures of blue emitting layer molecules and dopant molecules are below.

The triply doped white OLED fabricated showed a better performance than the device with single dopant and double dopants. A current efficiency of 6 cd/A and 3.11 lm/W was obtained, which was twice than that of the device with single dopant. The enhanced efficiency is attributed to a cascade energy transfer from host through co-dopant TBPe to DCJTB as well as the high fluorescent efficiency of C545. This triply doped device also showed a higher operating stability. A half lifetime of 22,245 hours at an initial luminance of 100 cd/m^2 with a small raise of voltage during ageing was reached.

4.3. Multi-Emitting Layer System

Among the many other approaches to achieve white emission, one is to use multi-emitting layer. In such devices, white emission is achieved by mixing two complementary colors (e.g. cyan + red) or three primary colors (red, green, and blue) in OLEDs. There are commonly two ways of achieving white OLEDs, one of which is the single-emitting layer white OLEDs[52,53] which also discussed in previous two sections and whose CIE x,y coordinates are independent to current density. This is because there is only one exciton recombination zone, but its EL efficiency is usually low. The other type is the multi-emitting layer white OLEDs[54], the major advantage of which is that it has much higher efficiency than that of the single emitting layer device. The drawback of multi-layered white OLEDs is that the variation of CIE x,y coordinates are often dependent upon drive current density which is due to the shift of exciton-recombination zone. Careful control of the location of exciton-recombination zone and the energy transfer between the host and dopant molecules have been shown to be critical in obtaining a balanced white emission of high efficiency and color with CIE x,y coordinates near (0.33, 0.33).

Hwang *et al.* reported a new white OLEDs structure incorporating a dual-layered emitting layer (EML) of blue and yellow to realize the white emission with high

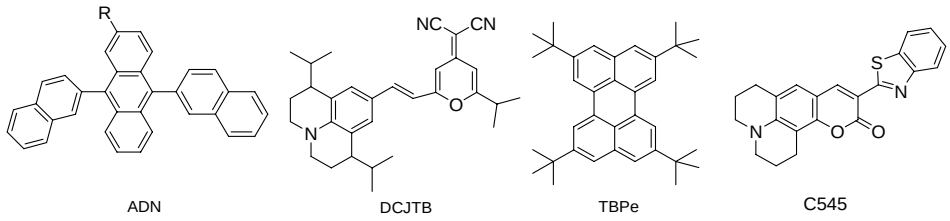


Fig. 7. Structure of blue emitting molecule (ADN, red dye(DCJTB), green dye(TBPe) and blue dye(C545)

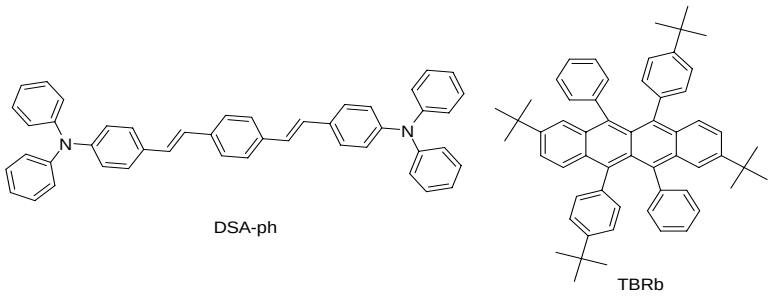


Fig. 8. Structure of blue dopant(DSA-ph) and yellow dopant(TBRb).

efficiency[55]. The blue and yellow dopants reported are *p*-bis(*p*-N,N-di-phenyl-aminostyryl)benzene (DSA-Ph) and 2,8-di (*t*-butyl)- 5,11-di[4-(*t*-butyl)phenyl]-6,12-diphenylnaphthacene (TBRb), respectively. The strategy of inserting both electron and hole blocking layers to control the recombination zone in the dual-emitting-layered white OLEDs is shown to be effective in suppressing the color-shift at different drive current density. In terms of operational stability, it was found that the white OLEDs is more stable than the blue component device. It seems that adding TBRb in NPB can improve the balance of carrier injection into emitting layer to reach longer operational durability. Structure of the novel dopants are below.

Similarly a three-layered emitting WOLEDs with three wavelengths were fabricated by separating the exciton recombination zone into three emitter layers, generating a sky blue, green, and red color by Yeom *et al*[56]. A sky blue emission originated from the 4,4′-bis(2,2′-diphenylethen-1-yl)biphenyl (DPVBi) layer. A green emission originated from a tris(8-quinolinolato)aluminum (III) (Alq3) host doped with a green fluorescent, 10-(2-benzothiazolyl)-1,1,7,7-tetramethyl-2,3,6,7-tetrahydro-1H,5H,11H- [1]benzopyrano [6,7,8-ij]-quinolizin-11-one (C545T) dye. An orange emission was obtained from the N,N′-bis(1-naphthyl)-N,N′-diphenyl-1,1′-biphenyl-4,4′-diamine (NPB) host doped with a red fluorescent dye, 4-(dicyanomethylene)-2-*tert*-butyl-6-(1,1,7,7-tetramethyljulolidyl-9-enyl)-4H-pyran (DCJTB). A white light resulted from the partial excitations of these three emitter layers by controlling the layer thickness and concentration of the fluorescent dyes in each emissive layer simultaneously. Structure of three emissive layers is given below.

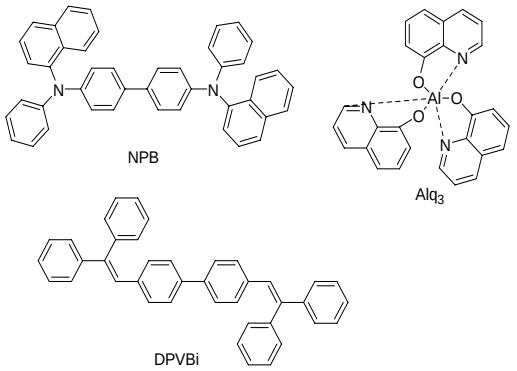


Fig. 9. Structures of emitting layer molecules

In this work, very bright WOLEDs with a maximum luminance exceeding 50,000 cd/m^2 and a high external quantum efficiency, η_{ext} of up to 2.65% was obtained. The white light was originated from the partial excitation of the three emitter species with simultaneous emission achieved by controlling the layer thickness and concentration of each

fluorescent dye in the emissive layers. The EL spectra of the white light-emitting device were not sensitive to the driving voltage investigated in this study, covering the three RGB peaks in the visible range. The color coordinates of the doped devices were well within the white range with increasing bias. The white device has a maximum luminance of approximately 53,300 cd/m^2. At a luminance of approximately 100 cd/m^2, the external quantum and power efficiencies were 2.62% and 3.04 lm/W, respectively.

5. Conclusion

Many organic, polymeric and inorganic white light emitting materials have been developed but their efficiency, CIE values and CRI are still not ideal. Among many approaches to achieve white emission, single component molecules is more desirable and demanding because of many advantages such as better stability, reproducibility and simple fabrication process. But only few single component white light emitting materials are reported, so there is a need of synthesis of new single component materials for white OLEDs. In host-guest system, single doped material is preferred as it significantly simplifies the structure of WOLED, relative to multi doped system. It also solves the problems associated with differential doping aging.

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