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Review

Materials and Devices toward Fully Solution Processable Organic Light-Emitting Diodes†

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Organic light-emitting diodes (OLEDs) have been successfully developed and have now entered the commercial marketplace. Besides the impressive performance as displays, one of the key advantages of OLEDs is that they can potentially be made entirely by solution process and thus are more suitable for low cost, large area flexible displays and white lighting panels. To realize this, many efforts have been devoted on the development of solution processable light-emitting materials and charge transporting materials as well as electrode materials, which have resulted in the successful demonstration of high performance fully solution processed OLEDs and have opened a way to achieve all printable roll-to-roll organic optoelectronic devices.

1. Introduction

Since the discovery of small molecule and polymer organic light-emitting diodes (OLEDs),[1,2] research from both academia and industry have been increasing steadfastly in this area for their great potential in large area flat panel displays and solid-state lighting panels. Impressive scientific and technological progress has been achieved in this field, and varieties of OLED products have now been commercialized. Compared to the other display technologies, OLEDs can be fabricated by solution processing technologies such as spin-coating or inkjet printing and are more suitable for flexible display applications. Therefore, OLEDs are considered as a competitive candidate for the next generation large-size high-resolution displays and solid-state lighting panels, combining the advantages of ease of fabrication and low production cost.

Because the organic semiconductors are critical to the performances of OLEDs, in the past decades numerous efforts have been put in the design and synthesis of new materials and many types of organic semiconductors (such as small molecules,[3,4] conjugated polymers,[5] dendrimers,[6,7] etc.) have been developed for OLEDs. Among them, conjugated polymers are considered as one of the most promising candidates due to their solution processable nature.[2,5] Later on, dendrimers, another important class of macromolecule, were also developed and successfully used in solution processed OLEDs.[6,7] Both conjugated polymers and dendrimers exhibited much better solution processabilty in OLEDs fabrication than most of light-emitting small molecules. More recently, by adopting novel molecule structure design or processing strategies, solution processed small molecule OLEDs were also successfully demonstrated and showed comparable performance to those vacuum evaporated processed counterparts.[8,9]

Although tremendous progress has been made in developing solution processable organic semiconductors, there are still big challenges to overcome for realizing "fully" solution processed OLEDs. OLEDs require a multilayer device structure to ensure good performance, which typically includes a transparent and conductive indium tin oxide (ITO) anode, a hole transporting/injection layer (HTL), an emissive layer (EML), an electron transporting/injection layer (ETL), and a metallic cathode (Figure 1)[10−12]. Thus, the anode and cathode materials should also be solution processable for fully solution processed OLEDs. Moreover, the interface mixing/erosion problems of HTL/EML, EML/ETL, and EML/Electrode interfaces during the solution process of OLEDs also need to be solved. As a consequence, developing solution processable electrode materials and solution processable functional organic semiconductors (such as cross-linkable organic semiconductors[12] and water/alcohol soluble materials[13,14]) suitable for fabrication of multilayer devices without interlayer mixing are becoming important topics of the field. Encouraging results have already emerged in these areas due to the continuous growth of research interest. By combining these research efforts, fully printable light-emitting devices have been successfully demonstrated.[15] Through further research, such design concepts should be able to be adapted to industrial scale technologies such as roll-to-roll process or inkjet printing, thus opening up opportunities to the commercialization of fully solution processed OLEDs. In this article, we will review the recent developments of materials and devices for fully solution processable OLEDs. We will mainly focus on the development of solution processable HTL/ETL materials and solution processable anode/cathode materials.

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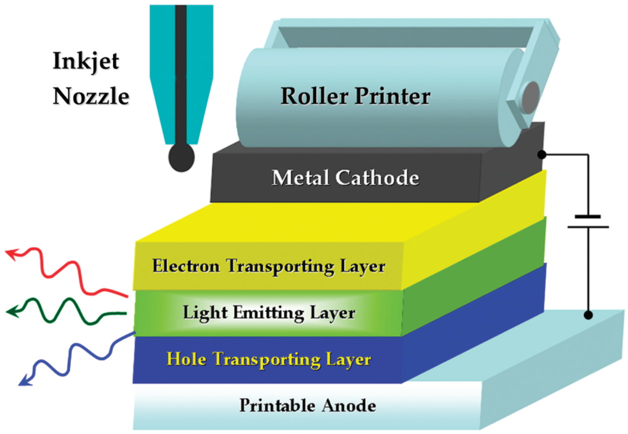

Figure 1. Schematic structure of solution processable OLED devices.

Advances in solution processable EML materials that address the multilayer device problems will also be reviewed.

2. Hole/Electron Transporting/Injection Materials

In order to achieve high power efficiency (PE) in OLEDs, effective and balanced charge injection and transport is necessary. HTL and ETL layers are commonly used to improve the charge injection from anode and cathode, respectively. Many ETL/HTL materials designed for the thermal evaporation process are also solution processable. However, since these materials and EML materials have similar solubility in common organic solvents used for solution processing, such as chlorobenzene or *p*-xylene, the interface mixing/erosion becomes a severe problem. Therefore, the fabrication of well-defined multilayer OLED devices through solution processing is a key challenge. Currently there are two ways to address this problem: the first one is to develop cross-linkable materials,[12] making the already processed cross-linked layer invulnerable to solvent erosion from the new layer. The second way is to develop water/alcohol soluble materials,[13,14] so that adjacent layers can be processed by orthogonal solvents, thus eliminating the interface mixing problem.

2.1. Cross-Linkable HTL Materials. Poly(3,4-ethylenedioxythiophene):poly(styrenesulfonic acid) (PEDOT:PSS, Figure 2) has relatively high conductivity and is almost transparent in the visible region with good stability. Moreover, PEDOT:PSS is a water-dispersed polyelectrolyte emulsion with good film-forming properties, which can avoid the interface erosion of commonly used low polarity organic solvent (such as toluene, chloroform, etc.) processed EML materials. The conductivity of PEDOT:PSS thin films can be tuned by changing the ratio between PEDOT and PSS or controlling the film morphology. Highly conductive PEDOT:PSS can be used directly as anodes without the need of ITO, and such usage will be reviewed in later sections. Consequently, PEDOT:PSS has been widely used as HTL in OLEDs.[16] However, there are several serious drawbacks for the use of PEDOT:PSS as HTL, such as high acidity which causes the corrosion of the ITO anode,[17] poor hole injection for most of the blue- and green-emitting materials due to the mismatched energy levels, and exciton quenching at the interface between PEDOT:PSS and emissive materials.[18] Furthermore, the question still remains as to whether PEDOT:PSS has adequate electron-blocking capacity. Aiming at better HTL materials in solution processed OLEDs, many efforts have been made in the development of cross-linkable HTL materials that can provide excellent solvent resistance after thermo- or photo-cross-linking.[12] The most straightforward way to develop cross-linkable HTL materials is to introduce efficient cross-linking functionalities into traditional HTL molecules, such as *N*,*N*′-(3-methyl-phenyl)-1,1′-biphenyl-4,4′-diamine (TPD), 4,4′-bis[*N*-(1-naphthyl-1)-*N*-phenyl-amino]-biphenyl (NPB), tri(*N*-carbazolyl)-triphenylamine (TCTA), and so forth. By using various cross-linking functional substitute groups, such as trifluorovinyl ether,[19] styrene,[20] siloxane,[21] oxetane,[22] acrylate,[23] and so forth, a large number of cross-linkable HTL materials were developed (Figure 3). Among these cross-linkable HTL materials, the trifluorovinyl ether and styrene-functionalized materials can be cross-linked upon heating without using initiators, while the oxetane-based materials will be cross-linked upon illumination by the ultraviolet (UV) light in the presence of the photoacid initiator. The chlorosilyl moieties on the siloxane-functionalized materials are highly reactive and readily undergo hydrolysis and condensation to form highly cross-linked films in the presence of moisture. After cross-linking, all of these materials can form robust HTL films with good solvent resistance for solution-processed multilayer OLEDs.

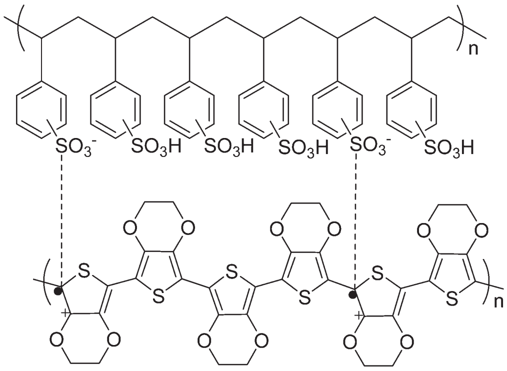

Figure 2. Chemical structure of PEDOT:PSS.

