Recent Developments in Flexible Organic Light-Emitting Devices

Yue-Feng Liu, Jing Feng, Yan-Gang Bi, Da Yin, and Hong-Bo Sun**

As the demand for display technology in consumer electronics and lighting panels increases, thin, light, high-quality, and more cost-effective light-emitting devices are required. Organic light-emitting devices (OLEDs), satisfying the criteria exactly, have been considered the most promising next-generation display and lighting technique. In particular, an OLED based on flexible substrate enables the device to be applied to curved displays, electronic newspapers, wearable displays, and conceptual lighting panels, has been always in the spotlight both in science and industry. Great advances on flexible OLEDs (FOLEDs) have been made over the past three decades. The fundamental elements of FOLEDs including substrates, electrodes, fabrication and encapsulation techniques, as well as the strategies of efficiency improvement are discussed herein. Moreover, emerging electrodes such as graphene, carbon nanotubes, metal nanowire network and their composite, flexible perovskite light emitting devices, and stretchable light emitting devices are also considered. Finally, the future challenges and prospects for these devices are put forward.

1. Introduction

The inherent advantages such as lightweight, high brightness, wide viewing angle, low power consumption, comprehensive color region, fast response time, and flexibility have attracted much more attention on organic light-emitting devices (OLEDs).[1–6] A large amount of great advances have been made in both scientific researches and commercial applications over the past three decades.[7–19] In particular, an OLED based on flexible substrate enables the device to be bent, rolled, folded, or even stretched, is pretty exciting as it opens up a fascinating world of possibilities: curved displays, electronic newspapers, wearable displays, and conceptual lighting panels. In addition, besides the excellent designs, a flexible OLED (FOLED)[7,20–40] has several other advantages, the displays and lighting panels are thinner, lighter, more cost effective, shatterproof, and durable compared to glass or silicon based OLEDs. They are impact resistance and less prone to break than glass. At present, both flexible displays and lighting panels are being mass produced (Samsung and LG on displays, LG and Konica Minolta on lighting). FOLEDs are becoming most promising and popular next-generation display technology in consumer electronics and lighting panels.

In order to develop the FOLEDs and realize their practical application, many great efforts have been conducted. The key components of the FOLEDs are flexible substrate, bottom and top electrode, organic functional layers, encapsulation layer, and optional light extraction layers. Differed from conventional OLEDs on rigid glass or silicon substrate, FOLEDs are fabricated on flexible substrates. Up to now, metal foil, flexible glass, and plastic film have been commonly used as flexible substrates for FOLEDs. On the other hand, actual fabric materials, natural silk fibroin films, bacterial cellulose, and rubbery poly (urethane acrylate) have also been developed to achieve the requirements of wearable and stretchable displays. The electrode is also very important for FOLEDs. Compared with the top electrode, more research has been conducted on the bottom electrode because its surface roughness, conductivity, and transmittance for bottom emitting OLEDs play a key role in the performance of FOLEDs. As conventional indium-tin-oxide (ITO) is not suitable for flexible devices as it is brittle, many great alternatives such as thin metal film, conducting polymer, dielectric–metal–dielectric (DMD) multilayers, metal nanowires, graphene, carbon nanotubes (CNTs), and their compound have been studied. It should be mentioned that the performance of organic layers has almost no difference with rigid OLEDs because of their inherent excellent ductility and identical working mechanism. Moreover, for practical and commercial applications, stability and efficiency are two factors of crucial importance. As a consequence, the encapsulation technique and light extraction of FOLEDs are also two research hotspots in recent years. This review will summarize the key components, discuss the method of implementation, highlight the recent research progresses, and conclude the challenges and prospects of FOLEDs.

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2. Fundamental Elements of FOLEDs

2.1. Flexible Substrates

As the foundation of the FOLEDs, flexible substrate demands good flexibility, smooth surface, and thermal and chemical stability. Metal foil,[41] flexible glass,[42] and plastic film[31,43–47] have been used as flexible substrates for FOLEDs. Metal foil and flexible ultrathin glass both have excellent capability of preventing moisture and oxygen permeation, adequate thermal and chemical stability, but the metal foil is lack of robustness of repeated bending and requires additional planarization and insulation steps, while the flexible ultrathin glass is brittle at large bending curvature. As a result, polyethylene terephthalate (PET)[39,43] and colorless polyimide[45,48] as an instance of plastic films have been most frequently used flexible substrate for FOLEDs due to their lightweight, thin thickness, high flexibility, and robustness. However, low barrier ability of moisture and oxygen, insufficient thermal and chemical stability of plastic films compelled them to combine with some inorganic materials as an instance of alumina (Al_2O_3).[49–51]

In recent years, several novel flexible substrates have been reported as shown in **Figure 1**. In order to improve the comfort level, sewability, and compatibility with clothes of wearable displays, actual fabric materials were developed.[52–54] Because a single fiber of off-the-shelf clothes is tens of micrometers and the surface roughness resulting from fabric weave construction is even larger, the planarization layer has to be used and then traditional FOLEDs could be fabricated on it. On the other hand, biodegradable and biocompatible natural silk fibroin films were also reported. Compared to typical plastic polymer and fabric clothes, the silk fibroin protein[55] as the raw materials can be easily obtained from the natural silkworm cocoons, which is more environmentally friendly. Bacterial cellulose[56,57] is also an environmentally friendly, renewable, and biodegradable nanocomposite. Moreover, good transparency in the visible region (up to 90%), excellent thermal stability, and mechanical property make them potential candidate materials of transparent substrate for FOLEDs. As increasing requirements of FOLEDs for application, flexible substrates need to be not just bending but stretching. Therefore, rubbery poly (urethane acrylate) (PUA),[21,58] etc., with the high transmittance and excellent stretchability were also used as stretchable substrate.

2.2. Electrodes of Flexible OLEDs

Flexible electrode is another key component in FOLEDs.[59–64] In the conventional OLEDs, ITO is the most frequently used electrode owing to its high transmittance and conductivity.[32,65–68] However, it is not an ideal choice for flexible OLEDs due to its poor mechanical robustness and fabrication incompatible with the plastic substrate because of the high-temperature deposition process. Therefore, thin metal film, conducting polymer, and DMD multilayers have emerged as leading candidate substitutes of the ITO electrode.

A continuous and thin metal film with high electric conductivity and mechanical robustness has been regarded as the most ideal alternative of the ITO in the early research.[29,61,62,69] Especially, it can be simply deposited by vacuum thermal evaporation and has excellent compatibility with the most organic materials. Due to superior ductility and conductivity, silver (Ag) and gold (Au) were developed more than other metal. Poly (3, 4-ethylenedioxythiophene):poly (styrenesulfonate)

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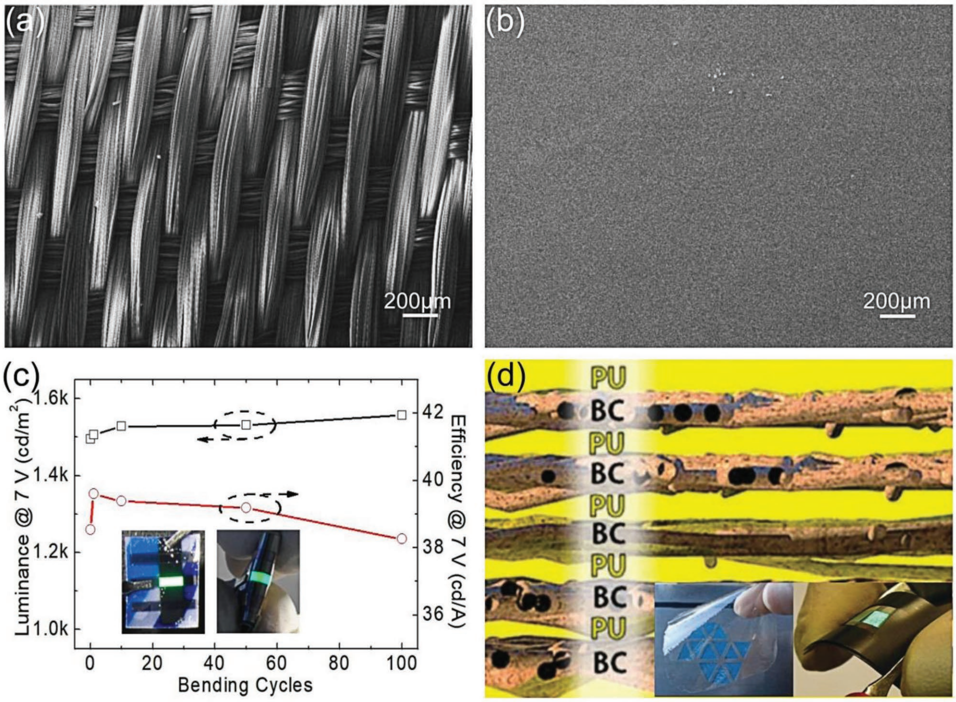