One of the key advantages of these cross-linkable hole-transporting materials is that they can be directly stacked layer by layer onto each other without intermixing using solution process. Thus, multiple HTLs with gradually changed HOMOs can be easily incorporated into a single OLED device to enhance the hole injection/transport (so-called cascade HTL structure). For example, trifluorovinyl ether based materials PS-TPD-TFV and TriTFA-TCTA (Figure 3) have been used as cascade HTLs in a blue-emitting phosphorescent OLED with configuration ITO/PS-TPD-TFV/TriTFA-TCTA/10% FIr6 in PVK/TPBI/CsF/Al, where bis(4′,6′-difluorophenylpyridinato)-tetrakis-(1-pyrazolyl)borate (FIr6) was doped into a poly(*N*-vinyl-carbozle) (PVK) host as EML and 2,2′,2′′-(1,3,5-benzenetriyl)tris[1-phenyl-1H-benzimidazole] (TPBI) was used as the ETL. The resulted device exhibited good device performance with a maximum external quantum efficiency (EQE)

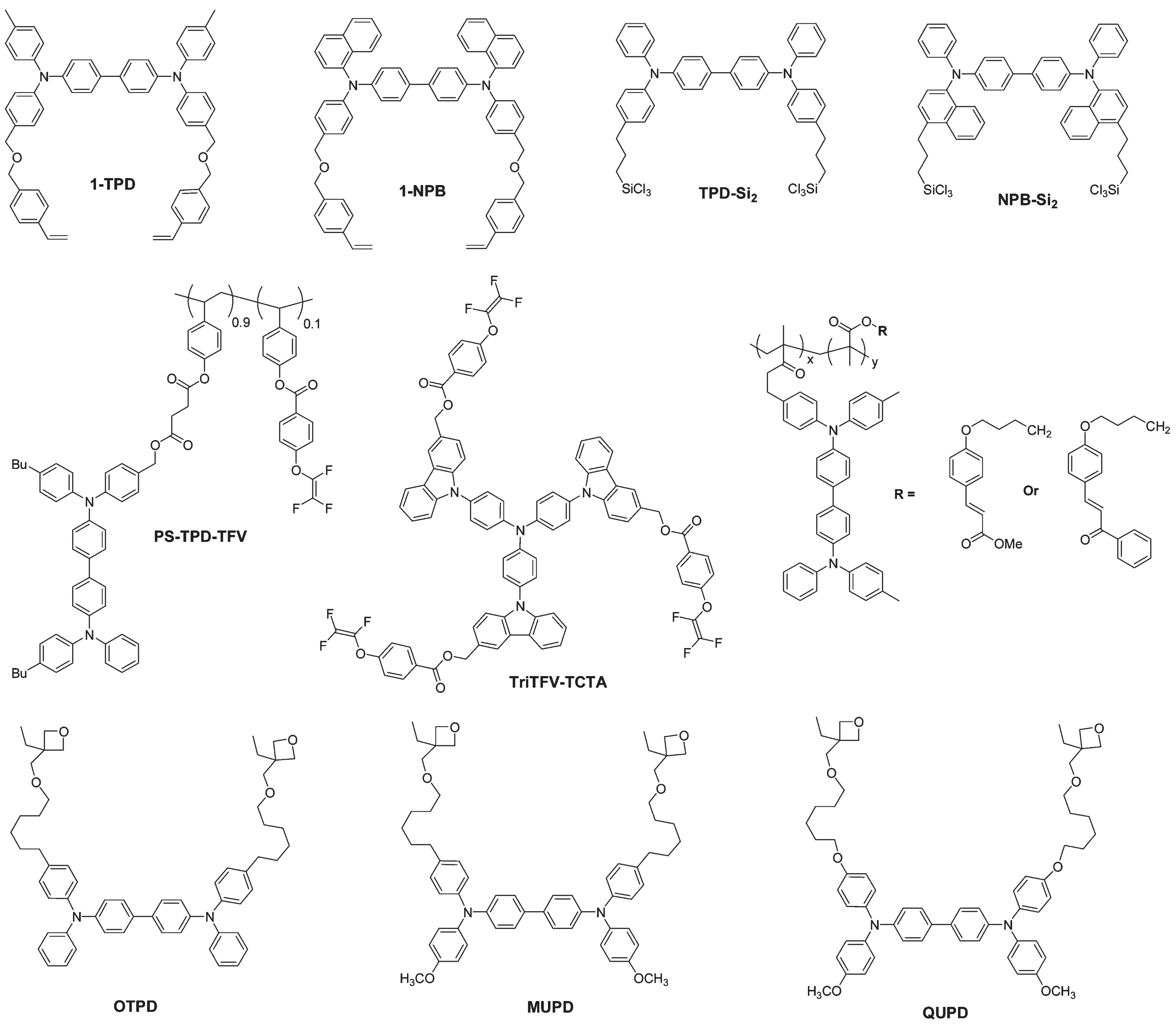

Figure 3. Examples of cross-linkable HTL materials.

of 3.0% and luminous efficacy (LE) of 6.1 cd/A (~5 cd/A at 1000 cd/m^2), which is much higher than that (1.2% and 2.4 cd/A, respectively, at ~20 cd/m^2) of the reference device with single PS-TPD-TFV HTL (all LE data in this review are forward viewing results unless specified).[24] Moreover, the conductivity of these HTLs can be greatly improved by redox chemical doping, which can dramatically reduce the driving voltage of the resulted device and greatly enhance the PE of the devices. By using $NO^+SbF_6^-$ doped QUPD as the first HTL and cross-linked OTPD as the second HTL, Neher and Meerholz et al.[25] reported a series of highly efficient polymeric electrophosphorescent LEDs. The second cross-linked OTPD HTL not only formed cascade HTLs with the first doped QUPD HTL but also functionalized as a separation layer to avoid the quenching of EML layer by the doped carrier transporting layer. The devices based on the doped cascade HTLs showed a much improved device performance compared to the device with undoped HTLs. For instance, the green-emitting phosphorescent OLED based on $Ir(mppy)_3$ (device structure: ITO/HTL/PVK:PBD: $Ir(mppy)_3$/CsF/Al) exhibited an maximum LE of 40 cd/A and PE of 38 lm/W (~30 cd/A and ~20 lm/W at 1000 cd/m^2) when using PEDOT:PSS/MUPD as the HTL, while the device's maximum LE and PE were improved to 67 cd/A and 65 lm/W (56 cd/A and 29.7 lm/W at 1000 cd/m^2), respectively, by using the doped QUPD/OTPD as cascade HTL. Similar enhancements were also observed in the red-emitting phosphorescent OLED, which showed a maximum LE of 11.7 cd/A and PE of 10.7 lm/W (6.7 cd/A and 2.5 lm/W at 1000 cd/m^2).[25]

2.2. Water/Alcohol Soluble HTL Materials. Besides the cross-linkable materials, new water/alcohol soluble HTL materials were also developed as high efficiency HTLs for PLEDs, due to their unique solubility, which offers them an erosion-resistant property to most of the commonly used EML materials. Since most of the commonly used conjugated polymer active materials, such as poly[2-methoxy-5-(2-ethylhexyloxy)-1,4-phenylenevinylene] (MEH-PPV),

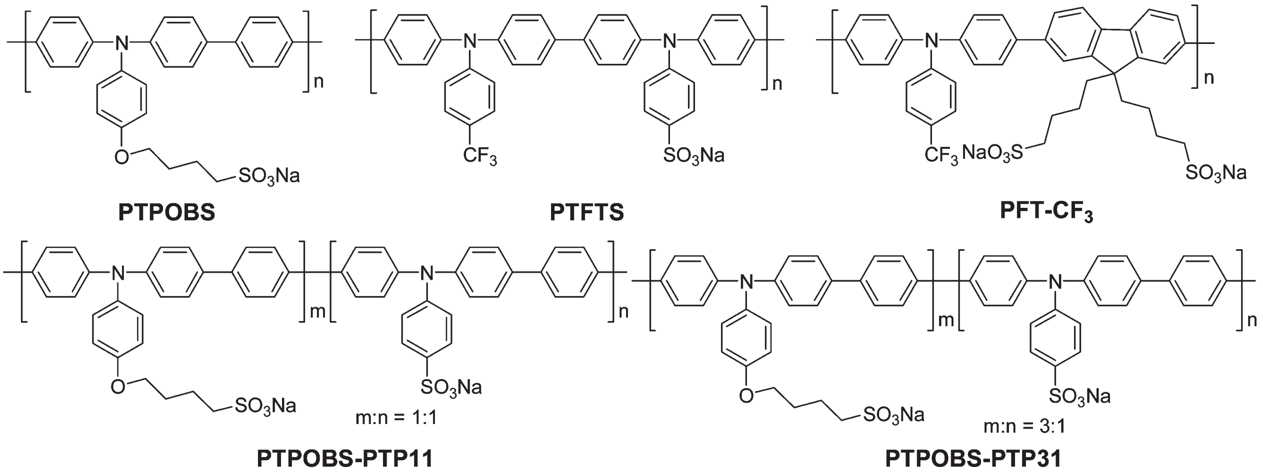

Figure 4. Molecular structures of water/alchohol soluble HTL materials.