Figure 1. SEM images of silk substrate a) before and b) after planarization with photopolymer. c) Luminance and efficiency at 7 V in condition of the different number of bending cycles. Inset: photographs of FOLEDs on the planarized silk substrate before and after bending. d) Cross-section view and transparency exhibition (left inset) of the bacterial cellulose and castor oil based polyurethane composite substrate with the construction of alternating layers of bacterial cellulose nanofibers filled with polyurethane and a FOLED (right inset) on it. (a–c) Reproduced with permission.[52] Copyright 2017 IEEE. (d) Reproduced with permission.[57] Copyright 2015 The Royal Society of Chemistry.

(PEDOT:PSS)[59,70] as a representative conducting polymer has been proposed the most promising material for cost-effective FOLEDs, which is compatible with mass production roll-to-roll process. The superiorities of PEDOT:PSS are high optic transmittance, excellent flexibility, and high work function, while its key drawbacks are insufficient conductivity and acidity which are adverse to the performance of FOLEDs. DMD electrode[71–77] is a type of multilayer structure consisting of a metal film between two high refractive index dielectric layers. It exhibits low sheet resistance and high flexibility due to the presence of thin metal film. In addition, the high refractive index dielectric layers enable to improve transmittance by reducing the reflection of the metal film ascribed to destructive interference. Up to now, several various structures of multilayer electrodes applied to FOLEDs have been reported. For instance, ZnS/Ag/ZnS,[78] ZnS/Ag/WO_3,[71] ZnS/Ag/MoO_3,[72] MoO_3/Ag/MoO_3,[73] WO_3/Ag/MoO_3,[79] Cs_2CO_3/Ag/ZnS,[80] InZnSnOx/Ag/InZnSnOx,[77] etc. Kim et al. have demonstrated a highly transparent FOLED which employed DMD anode as well as cathode.[76] The details of anode and cathode are [ZnS (24 nm)/Ag (7 nm)/MoO_3 (5 nm)] and [ZnS (3 nm)/Cs_2CO_3 (1 nm)/Ag (8 nm)/ZnS (22 nm)], respectively. **Figure 2** shows the photographs of flexible OLEDs with bending in the on and off states. Highly transparent and flexibility could be obviously observed. Moreover, metal/insulator/metal (MIM) structure has been used to replace the metal film in the DMD structure to improve the transmittance of DMD electrode furtherly. This is because destructive interference of the reflective wave cannot suppress the reflection thoroughly when the thickness of metal increases for higher conductivity. While, this problem could be solved by more effective interference deriving from resonance in the double metal layers systems of MIM structure.[74,75] It is notable that the sheet resistance of DMD electrode both thin metal layer and MIM structure between dielectric layers, after 1000 bending repetitions, remained at the same level as before bending. It proves the excellent mechanical flexibility and stability of DMD electrode as well as the metal electrode.

Recently, several kinds of emerging electrodes such as graphene, CNTs, metal nanowires, and their compounds have been researched and reported. Graphene[7,81–86] is an excellent choice as a flexible anode owing to its high transparency, high conductivity, high elasticity, low cost, and chemical stability. There are various methods to fabricate graphene thin film such as mechanical exfoliation, epitaxial growth, reduce functionalized graphene, and chemical vapor deposition (CVD).[87,88] Among them, mechanical exfoliation and epitaxial growth are not suitable for low-cost and large-area applications. Although reduced functionalized graphene is usually fabricated by solution-process,[7] which the graphene electrode was deposited on substrates by spin coating of an aqueous dispersive functionalized graphene followed by a vacuum annealing process to reduce the sheet resistance, it is not compatible with plastic flexible substrate owing to high temperature anneal. Therefore, chemical vapor deposition has been developed more. Monolayer graphene anode on a flexible PET substrate achieved using CVD has been demonstrated by Li et al. By using single layer graphene as a

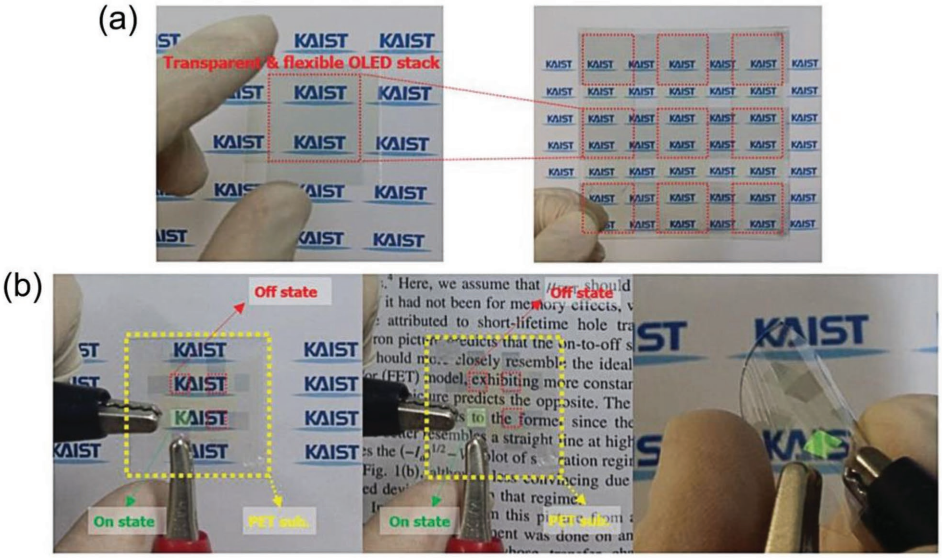

Figure 2. a) Transparent FOLEDs with 2.5 × 2.5 cm^2 area and enlarged view. b) The photographs of transparent FOLEDs in condition of on and off with letters as background (left and middle) and bending (right). Reproduced with permission.[76] Copyright 2015, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

transparent electrode, green and white FOLEDs with current efficiencies greater than 80 and 45 cd A^{-1} are realized.[84]

As another alternative anode for FOLEDs, CNTs have obvious advantage and disadvantage.[30,89–91] CNT network film is shown to have high work function, excellent electrical conductivity, and mechanical flexibility. In terms of depositing methods, CNTs could be fabricated on plastic flexible substrate by dip coating, spraying coating, meyer rod coating, polydimethylsiloxane (PDMS) stamp-based transfer printing (**Figure 3**a), etc.[30] However, the relatively high sheet resistance and rough surface morphology of CNTs