poly[2-(4-(3′,7′-dimethyloctyloxy)-phenyl)-*p*-phenylenevinylene) (P-PPV), etc., do not dissolve in highly polar solvents, water/alcohol soluble charge transporting layer materials have been extensively explored for fabricating multilayer devices with those traditional conjugated polymers. Shi et al[26−28] reported a series of triphenylamine-based conjugated polyelectrolytes that can function as HTLs in PLEDs (Figure 4). The sulfonate groups among these polymers offer them good solubility in polar solvents and can withstand the erosion of commonly used nonpolar organic solvents (such as toluene, chlorobenzene, and others). Thus, all of these polymers can efficiently avoid interface mixing when they are used as HTLs during the fabrication of multilayer PLEDs. The energy levels of these polymers can be well controlled by modifying the substitution groups on triphenylamine units. With the electron donating butyloxy groups, poly[*N*-(4-sulfonatobutyloxyphenyl)-4,49-diphenylamine-*alt*-1,4-phenylene] sodium salt (PTPOBS) has a relative high HOMO level of −5.08 eV, while the poly[*N*-(4-sulfonatophenyl)-4,4′-diphenylamine -*alt*-*N*-(*p*-trifluoromethyl)phenyl-4,4′-diphenylamine] sodium salt (PTFTS) HOMO level is at around −5.23 eV, indicating the electron density of the PTFTS backbone is greatly reduced by the electron withdrawing ($-CF_3$, $-SO_3^-$) groups. With the $-CF_3$ groups on the triphenylamine units, poly[9,9-bis(4-sulfonatobutyl)-fluorene-*alt*-*N*-(*p*-trifluoromethyl)phenyl-4,4-diphenylamine] sodium salt (PFT-CF_3) exhibited similar HOMO level of −5.22 eV to that of PTFTS. All of them showed similar LUMO levels, which are −2.21 eV, −2.24 eV, and −2.26 eV for PTPOBS, PTFTS, and PFT-CF_3, respectively. The relative high-lying LUMO levels indicate that all of these polymers have good electron-blocking property when used as HTLs in PLEDs. The hole-injection and transport properties of these polymers were verified by using them as HTLs in PLEDs with configuration ITO/HTLs/PFO-DBT15 or PFO-BT15/Ba/Al, where the red-emitting polymer poly(9,9-dioctylfuorene-*co*-4,7-dithien-2-yl-2,1,3-benzothiadiazole) (PFO-DBT15) or green-emitting polymer poly(9,9-dioctylfuorene-*co*-2,1,3-benzothiadiazole) (PFO-BT15) were used as the EMLs. The reference devices with PEDOT:PSS HTL were also made for comparison. All of the devices exhibited similar electroluminescence (EL) spectra exclusively from the EML materials, regardless of the HTL materials used, indicating that the recombination zone is well confined in the EML. Device with these newly developed HTLs exhibited much better device performances with lower turn-on voltages, higher brightness, and enhanced EQEs compared with the reference devices with bare ITO as anode or a device with PEDOT:PSS as HTL. For example, the device ITO/PTFTS/PFO-DBT15/Ba/Al exhibited a turn-on voltage of 3.2 V which is lower than those of devices with bare ITO anode (6.2 V) or PEDOT:PSS HTL (3.6 V). The devices' EQEs at the current density of about 33 mA/cm^2 were also improved from 0.3% for the bare ITO device and 0.9% for the device with the PEDOT:PSS layer to 1.5% for the device using PTFTS as HTL. The significant improvement in the device performance is mainly due to much better hole injection (lower HOMO) and electron blocking ability (higher LUMO) of the HTL materials compared to PEDOT:PSS and bare ITO. Moreover, it was found that the energy level of these HTL materials can be tuned by controlling the ratio of different triphenylamine units on the polymer main chain.[28] By increasing the sulfonated triphenylamine units' ratio, the HOMO can be changed from −5.08 eV for PTPOBS to −5.10 eV for PTPOBS-PTP11 and −5.16 eV for PTPOBS-PTP31, respectively. Both PTPOBS-PTP11 and PTPOBS-PTP31 also showed better device performance when used as HTLs in PLEDs compared to those of devices with PEDOT:PSS HTL.

Li et al. reported the use of a sulfonated poly(diphenylamine) (PDPA) as efficient HTLs in PLEDs.[29] Different from HTL materials in Figure 4, PDPA is a kind of self-doped conductive polymers, which usually have higher conductivities than those of undoped ones (Figure 5). PDPA was obtained by sulfonation of a neutral poly(diphenylamine) precursor polymer. The NMR, elemental analysis, and Raman spectra results showed that the $-SO_3H$ groups are bonded to every diphenyl ring of the polymer main chains and indeed exhibited a self-doping effect. Due to the high concentration of attached $-SO_3H$ groups, PDPA could be spin-coated from water, and the as-formed film exhibited a very smooth surface, a moderate conductivity (10^{-4} S/cm), and high transparency in

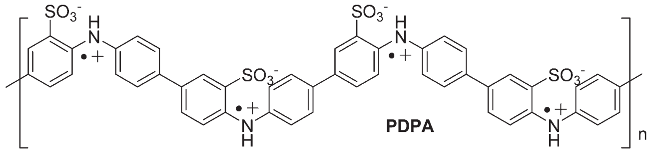

Figure 5. Molecular structures of sulfonated PDPA.

the visible region. The work function of PDPA is around 5.2 eV, which is determined by UPS measurement. The hole transport/injection ability of PDPA was examined in MEH-PPV based PLEDs with configuration: ITO/PDPA/MEH-PPV/Ca/Al. It was found that The device's LE reached 2.0 cd/A (~1.6 cd/A at 1000 cd/m^2), which is slightly better than that of reference device with PEDOT:PSS HTL.

2.3. Water/Alcohol Soluble ETL Materials. Development of suitable ETL materials for solution processed OLEDs was focused on synthesizing new conjugated polymers with solubility totally orthogonal to commonly used transporting/injection or active materials.[12,13] Cao and co-workers[30−33] reported highly efficient PLEDs based on amino-/ammonium-functionalized polyfluorene ETL and disclosed the unique electron injection properties of these water/alcohol soluble conjugated polymers for the first time. After that, more attentions have been made by other groups in this area, resulting in rapid development of water/alcohol soluble conjugated polymer ETL materials and deeper understanding on the mechanisms of their related devices.[13,14,34−42]

Figure 6 shows several typical examples of water/alcohol soluble conjugated polymer ETL materials. All of them have a conjugated main chain and polar pendant group substituted side chains. The conjugated main chain endows them with good conductivity for charge transporting, and the polar pendant group substituted side chain endows them with good solubility in highly polar solvents (such as methanol, ethanol, etc.) for solution processed multilayer engineering. Moreover, it was found that these water/alcohol soluble conjugated polymer ETL materials can also dramatically improve charge injection from high work-function metal electrodes into organic active layers resulting in significant enhancement in the device performance. As a result, these water/alcohol soluble conjugated polymers not only have the common functionalities as those traditional ETL materials, such as good electron transporting/hole blocking ability, eliminating the cathode quenching, etc., but can also enhance electron injection from high work-function metals (such as Ag, Au, etc.), which open a way toward the "fully" solution processed OLEDs.[15] For example, using poly[(9,9-bis(3′-(*N*,*N*-dimethylamino)propyl)-2,7-fluorene)-*alt*-2,7-(9,9-dioctylfluorene)] (PFN) as ETL, P-PPV exhibited LE of 11.6 cd/A at 3648 cd/m^2 (~10 cd/A at 1000 cd/m^2) with device configuration: ITO/PEDOT:PSS/P-PPV/PFN/Au, which is much higher than that of P-PPV in the plain Au cathode device, showing a LE of 0.006 cd/A at 0.78 cd/m^2.[33] Device physics studies showed that this unique property was attributed to the dipole formation between the water/alcohol soluble conjugated polymer ETL and metal cathode, while the movement of the counterions was also found to play an important role on the electron injection properties of conjugated polyelectrolyte-based ETL.[14] This unique electron injection property made it possible to use a solution processed metal cathode (such as Ag paste) in high efficiency OLEDs, resulting in the realizing of "fully" solution processed OLEDs.[15]

3. Solution Processable Electrode Materials

In most cases, ITO glass is used as the anode, and vacuum deposited metal is used as the cathode in OLEDs. ITO has many advantages as anode material, since it possesses high conductivity and high optical transparency simultaneously, and the work function is suitable for hole injection for certain emitting polymers of relatively high HOMO levels. However, ITO is not perfect for OLED applications.[43,44] First, it is not resistant to mechanical bending due to its brittleness, which is detrimental to the performance of flexible OLED devices. Second, the high processing temperature of ITO makes it difficult to form a high quality anode on flexible substrates such as the commonly used poly(ethylene-terephthalate) (PET). Third, ITO cannot be easily adapted to the solution process, while solution processable anode material is vital for the development of all solution processed OLEDs. Furthermore, the limited global reserve of indium resulted in ever-increasing prices of ITO, which will dramatically raise the manufacturing cost of OLEDs. Considering all these factors, it is urgent to look for substitute anode materials for ITO. The substitute material should be highly conductive and optically transparent like ITO, and it should also be solution processable. On the cathode side, the metal cathodes are exclusively processed by vacuum deposition for OLED these days, which is a big obstacle to fully solution processed OLEDs. Consequently, there is an urgent need to develop high conductivity solution processable cathode materials for OLED applications.

3.1. Carbon Nanotubes and Graphene. Carbon nanotubes (CNTs), especially single-walled carbon nanotubes (SWNTs), have been extensively studied due to their exceptional electrical and mechanical properties.[45,46] SWNTs have suitable work function (4.7−5.2 eV) for application as anodes in OLEDs. In flexible electronics applications, CNT films are superior to ITO because they can be bent to acute angles without fracture and can adhere strongly to plastic substrates such as PET and PMMA (poly(methyl methacrylate)) due to excellent surface energy matching. Furthermore, CNT can form stable dispersions in various solvents that are printable.[47,48] Zhang et al.[49]comparatively studied the transparent conductive thin films based on two kinds of commercial CNTs: high-pressure CO conversion (HiPCO) nanotubes and arc-discharge nanotubes. It was found that arc-discharge nanotubes exhibited better performances, and the optimized films show a typical sheet resistance of ~160 Ω/□ at 87% transparency. The OLEDs based on the optimized configuration SWNT/PEDOT:PSS/NPD/Alq$_3$/LiF/Al exhibited

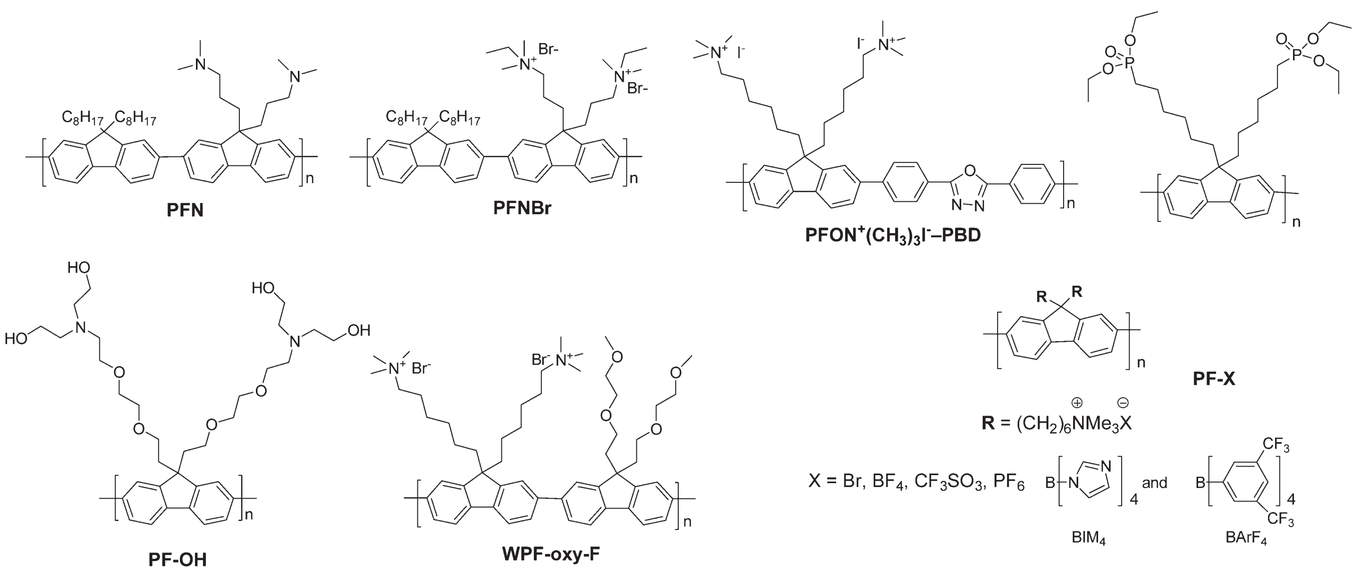

Figure 6. Examples of water/alcohol soluble conjugated polymer ETL materials.

EQEs from 0.21% to 0.34% within a wide bias range from 0.6 to 20 V. Li et al.[50] reported high efficiency polymer-based OLEDs using SWNT films on flexible PET substrates as transparent, flexible anodes, which showed a LE of 1.6 cd/A (~1 cd/A at 1000 cd/m^2) and a maximum brightness of 3500 cd/m^2. Ou et al.[51] reported high performance OLEDs with transparent and conductive CNT anodes after various modifications, including PEDOT:PSS, HNO_3 treatment, and polymer encapsulation. The device with PEDOT:PSS composite modified CNT anodes showed a maximum luminance of ~9000 cd/A and LE of ~10 cd/A at 1000 cd/m^2, similar with ITO-based OLEDs.

Graphene is another promising candidate for solution-processed electrode materials. Graphene is a single sheet of sp^2-bonded carbon atoms, which has very high conductivity and is nearly transparent. Wu et al.[52] reported OLEDs with solution processed graphene thin film transparent conductive anodes, where the graphene anodes were prepared using Hummers' method[53] followed by vacuum annealing to reduce the graphene oxide. The thickness of the resulted graphene film is ~7 nm with a sheet resistance of ~800 Ω/□ and 82% transparency at 550 nm, which is nearly 2 orders of magnitude higher than the ITO sheet resistance of the same thickness. Nevertheless, the OLEDs based on solution-processed graphene thin film with the configuration glass/graphene/PEDOT:PSS/NPD/Alq_3/LiF/Al exhibited a turn-on voltage of 4.5 V and a luminance of 300 cd/m^2 at 11.7 V, which is comparable to that of the reference device with ITO anode, showing a turn-on voltage of 3.8 V and a luminance of 300 cd/m^2 at 9.9 V. Moreover, the EQE and PE of graphene based OLED are comparable with that of the ITO control device despite the higher sheet resistance and different work-function of the graphene anode. It was proposed that the conductive PEDOT:PSS layer on top of the anode removed the effects of small differences in work function that may exist between ITO and graphene. It should be noted that the conductivity of such a graphene anode is far lower than theoretical limits of ideal graphene since solution processing of functionalized graphene contains multiple grain boundaries and incorporates lattice defects and oxidative traps that limit charge carrier transport. These problems must be addressed in future research.

3.2. Solution Processed Layer-Structure Transition Metal Dichalcogenide. Solution-processable anodes can also be developed from inorganic materials, using layer-structure transition metal dichalcogenide (LTMDC), in which one hexagonally packed sheet of metal ions is sandwiched between two hexagonal sheets of chalcogen. Friend's group first reported the use of aqueous solution processed LTMDC as hole-injecting and electron-blocking layers for PLED.[54] MoS_2 was chemically processed into thin film through *n*-BuLi intercalation and exfoliation in water. To improve wetting of the LTMDC surface by the polymer and to shift the emission zone away from the electrodes, a transition metal oxide (TMO) layer with different thickness was introduced through simple oxidation of MoS_2 with different O_2 plasma treatment time. The high work function of the MoS_2 layer reduces the barrier for hole injection from the ITO anode into the polymer, and consequently, high hole injection efficiency was achieved. The oxide layer prevents electron extraction from the anode, and the consequential build up of space charge enhances hole injection. The performance of devices with a structure of ITO/MoS_2 (O_2 plasma treated)/polyfluorene/Ca/Al showed LE and PE as high as 7.3 cd/A and 9.6 lm/W, respectively, at 1000 cd/m^2 with applied bias of 2.4−2.5 V. The OLED operation was also modeled using experimentally determined parameters, including work functions, carrier mobility, and barrier properties.[55] The good agreement between experiment and model corroborated the hypothesis that the MoS_2/MoO_3 anode acts as a high work function hole injection layer coupled with an electron blocking layer. Later, X-ray photoelectron spectroscopy studies and optical transmittance spectrum studies confirmed the existence of the TMO layer on the top of the LTMDC layer introduced by plasma treatment, and it was found that the thickness of the TMO layer increases with plasma treatment time, and thus the electron blocking ability of the anode is controllable.[56]

On the basis of these works, they extended the use of LTMDC further by using the metallic compounds $NbSe_2$ as semitransparent anodes in PLEDs replacing ITO.[57] $NbSe_2$ films were deposited through intercalation and exfoliation onto oxygen plasma treated glass substrates. The LTMDC films have a sheet resistance of $\sim 1000\ \Omega/\square$, and a transmittance of greater than 60% over the range of 400−800 nm. Typical diodes with $NbSe_2$ anodes show high luminance and PE of over 1 lm/W. Some $NbSe_2$ films were oxygen plasma treated to improve the wetting by the polymer. The Nb_2O_5 that is formed acts as an electron blocking layer and significantly reduces leakage currents, which is similar to the effect of MoO_3. Moreover, $NbSe_2$ films were also deposited on oxygen plasma treated plastic substrates such as polyethylene, poly(ethylene terephthalate), and poly(vinyl phenol) (PVP). However, the devices showed high leakage currents due to the high surface roughness of the plasma treated substrate.