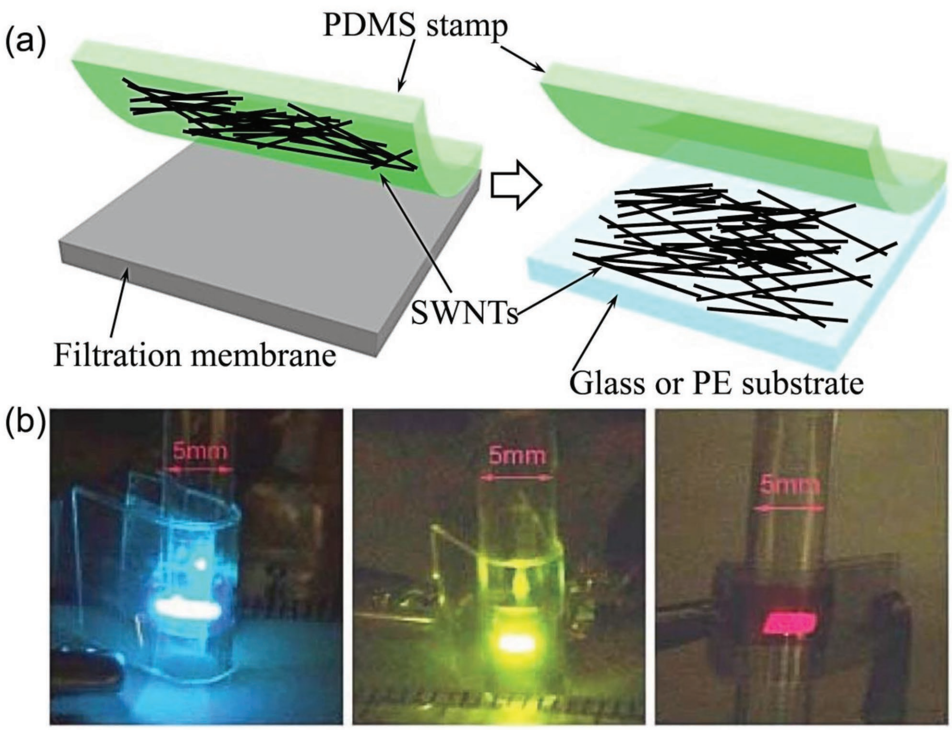

Figure 3. a) Illustration of the dry transfer process of SWNT electrode: the SWNT film is peeled off from the filtration membrane using a PDMS stamp and subsequently printed onto a rigid or flexible substrate. b) Photographs of blue, yellow, and red operating polymer light-emitting devices at 10 V, which are bent around a glass tube with 5 mm diameter. (a) Reproduced with permission.[30] Copyright 2006, American Chemical Society. (b) Reproduced with permission.[90] Copyright 2011, SPIE.

networks are two major issues.[91] In spite of high conductivity of individual CNT, the network CNTs thin film has mediocre conductivity because of the barrier between the different CNTs. Another challenge to CNTs anode is poor surface morphology and their surface roughness is typically about 10 nm arising from high aspect ratios of CNTs. Although this rough surface roughness is probably modified by spin-coating holes injection or transport materials such as PEDOT:PSS, the hydrophobic nature of CNTs causes the physical interface contact between the CNTs network and the adjacent PEDOT:PSS layer is doubtless poor. Even then, FOLEDs with CNTs anode have already reported by several research groups. As shown in Figure 3b,[90] flexible blue, yellow, and red polymer light emitting devices with CNTs as both anode and cathode have been demonstrated. The devices exhibit a low turn-on voltage, modestly high efficiency, and mechanical flexibility. No failure was observed after repeated bending to a 2.5 mm radius and unbending for 50 cycles. However, perhaps due to the aforementioned limits, CNT anode for FOLEDs is being slowed down a little bit compared to other emerging anodes.

Metal nanowire network electrode, particular silver nanowires (AgNWs) as a solution processable material have been developed for FOLEDs because of their superior optical, electrical, and mechanical properties.[21,45,48,55,92–96] Excellent sheet resistance and transparency can be obtained via the use of high aspect ratio AgNWs. Further, post-treatment processes, such as high temperature annealing (commonly higher than 180 °C), plasmonic welding, and high-force pressing are effective for reducing the resistance of AgNWs network by fusing the junctions of AgNWs because the sheet resistance of the AgNWs percolation network is dominated by the contact resistance of inter-AgNWs owing to the inherent high conductivity of silver.[21] Also because of the presence of junctions, the roughness of AgNWs percolation network is at least tens of nanometers.[45,93] It is the most challenging issue to apply the AgNWs to FOLEDs. We will discuss the approach of improving the surface morphology of AgNWs electrode in Section 3.1.

In order to improve the performance of electrodes for FOLEDs, compound electrodes with different abovementioned materials have been revealed because compound electrodes not only could maintain the advantages but also eliminate the disadvantages of individual electrode materials. Most of these compound electrodes are two species including laminated electrode and hybrid electrode. The laminated compound electrodes including AgNWs/ITO,[97] AgNWs/Graphene,[98] AgNWs/TiO_2,[45] AgNWs/ZnO:Al,[99] Al/multi-walled CNTs (MWCNTs)/Al,[100] Graphene/Ag thin film,[101] Graphene/Ag/ZnO:Al,[102] Ag thin film/PEDOT:PSS,[22] etc., the hybrid compound electrodes including PEDOT:PSS and single-walled CNTs (SWCNTs),[103] PEDOT:PSS and AgNWs,[104] AgNWs and SWCNTs[105] are respectively researched. **Figure 4**a,b[98]

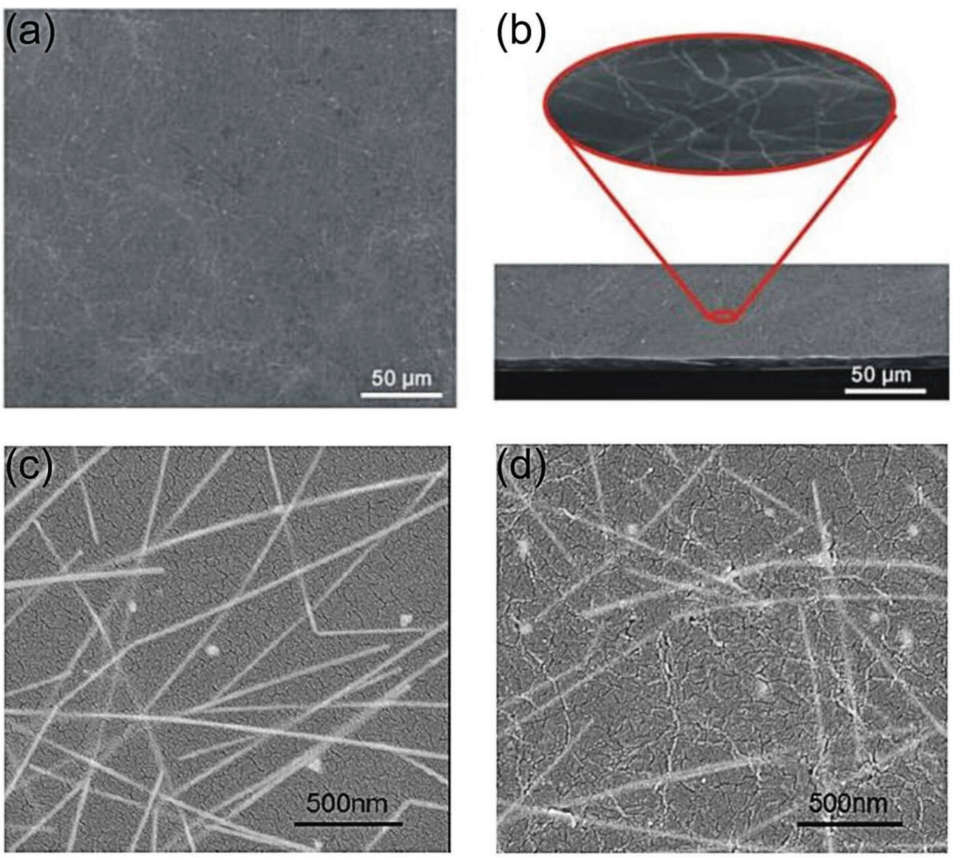

Figure 4. a) SEM top view image and b) cross-section image of laminated compound electrode with monolayer graphene and AgNWs embedded in polymer film (3.75 mg mL^{-1}). SEM image of hybrid compound electrode of c) an AgNW-nanoparticles film and d) an SWNT/AgNW nanoparticles film. (a,b) Reproduced with permission.[98] Copyright 2016, American Chemical Society. (c,d) Reproduced with permission.[105] Copyright 2014, Springer Nature.

has shown a laminated electrode which is the AgNWs network covered by a monolayer graphene. Graphene could provide an effectively protective screen for AgNWs and improve the extra electron pathway for the compound film. This unique electrode exhibits the superior optoelectronic characteristics (sheet resistance of 8.06 Ω □$^{-1}$ at 88.3% light transmittance). As shown in Figure 4c,d,[105] it is a typical hybrid electrode comprising AgNWs, single walled carbon nanotubes (SWNTs), and nanoparticles of barium strontium titanate. Combined with the excellent conductivity of AgNWs and SWNTs and the light scattering of nanoparticles, the FOLEDs with this hybrid electrode exhibit a maximum current efficiency of 118 cd A^{-1} at 10 000 cd m^{-2} with the calculated external quantum efficiency being 38.9%. As the improvement of materials and fabrication techniques, the compound electrodes will be certain to keep ongoing evolution and progress.

2.3. Fabrication Technologies of Flexible OLEDs

In the traditional process, small molecule based OLEDs and polymer based polymer light emitting devices are fabricated by vacuum thermal evaporation and spin-coating, respectively. At present, these two technology processes are also most commonly used for both laboratory research and industrial production. In terms of FOLEDs, the processes are identical with conventional rigid OLEDs when the devices are fabricated by thermal evaporation or spin-coating. It is notable that the detachment transfer approach[106,107] of FOLEDs from the rigid substrate to the flexible substrate is also fabricated by these two technology processes. Therefore, we do not repeat them in detail, while just introducing some processes for FOLEDs. With goal for low cost and high throughput fabrication of FOLEDs, it comes as no surprise with numerous scientific and company focus on roll-to-roll coating and printing techniques, such as inkjet printing, gravure printing, slot-die coating, and screen printing (**Figure 5**).[108–111] Inkjet printing[111–113] enables thin film deposition and patterning simultaneously and avoids any chemical processes which degrade the performance of organic materials beneath. The small amounts of functional materials with almost any low-viscosity liquid can be deposited without contact with the substrate. Therefore, very thin films can be fabricated and it is not sensitive to substrate defects. However, the inkjet printing technology also has some intrinsic limits, such as the roughness of printed film generated by joined drops due to the technical difficulty to keep the nozzle clear and the degradation in thickness uniformity because of drying processes.[112] Gravure printing[109,114] is a cost-effective and high throughput roll-to-roll printing technology. Combined with low temperature processing, gravure printing has become one of the most favourable methods for the quantity production of organic films. On the other hand, highly efficient OLEDs fabricated by screen printing have been also demonstrated.[108,115] The ink is transferred using a squeegee through the open networks onto the substrate by shifting the squeegee and touching the substrate momentarily. It allows the pattern to easily define which parts of the substrate receive deposition. Unfortunately, both gravure printing and screen printing usually require high ink viscosity, which makes these two printing techniques still challenging for

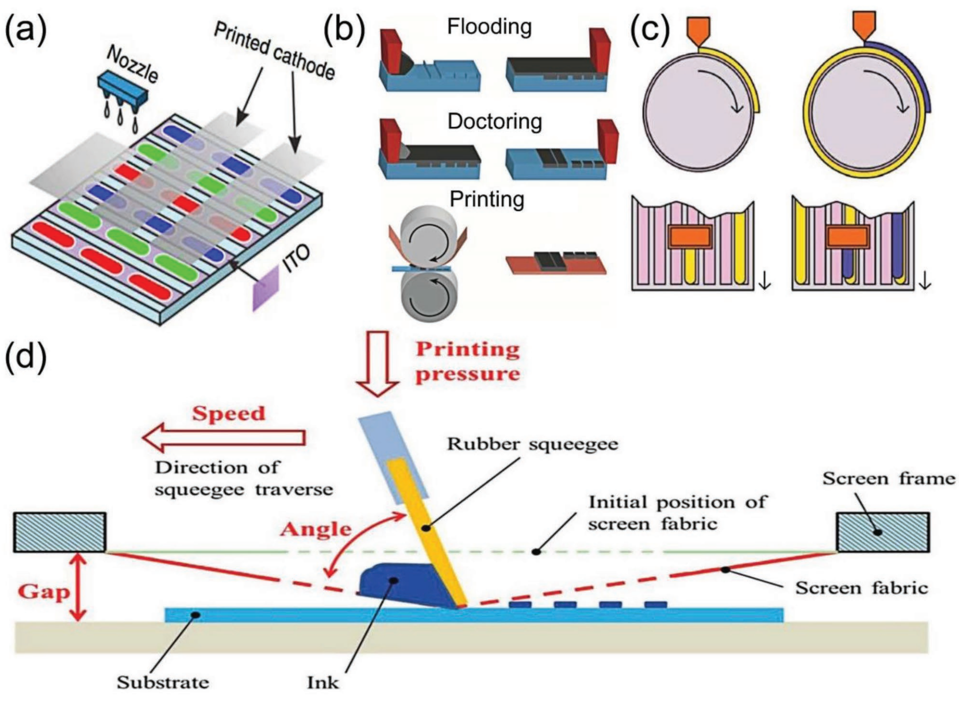

Figure 5. a) Schematic illustration of the full-color display with cathode using ink print, b) the gravure printing process decomposed into three steps, c) the slot-die roll coating of the (yellow) active layer and the (blue) semitransparent anode on top of a (pink) flexible cathode-coated substrate, the ink is transferred from an external container via a pump to the slot-die head (orange), and d) screen printing process. (a) Reproduced with permission.[111] Copyright 2013, Springer Nature. (b) Reproduced with permission.[109] Copyright 2014, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (c) Reproduced with permission.[110] Copyright 2012, Springer Nature. (d) Reproduced with permission.[108] Copyright 2018, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

practical application of FOLEDs. On one hand, the high viscosity of the inks will obviously give rise to thick film while FOLEDs need thin organic functional layers to make the carriers transport effectively. On the other hand, additives have to be used to increase the viscosity of the ink because of the low viscosity nature of the functional inks. It is adverse to keep the purity of the organic layer, and will influence the performance of FOLEDs consequently. In regard of slot die coating[110] with the good homogeneity of coated films, an ink over a wide range of viscosity is allowed directly to apply through a pump feeding slot orifice to a grid supported by a backup roll. The adjustment of the pump pressure is an effective method to control the operating range of the viscosity due to the upper limit of the viscosity depending on the pump. It should be mentioned that the slot die coating has an obvious shortage which is the incompatibility with patterning.