3.3. PEDOT:PSS and PANI. Currently, there are two types of polymer materials that have the potential to replace ITO as the transparent anode for OLED applications: PEDOT:PSS and polyaniline (PANI). These two materials are well studied conjugated polymers with excellent mechanical stability and flexibility by nature, which is very suitable for flexible display applications. More importantly, they can achieve high conductivity or high transparency, which are crucial factors for OLED applications. However, there are currently two problems that prevent these two materials from becoming practical anode materials. First, there is a trade-off between conductivity and transparency in these two materials, which means it is difficult to achieve high conductivity ($>10^3$ $S\cdot cm^{-1}$) and high transparency ($>90\%$ in the visible region) simultaneously for these materials. Second, the highest conductivity of these materials is still not comparable to ITO (10^3-10^4 S/cm), which will greatly reduce the PE of OLED devices due to higher driving voltage. The current research of PEDOT:PSS and PANI anode focus on addressing these two issues.

PANI is insoluble in common solvents in its intrinsic form. It became a potential candidate for a solution processable transparent anode when Cao et al.[58,59] discovered that camphor-sulfonic acid doped PANI (PANI:CSA) is soluble in *m*-cresol or chloroform and the solution cast films are highly conductive and optically transparent. Gustafsson et al.[60] then successfully fabricated PANI:CSA anodes on PET substrate without ITO and invented the first flexible PLED device, which attracted great attention to the OLED area. Yang and Heeger[61] later used PANI:CSA as anode for MEH-PPV PLED devices and achieved higher quantum efficiency than ITO anode devices. However, independent PANI:CSA anodes must be thicker than 250 nm to achieve high device efficiency, while the transparency is reduced to 70% under such thickness, which limits further improvements in efficiency. Recently Fehse et al.[62] reported the use of a new solution processable PANI formula D1033 as anode which has conductivity of 200 S/cm and lower absorbance in the visible region than PANI:CSA. The multilayer structured OLED devices using D1033 anode showed high PE and low turn on voltage. However, the transparency of D1033 anodes is still inferior to ITO anodes, especially in the red spectrum region due to the absorption of PANI.

PEDOT:PSS was developed by Bayer AG first as solution processable antistatic coating material.[63] It was then introduced into PLED devices as the hole injection layer (HIL) between the active layer and ITO anode.[64] Hence, it became the standard HTL material for PLED devices and proved to be an effective HTL material for small molecular OLED devices as well.[65] It was later discovered that adding a small proportion of high boiling point polar solvent such as polyalcohol[66,67] or dimethyl sulfoxide (DMSO)[68] into the PEDOT:PSS aqueous solution will significantly enhance the conductivity of the solution processed PEDOT:PSS thin films without compromising the transparency. Kim et al.[69] first demonstrated that such highly conductive PEDOT:PSS can serve as independent anode materials for OLED devices without the need of ITO. However, the conductivity of PEDOT:PSS anodes made by Kim et al. is still magnitudes lower than ITO, therefore crippling the efficiency of the device. Nevertheless, it was a clear demonstration of the potential of PEDOT:PSS as anode material. Ouyang et al.[70] used ethylene glycol as polar additive and acquired PEDOT:PSS thin films with conductivity as high as 160 S/cm, and the MEH-PPV PLED devices using such PEDOT:PSS films as anode showed comparable performance with ITO/PEDOT:PSS devices. More recently, Fehse et al.[71] implemented a commercially available high conductivity PEDOT:PSS Baytron PH500 (conductivity reaches 500 S/cm when DMSO is added) as anode in small molecular OLED devices, where the green and blue emitting devices showed superior performance to ITO anode devices in terms of PE.

It is necessary to enhance the conductivity of PEDOT:PSS to the 10^3 S/cm region without losing the optical transparency for replacing ITO. To realize this, the deeper understanding of the conducting mechanisms of PEDOT:PSS thin films is necessary. PEDOT is a highly conductive conjugated polymer built from ethylenedioxythiophene (EDOT) monomers. Just like PANI, intrinsic PEDOT is insoluble in common solvents and unstable in its neutral state. Polystyrene sulfonate (PSS) is added as charge balancing dopant to form PEDOT:PSS aqueous dispersion,[16] which helps PEDOT stably dispersed in water. As a result, PEDOT:PSS can be readily processed from solution. However, the presence of PSS in the PEDOT:PSS thin film significantly lowers the conductivity due to the insulating nature of PSS. Thus, it is desirable to develop new solution process routes for PEDOT that do not require the use of PSS or any other insulating charge balancing dopants. One way is in situ polymerization, which includes the spin-coating EDOT monomer thin film and oxidative polymerization on film to form transparent PEDOT films,[72] thereby eliminating the need of charge balancing dopants. Ha et al.[73] optimized oxidative polymerization conditions and yielded PEDOT thin films with conductivity of 700 S/cm and transparency exceeding 80%. By changing the monomer's structure, the conductivity of PEDOT can even be improved to 900 S/cm. PEDOT thin films with even higher conductivity can be fabricated by vapor phase polymerization (VPP),[74,75] as the PEDOT prepared by this method has much longer conjugation length compared to other synthesis routes. Levermore et al.[76,77] reported VPP PEDOT anodes with conductivity as high as 1180 S/cm, which is the highest conductivity value of PEDOT reported to date. The PLED devices using such anodes have PEs equivalent to those of ITO anode devices. However, both in situ polymerization and VPP suffer from long processing time and residual reaction agent problems. Moreover, VPP requires an additional low vacuum processing step which is not compatible with large scale solution processes. These problems must be overcome before adapting these methods to mass production of solution processing technologies such as roll-to-roll coating.

3.4. Metal Nanowire Mesh. Metal nanowire mesh (MNM) electrode is a potentially useful anode design for OLED applications. Similar to CNT mesh electrode, it takes advantage of the high conductivity of metals and meanwhile ensures good optical transparency by enabling light to pass through the meshes. More importantly, metal nanowires can be well dispersed in organic solvents and thus solution processable. MNM electrodes can achieve higher conductivity than CNT counterparts of same thickness not only because of the obviously higher bulk conductivity in metal but also because contact resistance between metal nanowires is much less than between CNTs. Therefore, MNM is considered a competitive candidate for ITO substitute besides PEDOT and PANI.[78] Like ITO anodes, MNM anodes require HTL such as PEDOT:PSS to ensure good hole injection.

Lee et al.[79] first demonstrated the potential of MNM electrode by fabricating silver nanowire mesh electrodes on glass substrate. The best electrode has a sheet resistance of 16 Ω/□ and total optical transmittance of 86% between wavelengths of 400 and 800 nm, which is comparable to ITO anodes. De et al.[80] optimized film quality of silver MNM electrodes and achieved a sheet resistance of 13 Ω/□ (conductivity 5×10^4 S/cm) and optical transmittance of 85%. However, simple solution process can only generate random spatial distribution of metal nanowires, which scatters light in oblique incidence angles and may therefore compromise the angle characteristics of OLED emission.[77] Thus, it is desirable to align metal nanowires into more ordered patterns. There are already reports of transparent anodes using well-defined nanosized gold gratings generated by nano-imprint lithography;[81] however, it is still challenging to incorporate such high precision nanopatterning techniques to improve the degree of order in MNM electrodes without compromising the low-cost, high-throughput nature of the solution process.

3.5. Solution Processed Metal Cathode Materials. The cathodes of OLED devices are typically low work-function metal thin films formed via vacuum deposition techniques. Due to high air- and moisture-instability of alkali and alkaline metals in solution and dispersion, high work-function noble Au and Ag metals are necessary for solution processing. In this case barrier height for electron injection becomes a serious problem.[82] There are few reports on solution processed cathodes for OLED applications, since it is difficult to produce a high quality cathode layer with high conductivity and low electron injection barrier through solution processing techniques. The injection barrier problem can be circumvented by introducing ETL between cathodes, which will be discussed in later parts, and the key issue is to develop highly conductive and solution processable or at least printable (in the form of paste or ink dispersion) cathode materials.

Ag is the most studied printable conducting material, because it is one of the most conductive materials among metals. Solution processing of Au[83] and Cu[84] are also studied; however, they receive less attention since Au is less competitive in terms of cost and solution processing of Cu is too complicated. With proper device design, Ag can serve as an effective cathode in OLED devices. There are several technologies that enable solution processing of Ag. The first way is to disperse micrometer sized Ag particles into highly viscous pastes and process it through screen printing.[85] This is the simplest and most reliable way to solution process Ag anodes, since it is a mature technology, which is fully compatible with the roll to roll process, and there are many types of commercial Ag pastes available. The second way is to make Ag inks with Ag nanoparticles monodispersed in low viscosity organic solvents and process through inkjet printing. This method takes full advantage of inkjet printing technology in terms of efficient use of materials, maskless pattering, and high throughput.[86] The printed electrodes can achieve very high conductivity, as Ag electrodes with conductivity comparable to the bulk value are reported.[87] However, the as-printed film is insulating because of the organic or polymeric surfactants wrapped around the Ag nanoparticles, and sintering is needed to decompose the surfactants and melt the nanoparticles together to achieve high conductivity. Although the sintering temperature can be lowered by reducing the size of Ag nanoparticles,[88] the state of the art Ag ink still requires ~150 °C sintering temperature,[89] which is higher than the glass transition temperature of many flexible substrates such as PET and may cause efficiency loss in the active layer. Thereby, the technology for Ag inks with lower sintering temperature needed to be developed for OLED cathode fabrication. The third way is forming Ag through reduction of Ag salts, where a reducing agent is introduced into the Ag salt solutions and the resulted Ag film is formed and deposited onto substrates through the redox reaction. The advantage of this method is that Ag film with high conductivity can be formed at a relativey low temperature. Xia et al.[90] used a commercial formula of Ag salts and reagents to form 40 nm thick Ag electrodes with half bulk conductivity on glass and silicon substrates. Recently

Valeton et al.[91] developed a similar route to solution process Ag electrodes using redox reaction between silver neodecanoate and hydroquinone. High quality Ag films are quickly formed by exposing the solution to UV exposure and no heat treatment is required, and 10% bulk conductivity can be achieved. Although currently there are no reports on the use of such reduction formed Ag electrodes as cathode for OLED devices, it is demonstrated that uniform Ag electrodes can be formed on polymer substrates such as polycarbonate and polystyrene using this method,[92] which may apply to OLEDs in the near future. The effects of residual reagents and solvents on the interfacial properties of solution processed Ag cathodes are crucial to the efficient operation of OLED devices and should be addressed if they are to be incorporated into OLEDs design.