2.4. Encapsulation of Flexible OLEDs

The lifetime of FOLEDs is very important factor for their commercial application. It is relative to the degradation of OLEDs, such as some undesirable internal processes such as chemical reactions, morphological (phase changes, crystallization, and delamination processes), and other physical (e.g., charge accumulation) changes. Moreover, it is well known that organic materials are very sensitive to moisture and oxygen so that the OLEDs are easily degraded and shortened the lifetime in the ambient atmosphere.[13,116,117] But the commercial requirement of FOLEDs display for lifetimes should exceed 10 000 h, so the encapsulation of FOLEDs plays the most important role for the lifetime of FOLEDs in the practical application. In the conventional OLEDs, they are commonly encapsulated by hollow rigid glass because of its impermeability and reliability. But it could not be bent, giving rise to not suitable for use in FOLEDs. To keep the high flexibility of FOLEDs, flexible encapsulation process should be developed as well as flexible substrates and electrodes. As the degradation mechanisms of vapor permeation and the measurement of permeation rates of OLEDs have been discussed in detail in previous review article,[116,118] no more detailed descriptions are conducted here. Up to now, the well-known approach is thin film encapsulation for FOLEDs.[49–51,118–125] A hybrid organic–inorganic alternating multilayer barrier coating has demonstrated that it is suitable for FOLEDs because of its adequate ability to protect the permeation of moisture and oxygen.[49,119] The polymeric layers make the substrate smooth so as to reduce mechanical damage and increase thermal stability of the nucleation surface, while inorganic metal oxide layers are using as thin barrier layers. Meanwhile, an atomic layer deposition (ALD) layer such as Al_2O_3 has been reported as the excellent barrier property for polymeric substrates. ALD could realize fabricating very thin, conformal film with control of the thickness and composition of the diffusion barrier film possible at the atomic level. It is very attractive to satisfy the criteria of barrier property of moisture and oxygen.[50,51,121,122,124,125] However, the drawback of both organic–inorganic alternating barrier and ALD is low production throughput. To overcome this problem, flexible lamination with metal foil and PDMS encapsulation process including treatment with a self-assembled monolayer, spin coating, and thermal cross-linking has been reported (**Figure 6**).[126] The water vapor transmission rate (WVTR) measurement proved that the flexible lamination encapsulation has the excellent

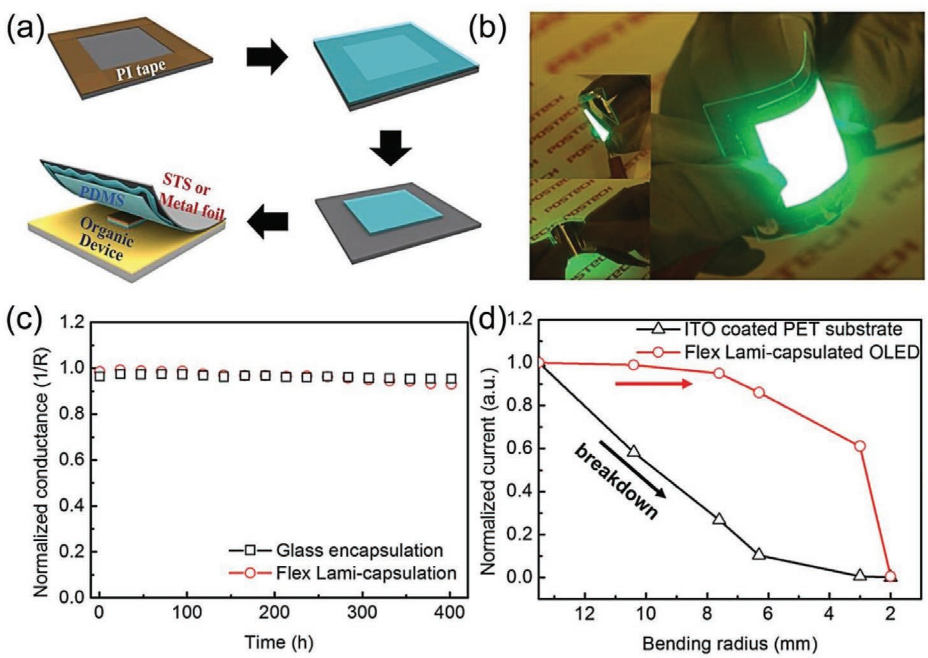

Figure 6. a) The flexible lamination encapsulation process. b) Photograph of large area FOLEDs encapsulated with 40 µm thick Fe–Ni alloy foil (Substrate area: 5 cm × 5 cm; light area: 3 cm × 3 cm). Insets: side view (upper) and top view (bottom) of FOLEDs. c) Measurement of WVTR at 25 °C, 40% relative humidity (RH) (glass encapsulation: WVTR = 5.9×10^{-4} g m^{-2} d^{-1}; flexible lamination encapsulation: WVTR = 5.5×10^{-4} g m^{-2} d^{-1}). d) Mechanical stability measurements of ITO-coated PET substrate and flexible lamination encapsulated phosphorescent OLED fabricated on ITO-coated PET substrate as a function of the bending radius. Reproduced with permission.[126] Copyright 2015, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

ability to block the moisture and oxygen. In addition, flexible lamination encapsulated FOLEDs showed good flexibility and mechanical stability. This approach is also compatible with cost-effective roll-to-roll process. However, as the metal foil is opaque, further research and development are required for the commercialization of the flexible lamination encapsulation.

3. Efficiency Improvement of Flexible Organic Light-Emitting Devices

According to the significance of high efficiency, efficiency improvement of OLEDs is a constantly pursuing goal. Although the quantum efficiency of luminescent material used in advanced OLEDs can reach approaching 100%, there are also several critical factors for the efficiency of FOLEDs, such as surface morphology of the bottom electrode, interface between the electrodes and organic layers, light extraction (efficiency enhancements have been summarized in **Table 1**), etc. The effective approaches to improve the efficiency of FOLEDs from abovementioned aspects have been discussed in this section.

3.1. Surface Morphology Manipulation of Electrodes

The surface morphology of the electrode including the roughness, continuity, and grain size is a directly acting factor for the performance of FOELDs. As abovementioned in Section 2.2, Au and Ag are widely used due to their excellent conductivity and ductility. But depositing the Au and Ag films directly on the substrates exhibits Volmer–Weber growth mode and tends to form 3D island.[34,127,128] It is unbeneficial to fabricate the smooth film and lead to additional optical loss, even causing nonconductive film if the islands could not form percolation paths. To overcome this problem, there have been tremendous efforts including surface treatment before metal deposition,[129] doping,[130] and adding seed material deposition.[131,132] A silver film with subnanometer surface roughness (0.322 nm) has been demonstrated by template stripping.[69] It is an effective and simple technique to generate ultrasmooth metallic films on flexible substrate. In the typical process of template stripping (**Figure 7**a),[62,69,70,104,133–135] substrates with ultrasmooth surface such as silicon, glass, and mica are used as master templates. Despite the evaporated metal film has a rough surface on the templates, a smooth surface at the interface between metal and template is formed and the smoothness of the opposite surface is close to that of the template. A photopolymer (e.g., NOA, SU8) as backing layer was spin-coated on the substrates and cured. Finally, the cured photopolymer film is peeled off and the flexible substrate with ultrasmooth electrode is acquired. This method exhibits a special advantage in FOLEDs because not only the electrode smoothness can be improved as shown in Figure 7b[69] (the root mean square roughness of the as-evaporated Ag and the template-stripped Ag surfaces are 1.04 and 0.322 nm, respectively), but also the supporting layer itself has good flexibility and is suitable to be used as the flexible substrate. It is notable that this method is universally available to various electrodes, just requires the electrodes have better adhesion with photopolymer compared with the template to ensure they could be peeled off along with the photopolymer. The Ag electrode and PEDOT:PSS electrode have been all fabricated by this method in our previous work. Compared with as-evaporated or spin-coated electrodes, FOLEDs with the peeled-off ultrasmooth electrode show the improved efficiency obviously and high flexibility (Figure 7c,d).[62,69] On the other hand, an ultrathin Au film (7 nm) with excellent surface morphology, high conductivity, and applicable optic transparency was obtained on SU8 flexible substrate through chemical bond interactions between the SU-8 film and Au atoms, providing more dense nucleation centers for the subsequently following Au atoms and suppressing the formation of large isolated Au islands during the deposition process, finally leading to the continuous, ultrasmooth, and ultrathin Au film (**Figure 8**).[61] When a SU-8 film was used to modify the glass surface prior to Au thermal deposition, the ultrathin Au film showed a uniform, continuous, and smooth surface morphology. The roughness of the Au film on SU-8 is about 0.35 nm, which is nearly one order of magnitude lower compared with the Au film on the glass. Similar with the effect of SU8, ZnS, tungsten trioxide (WO_3) and molybdenum trioxide (MoO_3) could be also used as seed layer materials to affect the formation of the evaporated metal film. Han et al. reported a comparative study on the surface morphologies of Ag films with ZnS, WO_3, and MoO_3.[72] The sheet resistance, transparency and surface roughness of different flexible electrodes have been summarized in **Table 2**. The better thermal stability of Ag atom on seed layer allows

Table 1. Reported enhancement in the outcoupling efficiency of FOLEDs with different light extraction technique.

Technique of light extraction	The mode of being coupled out	Efficiency enhancement	Ref. No.
	Internal nanostructures:		
Quasi-random structure	Waveguide and surface plasmon modes	76% (EQE)	[143]
Periodic corrugated structure		70% (EQE)	[156]
Periodic nanomesh		30% (EQE)	[145]
	External Nanostructures:		
Inverted conical structure	Substrate mode	160% [cd A^{-1}]	[148]
Spherical refractive microlenses		70% [cd A^{-1}]	[147]
Single-crystal AgCl nanorods		33% [cd A^{-1}]	[158]
	Nonstructural techniques:		
High-index Ta_2O_5 optical coupling layer	Waveguide mode	60% (EQE)	[162]

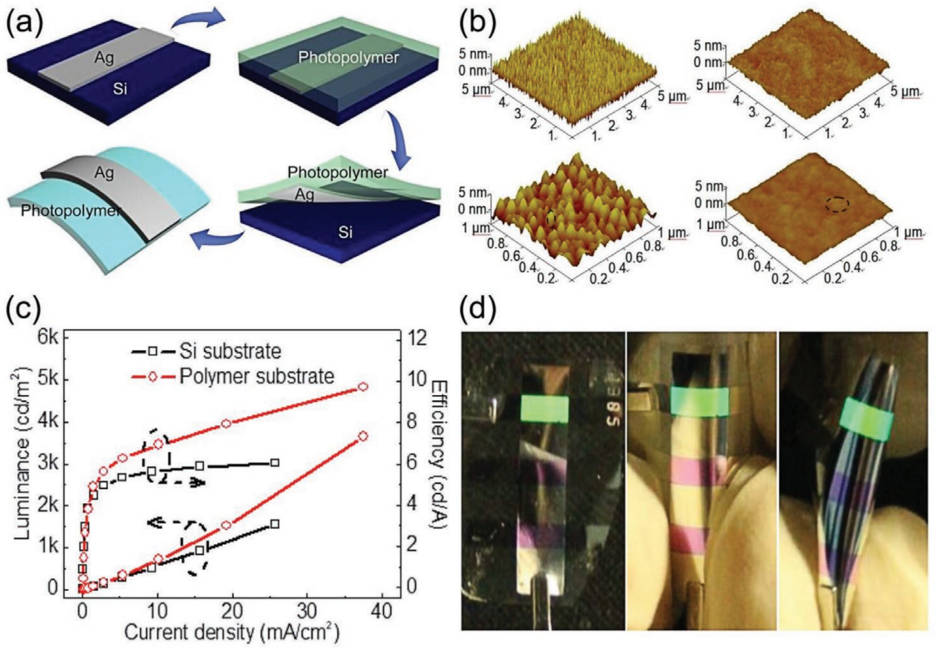

Figure 7. a) Scheme of ultrasmooth Ag film fabricated by template stripping. b) AFM images for the as-evaporated Ag film (left) and template-stripped Ag film (right), scan area: 5 mm × 5 mm (upper) and 1 mm × 1 mm (bottom). c) Luminance–current density–efficiency of flexible TOLEDs on Si and photopolymer substrates. d) Photographs of the flexible TOLEDs at different bending radius. Reproduced with permission.[69] Copyright 2013, The Royal Society of Chemistry.

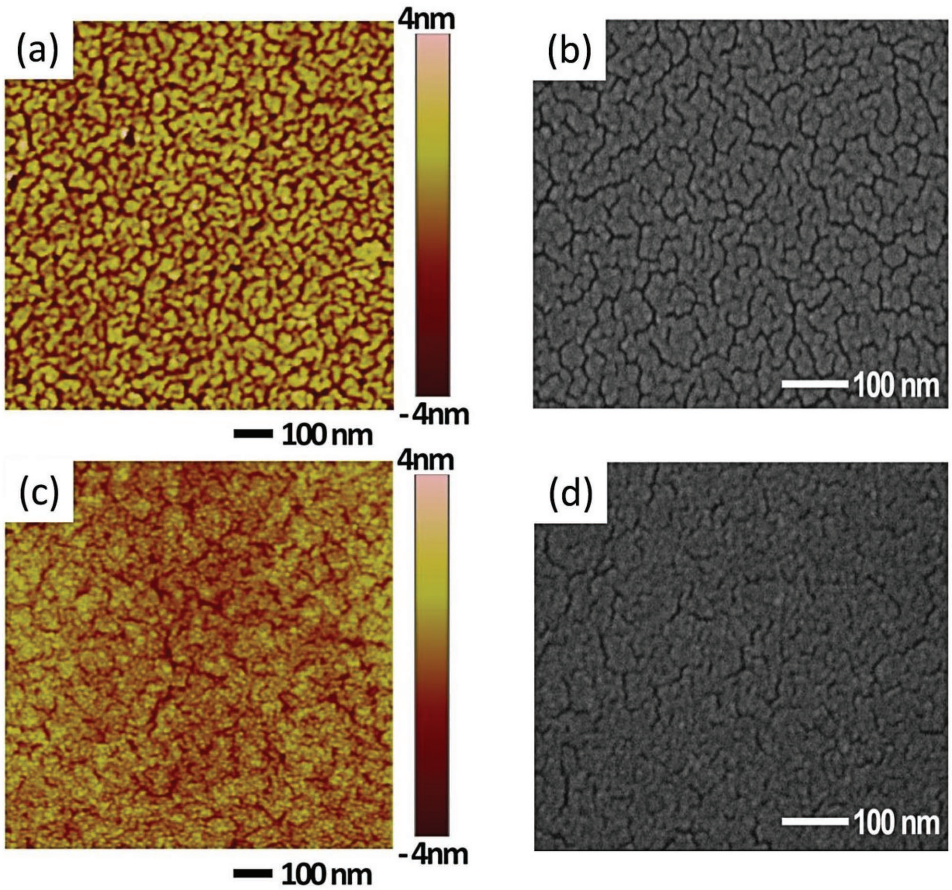

Figure 8. a,c) AFM images and b,d) SEM images of surface morphology of the 7 nm ultrathin Au film deposited a,b) on the glass substrate and c,d) on the SU-8 modified glass substrate. Reproduced with permission.[61] Copyright 2016, The Royal Society of Chemistry.

Table 2. Reported parameters of different flexible electrodes.