4. Solution Processed Light-Emitting Materials

Organic EML materials can be divided into small molecules, polymers, and dendrimers. In most cases, small molecule based OLEDs are processed via vacuum evaporation, while solution processing of such materials is usually difficult due to the much higher crystallization tendency during solvent drying. Nervertheless, it was found that small molecule also can form good films under proper conditions. In this section we will briefly review the advances in these three types of EML materials.

4.1. Light-Emitting Conjugated Polymers. Conjugated polymers is the most studied family of solution processable EML materials due to their excellent processability in nature. The synthesis, device characterization, and optoelectronic properties of these materials are covered in previous reviews and will not be discussed here in detail.[5] In this section we focus on the development of high efficiency polymer EML materials and the strategies to realize multilayer structure with polymer EML materials. Though many green and red polymer light-emitting materials with excellent LED performance are successfully synthesized, it is hard to develop highly efficient blue light-emitting polymers.[93−95] In most cases, those blue-emitting homopolymers have a large band gap and emit pure blue light in solution and thin films when excited by UV light, while they always exhibit poor color stability and low EL efficiency because of aggregate/excimer[96] or defect formation.[97]

Despite their poor device performance, these large band gap blue emitters can be developed into high-efficiency EML materials by applying dopant−host strategy (Figure 7). Among them, polyfluorenes (PF) are the most extensively explored hosts due to their high photoluminescence (PL) quantum efficiency and good chemical and thermal stability.[93] It was found that by incorporating small amounts of narrow band gap units (dopants), such as 2,1,3-benzothiadiazole, 4,7-di-2-thienyl-2,1,3-benzothiadiazole, 2,1,3-benzoselenadiazole, and so forth, into the PF (hosts) main chain, the PF derivatives' device performance can be greatly improved due to charge trapping and energy transfer mechanisms. By this way, polyfluorene-based high-efficiency green-, red-, and blue-emitting (RGB) polymers have been reported.[98−107] Besides PFs, polycarbazoles and polydibenzosiloles are also used as hosts for high efficiency dopant−host light-emitting polymers.[108,109] Later on, phosphorescent complexes were introduced to polymers' main chain or side chain as dopants to further enhance EL efficiency.[110−114] One obvious advantage of dopant−host copolymer design is that the emission color and device performance can be easily tuned by choosing different dopants or controlling their ratios in the blend, without complicated synthesis procedure. Such fine-tuning could result in ideal white emission.[115] Therefore, a large amount of white emission polymers were developed for potential applications in solid-state lighting.[116]

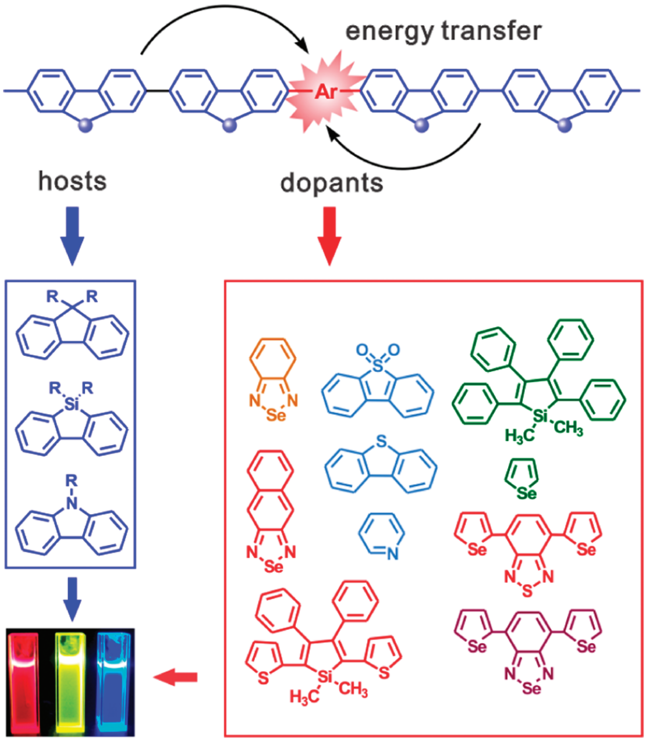

Figure 7. Schematic structures of RGB emitting polymers based on dopant-host strategy.

Light-emitting polymers can adopt similar strategies described in Sections 2.2 and 2.3 to realize multilayer device structure. The first approach is to develop water/alcohol soluble light-emitting polymers (Figure 8).[13,117,118] Combining charge transport materials that are solution processed with common organic solvents, multilayer devices can then be fabricated. Amino-/ammonium-functionalized fluorene-based water/alcohol soluble polymers developed by Huang et al[32,119,120] were successfully utilized as EML layer in PLEDs. The emission of the polymers could be tuned by doping different narrow band gap monomers in conjugated main chains. Moreover, it was found that these polymers displayed good devices performance with high work function metal Al as a cathode. For example, the green-emitting polymer exhibits the highest QE of 3.24% with a luminance of 494 cd m^{-2} at a current density of 13 mA cm^{-2} in the device configuration ITO/PVK/polymer/Al. This phenomenon indicates that these polymers

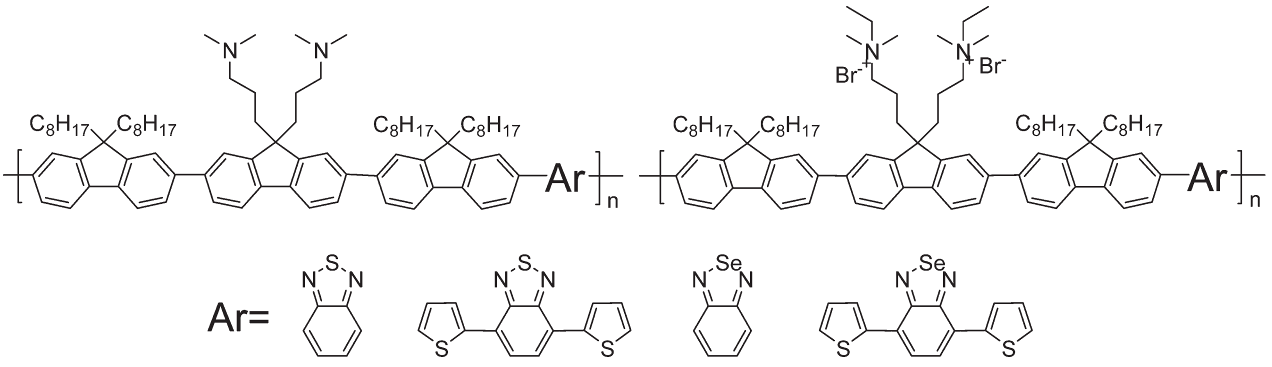

Figure 8. Examples of water/alcohol soluble light emitting polymers.

have the ability to improve the electron injection from high work-function metal cathodes as described in Section 2.3.

The second approach is to develop thermo- or photo-cross-linking light-emitting polymers. For example, Meerholz's group reported a series of photo-cross-linkable materials as EML in OLEDs (Figure 9).[121,122] The OLEDs fabricated from the cross-linked blue-emitting polymer and red-emitting polymer exhibited slightly improved efficiency compared to their non-cross-linked references with a highest LE of 3.0 and 1.0 cd/A, respectively. Cross-linkable phosphorescent orange-emitting dopant and host materials were later developed. A highest luminance efficiency of 18.4 cd/A was achieved at an operating voltage of 5 V and a brightness of 100 cd/m^2 (15 cd/A at 1000 cd/m^2).[123] It should be noted that photoinitiators, which are introduced during the photo-cross-linking process, will yield radical cations and potentially quench electroluminescence. Therefore, developing novel photo-cross-linking routes that use less or even no photoinitiator can greatly improve the efficiency and lifetime of photo-cross-linking EML materials. An example would be cationic ring-opening polymerization (CROP), which is a photoinitiator-free cross-linking process developed by Köhnen et al.[122] PLED devices prepared by CROP displayed similar performance compared to conventional photo-cross-linking EML devices, while the lifetime is improved by a factor of 2 (increased from 500 h to >1000 h at 100 cd/m^2 for blue emitters) to 3 (increased from 2000 to 6200 h at 100 cd/m^2 for red emitters).