Transparent electrode	Sheet resistance [Ω □$^{-1}$]	Transparency [%]	Surface roughness [nm]	Work function [eV]	Ref. No.
Ag (6 nm)	<15	76 (420–680 nm)	N/A	4.26	[129]
Au (7 nm)	23.75	73 (@550 nm)	0.35	5.1	[61]
PEDOT:PSS	54.39	>90 (@500 nm)	0.468	5.2	[70]
ZnS/Ag/MoO3	9.6	83 (@550 nm)	1.5	5.3	[72]
ZnS/Ag/ZnO/Ag/WO_3	2.17	83.37 (@550 nm)	N/A	4.9	[75]
Graphene	<200	97 (400–700 nm)	0.35	5.1	[84]
CNTs	300	90 (@550 nm)	8-10	4.7-5.2	[91]
AgNWs	7.2	78 (@550 nm)	1.58	5.9	[95]

the Ag deposition to acquire a thinner continuous film. The percolation threshold first occurred on ZnS, as the Ag film increased in thickness and a quasiperfect continuous Ag film (7 nm) with a surface coverage of 99.6% and sheet resistance of 9.2 Ohm sq^{-1} was achieved on ZnS. Except for using a seed layer, the incorporation of a small quantity of Al also could suppress the 3D islands growth of Ag and facilitate smooth surface morphology and ultrathin continuous Ag film formation.[127]

To date, AgNWs are always embedded in substrates by a peeled off process to planarize the surface morphology (**Figure 9**a).[45] The surface roughness of the peeled off AgNWs electrode is lower than 1 nm, while a AgNW embedded in other transparent polymer substrates via mechanical pressing typically shows a surface roughness on the order of 10 nm. This AgNW electrode also has high mechanical stability and no loss in conduction was observed even more than 20 000 bending cycles at 1 mm bending radius. In addition, PEDOT:PSS, graphene oxide (GO), etc., anode modified materials are used on the top of AgNWs to reduce the roughness further (Figure 9b).[21] The charged and flexible GO sheets could adhere and wrap around the AgNWs and solder the inter-nanowire junctions, causing significant reduction of the inter-nanowire contact resistance without heat treatment or high-force pressing. Moreover, the surface morphology also could be improved due to without high temperature fused junctions which are adverse to obtain a smooth surface. Also, Lee et al. reported a film with dual-scale AgNWs percolation networks, which showed superior electrical and optical characteristics to demonstrate a high efficiency FOLED (**Figure 10**).[94] The secondary AgNWs networks can be formed on the voids between longer AgNWs percolation networks and fill the large fluctuation of longer AgNWs networks. It does not affect the transmittance obviously, but enhances the effective electrical area significantly.

3.2. Energy Level Modification of Electrode/Organic Interface

An electrode with good carrier injection characteristics is desired for high efficiency of FOLEDs. Numerous kinds of methods have been proposed to improve the carrier injection, such buffer layer, p-doped for anode, n-doped for cathode, p–n junction structure as an efficient and cathode independent electron injection layer, etc..[35,136–140] Generally, PEDOT:PSS and transition metal oxide for an instance of MoO_3 are used to enhance the holes injection as anode buffer layer or p-dopants, while alkali metals and alkali metal fluoride or carbonates for an instance of lithium (Li), lithium fluoride (LiF), and cesium carbonate (Cs_2CO_3) are used to enhance the electrons injection as cathode buffer layer or n-dopants. These materials all have the ability to reduce carrier injection barrier, give rise to improve the efficiency of FOLEDs. An organic p–n junction as an efficient electron injection layer for inverted FOLEDs has been also demonstrated[138]

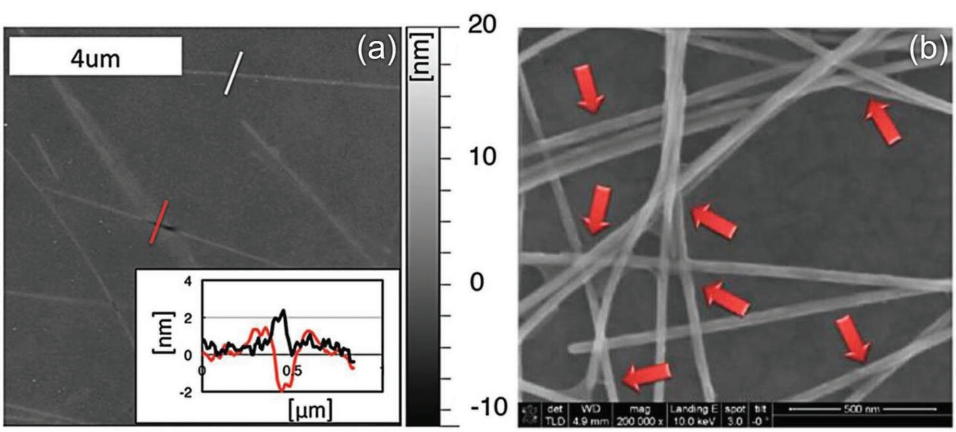

Figure 9. a) AgNWs embedded in colorless polyimide. b) GO-soldered the junctions of AgNWs network (indicated by red arrows). (a) Reproduced with permission.[45] Copyright 2015, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (b) Reproduced with permission.[21] Copyright 2014, American Chemical Society.

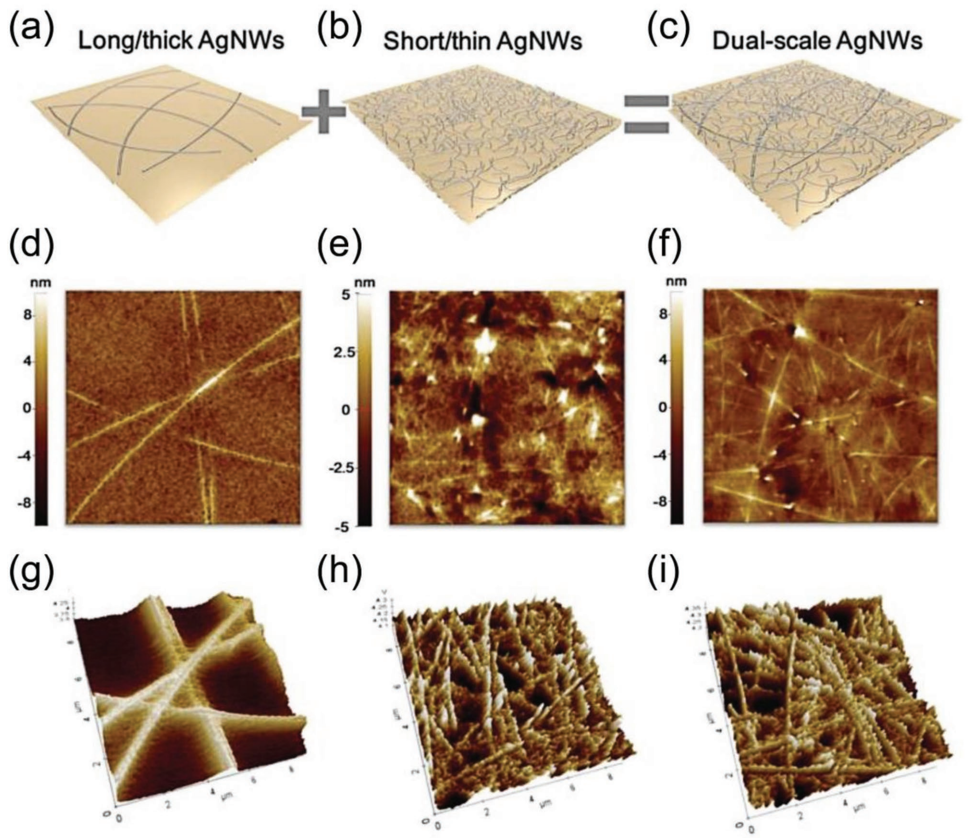

Figure 10. a–c) Schematics of the dual-scale AgNWs network transparent conductive films. d–f) AFM images and g–i) electrostatic force microscopy analysis (measured at +4 V bias from the sample side) of long/thick, short/thin, and dual-scale AgNWs. Reproduced with permission.[94] Copyright 2017, The Royal Society of Chemistry.

(**Figure 11**). The organic p–n junction composed of p-doped copper phthalocyanine (CuPc)/n-doped 4,7-diphenyl-1,10-phenanthroline (Bphen), is known to generate electrons and holes under the condition of reverse bias by electron tunneling. The generated electrons and holes at the junction move toward undoped electron transport layer (ETL) and the adjacent electrode, respectively. The injection barrier for electrons (Bphen/ETL) and holes (ITO/CuPc) will be much lower than that of the direct electron injection from the electrode to the n-ETL, leading to an efficient electron injection. Most importantly, this method is independent of the work function of the cathodes, which is a valuable advantage for FOLEDs to choose the cathodes.