4.2. Light-Emitting Dendrimers and Solution Processable Small Molecules.

Dendrimers can also be solution-processed due to their good solubility and appropriate viscosity.[6,7] In contrast to linear polymers, dendrimers typically comprised a well-defined branched architecture including a core, surrounding dendrons, and surface groups (Figure 10).[7] The emissive chromophores are usually used as the core for light-emitting dendrimers, while surface group functionalized charge transport units are used as the dendrons to improve the charge transporting ability and the solubility of the dendrimers. Early studies of light-emitting dendrimers focused on the fluorescent materials and established that the dendrimers with higher generation exhibited much better device performances than those first-generation dendrimers.[124] Moreover, the aggregation of the emissive chromophore cores can be effectively suppressed by the introduced dendrons to ensure good color purity. In order to develop highly efficient light-emitting dendrimers, phosphorescent complexes were used as the core in the dendrimers. The key advantage of the resulted phosphorescent dendrimers is that they can be deposited from solution without using any host materials.[125] State-of-the-art light-emitting dendrimers can reach a maximum LE of 34.7 cd/A (~20 cd/A at 1000 cd/m^2) for green emitters,[126] 9.2 cd/A (5.7 cd/A at 1000 cd/m^2) for red emitters,[127] and 5.3 cd/A (4.9 cd/A at 1000 cd/m^2) for blue emitters.[128]

Small molecular OLEDs (smOLEDs) were usually processed by vacuum evaporation,[1] or blended into polymer hosts so that they can also be processed by solution-based deposition.[129] In order to take the advantages of both the macromolecular light-emitting materials (solution processability, low cost) while keeping the key advantages of small molecular light-emitting materials (easy synthesis and purification), many efforts have been made on the development of solution processable smOLEDs. For examples, it has been reported that binuclear gallium complexes[130] or aluminum chelates[131] can be solution-processed as EML in smOLEDs. Zhu et al. reported a series of solution processable small molecular fluorescent blue and red emitters based on dendrimer-like 9,10-disubstituted anthracene derivatives[132] and end-caped 4,7-di(thiophen-2-yl)-2,1,3-benzothiadiazole derivastives,[133] respectively. Crystallization of thin film during solvent drying is suppressed by introducing the bulky dendron-like substituted groups; thus, all of these materials can form stable amorphous films by solution-processing, with excellent morphological stability and good photoluminescence efficiencies. Recently, He et al.[134] reported solution processed smOLEDs based on 4,4′-bis-[*N*-(1-naphthyl-1)-*N*-phenyl-amino]-biphenyl (NPB) and Alq_3. Although the performance is not as high as the corresponding vacuum-deposited OLEDs, it is interesting to note that, according the authors, a good thin film can be obtained from solution-processed NPB/Alq_3 blends, while NPB and Alp_3 are usually vacuum deposition so far as HTL and EML in typical OLEDs. On the other hand, by developing novel solution processable small molecular hosts, high efficiency solution-processed phosphorescent smOLEDs were also realized.[9,135,136]

There are relatively few reports on multilayer realization for dendrimers and solution processable small molecules. Son et al. successfully fabricated multilayer dendrimer thin

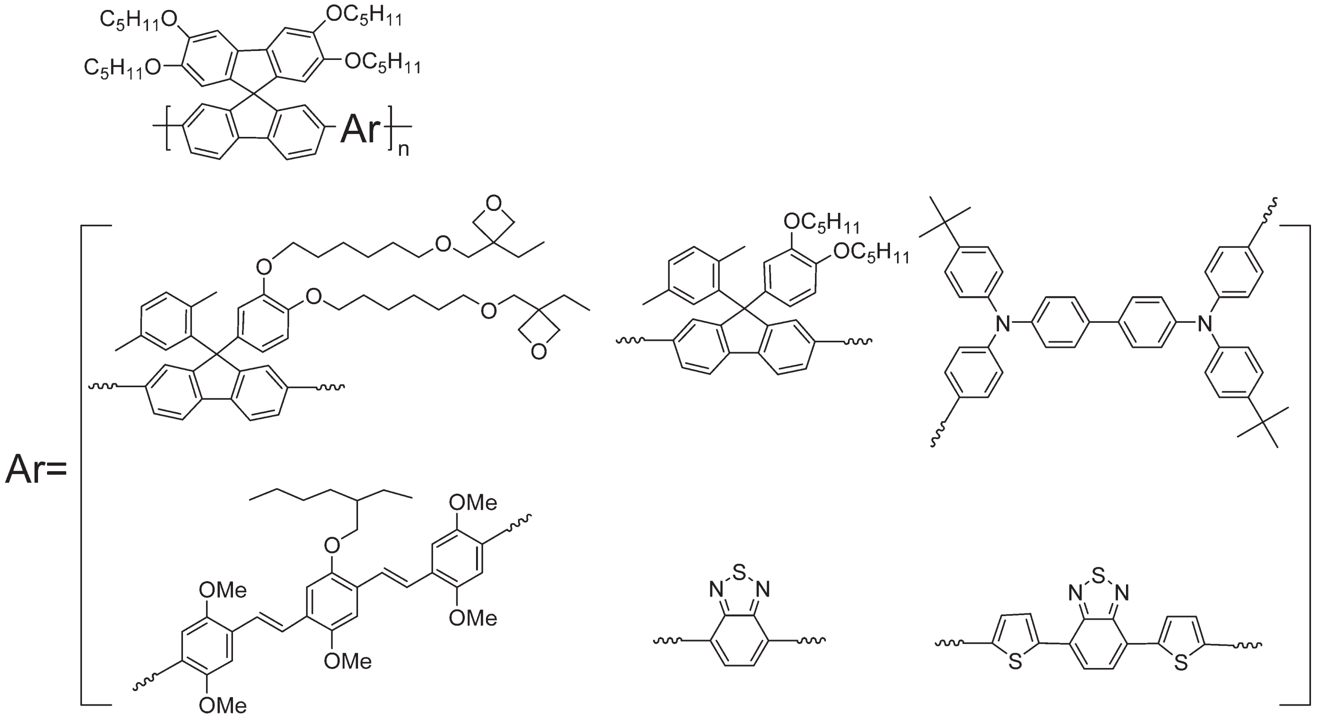

Figure 9. Examples of cross-linkable light-emitting polymers.

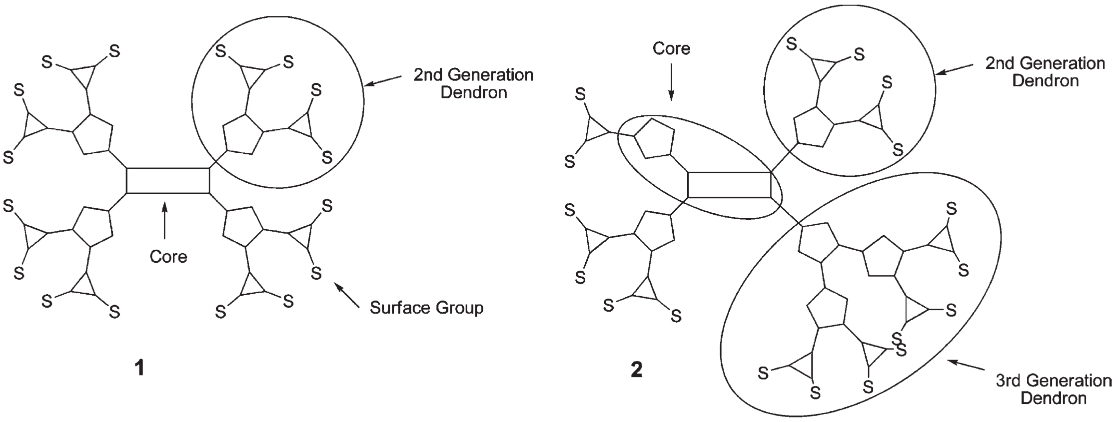

Figure 10. Schematic structures of dendrimers. Reprinted with permission from Lo and Burn.[7] Copyright 2007 American Chemical Society.

films by electrochemical deposition;[137,138] however, no complete OLED device reports are available yet in literature. Rehmann et al.[8] reported highly efficient solution-processed multilayer smOLEDs based on a low-molecular-weight host, bis(9,9′-spirobifluorene-2-yl)-phenylphosphaneoxide doped with a green phosphorescent Ir complex. By introducing a cross-linked hole-transporting double layer, the device exhibited a maximum LE of 59 cd/A at 200 cd/m^2 (55 cd/A at 1000 cd/m^2). Very recently, Yook et al.[139] reported a highly efficient solution processed blue phosphorescent smOLED design, in which the active layer with a spirobifluorene based phosphine oxide small molecule (SPPO13) as host and iridium(III) bis(2-(4,6-difluorophenyl)-pyridinato-N,C2) picolinate (FIrpic) as dopant were spin-coated by alcohol. Stacked with a chlorobenzene processed PVK:FIrpic layer, the multilayer device can reach maximum LE of 23.9 cd/A (19.3 cd/A at 1000 cd/m^2). Like in the case of He et al.,[134] SPPO13 and FIrpic were originally designed for vacuum deposition,[140] and it is remarkable that they maintained most of their performance when directly switched to the solution process without any chemical modification.

5. Fully Solution Processed OLEDs

Although significant efforts and progress have been made on the development of solution processable light-emitting materials, charge transport materials, and electrode materials, there are only a few examples that can combine these achievements together to produce real “fully” solution processed OLEDs. Since most of OLEDs were fabricated on commercial precoated and patterned ITO glass, the cathode is a major challenge in realization of a fully solution processed OLED, namely, to achieve efficient electron injection from cathode to active layer using only solution processable materials. Although Au, Ag, and Cu can be solution processed by means described above, they alone cannot give high performance OLED devices, since their work functions are high and their Fermi levels are typically 1−3 eV below the LUMO level of the active layer (depending on the specific active layer material), leading to a high electron injection barrier and hence poor electron injection. Low work function metals such as Ba, Ca, or Mg must be evaporated between cathode and active layer or coevaporated with the cathode materials in order to achieve efficient electron injection. This poses no problem for the vacuum evaporation process; however, it is not practical for the solution process, since low work-function metals are unstable in air. Therefore, the electron injection barrier problem adds new difficulties into solution processing cathode design. The newly developed water/alcohol soluble conjugated polymer ETL materials offer a useful way to resolve this problem.[13] Since it has been established that these new water/alcohol soluble conjugated polymer ETL materials can be processed from highly polar solvents such as methanol to prevent intermixing between EML and ETL layers and can efficiently enhance the electron injection from those high work-function metals to the EML materials, resulting in a comparable or even superior device performance to common Ba/Al cathode devices, it is possible to realize fully solution processed OLEDs based on these water/alcohol soluble conjugated polymer ETL materials and those solution processed high work-function metal cathodes.

Zeng et al.[15] successfully fabricated highly efficient red, green, and blue PLED devices with Ag paste cathodes and PFN as ETL (Figures 11 and 12). The properties of Ag paste, including the curing condition, the type of Ag particles, and the composition of Ag paste (Ag particle size, and content, adhesives, solvent, etc.), play an important role in optimizing the performance of PLED devices. After comparing several types of Ag pastes including commercially available ones, the authors concluded that their homemade formula type 262 was most suitable for solution processed cathode fabrication, and the devices using this type of Ag paste showed comparable quantum efficiency with vacuum deposited Ba/Al or PFN/Al electrode devices (Figure 12). The difference among various types of Ag paste is mainly caused by the different Ag particle distribution in the cathode layer. The Ag particles were most concentrated near the cathode/PFN interface for type 262 cathodes. Therefore, these devices exhibited lowest turn on voltage and highest quantum efficiencies. The optimized green-emitting device with configuration ITO/PEDOT:PSS/P-PPV/Ag paste showed a maximum LE of 7.8 cd/A (∼5 cd/A at 1000 cd/m^2) and a maximum brightness above 10000 cd/m^2 at 12.5 V. Similar excellent device performances based on PFN/Ag paste cathodes were also observed among PLEDs with blue-emitting or red-emitting polymer EMLs. To the best of our knowledge, this is the first and only successful attempt in literature to incorporate solution-processed cathodes into actual OLED devices.

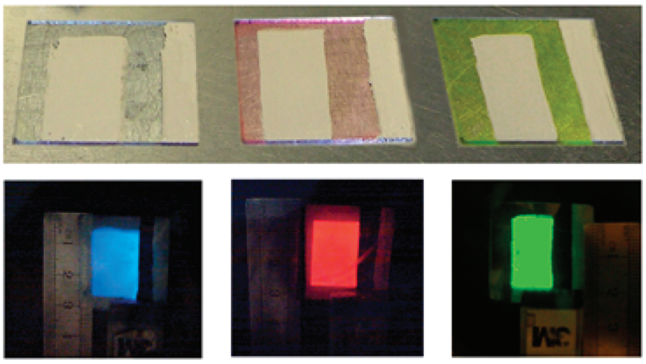

Figure 11. Photographs of RGB PLED devices (3 cm × 3 cm active area) fabricated with a printed Ag-paste cathode. Reprinted with permission from Zeng et al.[15] Copyright 2007 John Wiley & Sons.

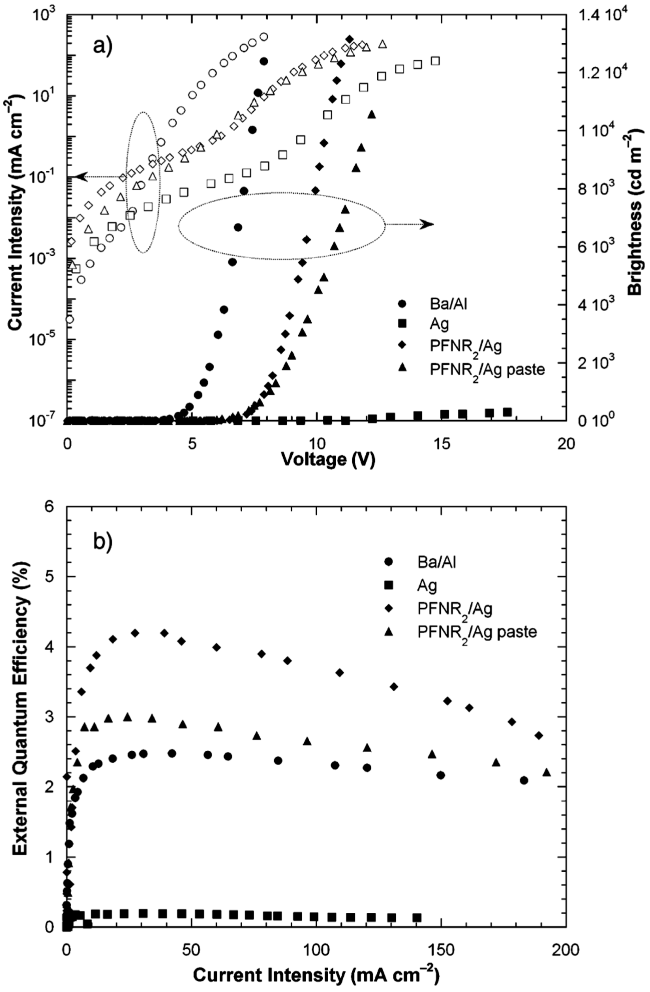

Figure 12. (a) *J*−*L*−*V* and (b) *QE*−*J* characteristic of P-PPV devices with Ba/Al, Ag, PFN/Ag, and PFN/Ag-paste cathodes. Reprinted with permission from Zeng et al.[15] Copyright 2007 John Wiley & Sons.

6. Summary and Outlooks

The solution processability of organic semiconductors endow them great potential for realizing low cost, large area electronic devices by spin-coating or inkjet printing, which is the key advantage of organic electronic devices (such as OLEDs) compared to traditional inorganic semiconductor based electronic devices. To develop solution processed OLEDs, numerous efforts have been made on developing solution processed light-emitting and charge transporting materials, resulting in a large variety of solution processed organic semiconductors, including polymers, dendrimers, and small molecular semiconductors. Polymer and dendrimer semiconductors are promising candidates for solution processed electronic devices due to their macromolecular nature, while recent interesting results have demonstrated that small molecule semiconductors also can form good films via solution process under proper conditions. Besides the light-emitting and charge transporting materials, extensive researching efforts have resulted in the development various high conductivity solution processed anodes and cathodes, which offer versatile ways toward real "fully" solution processable OLEDs.

One of the main challenges why polymer-based OLEDs is less efficient than vacuum deposited small molecule OLEDs is the difficulty in building multilayered devices using solely a solution process, since the processing of a new layer will inevitably affect the underlying layer. Multilayered design is critical to further improve the efficiency of all solution processed OLEDs to the level of state-of the-art vacuum evaporated OLEDs. To address this, both cross-linkable and water/alcohol soluble semiconductors are developed, which offer useful ways for efficient multilayer engineering of solution processed OLEDs. Most importantly, breakthroughs have been made by using water/alcohol soluble semiconductors as ETL in solution processed OLEDs that allow the use of stable high work-function metal cathodes (such as Ag, Au, etc.) for high performance devices. Finally, by using the water/alcohol soluble semiconductors as ETL and Ag paste as cathode, the first fully solution processed OLEDs were realized, which exhibited excellent device performance comparable to the reference devices with vacuum deposited Ba/Al cathodes.

Besides developing new solution processable materials for high efficiency OLEDs, the device fabrication process by varieties of printing techniques deserve much attention. New problems arise when lab results are to be reproduced in industrial scale processes. For example, the size of the droplet and drying condition during inkjet printing will dramatically influence the uniformity and EL efficiency of the printed thin films. Thus, it is a challenge to maintain the high efficiency when using inkjet printing technology.[86] Also, the wettability of the surface must be improved if the solution process is to be carried on flexible substrates.[141] Therefore, for all the new materials and designs covered in this review to be put into practice in a real large scale roll-to-roll process, much work still has to be done in the future.

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