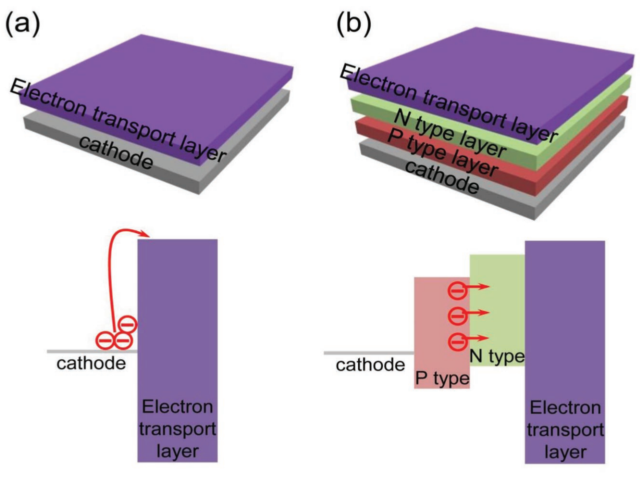

Figure 11. Schematic illustrations of the electron injection mechanism in a) a normal electrode/organic junction and b) an electrode/p–n junction.

As the inherent limit of graphene electrode is that its relatively low work function (about 4.4 eV, work function of different flexible electrodes have been summarized in Table 2) causing the inadequate holes injection, it also needs to be modified to improve carrier injection. Han et al. used a self-organized polymeric hole injection layer (HIL), which has the gradient work function to modify the graphene anode (**Figure 12**).[85] The gradient work function enables holes to be injected easily into the organic layer despite the high hole-injection barrier at the interface between the graphene anode and the organic layer. As a result, the FOLEDs with four layer graphene anode doped with $AuCl_3$ or HNO_3 showed extremely high maximum current efficiencies, 30.2 cd A^{-1} for fluorescent devices (Figure 12b) and 98.1 cd A^{-1} for phosphorescent devices (Figure 12c), and maintained almost the initial current density even after 1000 bending. Also, a 5×5 cm^2 bright and uniform white FOLED has been shown in Figure 12d, which demonstrated the promising application of graphene anodes in future flexible solid-state lighting sources.

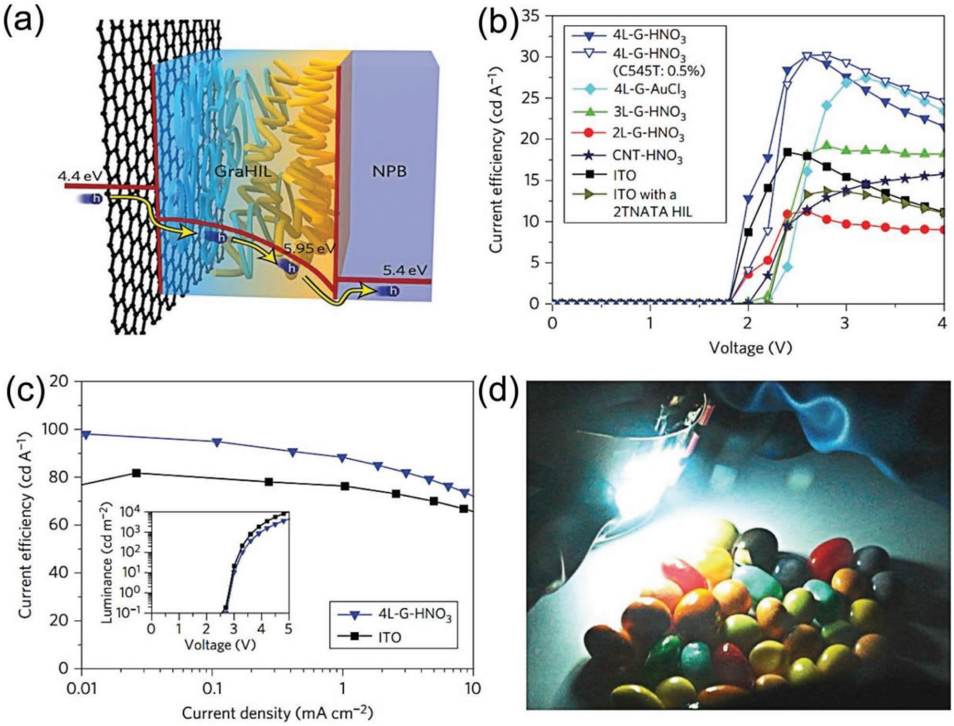

Figure 12. a) Schematic illustration of a hole-injection process from a graphene anode via a self-organized HIL with work-function gradient (GraHIL) to the NPB layer. b) The current efficiency of green fluorescent and c) phosphorescent FOLEDs with graphene doped with HNO_3 (modified with GraHIL) and ITO anode. d) Large-area FOLED with a graphene anode on a 5 cm × 5 cm PET substrate. Reproduced with permission.[85] Copyright 2012, Springer Nature.

3.3. Light Extraction of FOLEDs

Light extraction efficiency has always been the focus and key-point of the FOLEDs due to the significant radiation loss in conventional OLEDs. About 80% of internally generated photons are trapped in different forms including surface plasmon polaritons (SPPs) mode associated with the interface between metal electrode and organic, the substrate mode owing to the total internal reflection at the substrate/air interface, waveguide (WG) modes in ITO anode and organic layers.[141] Thus, light extraction efficiency of the conventional OLEDs is commonly less than 20%. In spite of this, as tremendous research works have been conducted on improving the light extraction of FOLEDs, the highest value of light extraction efficiency is more than 60% at present. This value will be expected to reach 70% by 2020.

As the aforementioned frangibility of ITO, FOLEDs could not be considered the WG modes in ITO because of the little presence in FOLEDs. It is therefore not surprising that the work focused on the improvements of the light extraction by eliminating the SPPs mode, substrate mode, and WG mode in organic layers. Integrating the internal or external micro/nanostructures has demonstrated an effective approach to improve the light extraction efficiency of FOLEDs.[142–159] In case of the internal micro/nanostructures, they are fabricated on a substrate or holes injection layer for an instance of PEDOT:PSS, and then replicate to atop organic materials and electrode layer by layer. The integrated structures enable an additional wave vector for the momentum-matched to couple out WG mode in the organic layer and SPPs mode.[143,145,146,150,156,160] It should be noted that periodic structures will cause an angular dependence of the EL spectra due to the oscillating behavior. Therefore, quasi random nanostructures have been highlighted to solve this problem.[146,150] While in case of external micro/nanostructures with different feature size and geometries, microlens array, conical pillars, micrometric, pyramids, etc., have already been developed in the direction of light emission and demonstrated the ability of improving the light extraction theoretically and experimentally.[147–149,152,154,158,159] Recently, the flexible plastic substrates combined with light scattering particles have started to show their potential on enhancing light extraction of FOLEDs for lighting panel application.[155,157] In the practical application for lighting panel, the substrate with high haze and acceptable transmittance is favorable. Efficient light extraction of FOLEDs with light scattering layer has been demonstrated and nanoparticle doping is apparently cost-effective method.

In terms of fabrication techniques, Feng et al. have discussed that integrating the micro and nanostructures into the conventional OLEDs in detail in the other review.[141] Holographic lithography, nanoimprint lithography, mold transfer process, colloidal lithography, E-beam lithography, and spontaneously formed random patterns due to the anisotropy of materials are all effective techniques. But in this review, we would like to emphasize the method which is more suitable for FOLEDs. Up to now, mold-assisted soft nanoimprinting lithography (SNIL) is the most promising for FOLEDs because it is cost

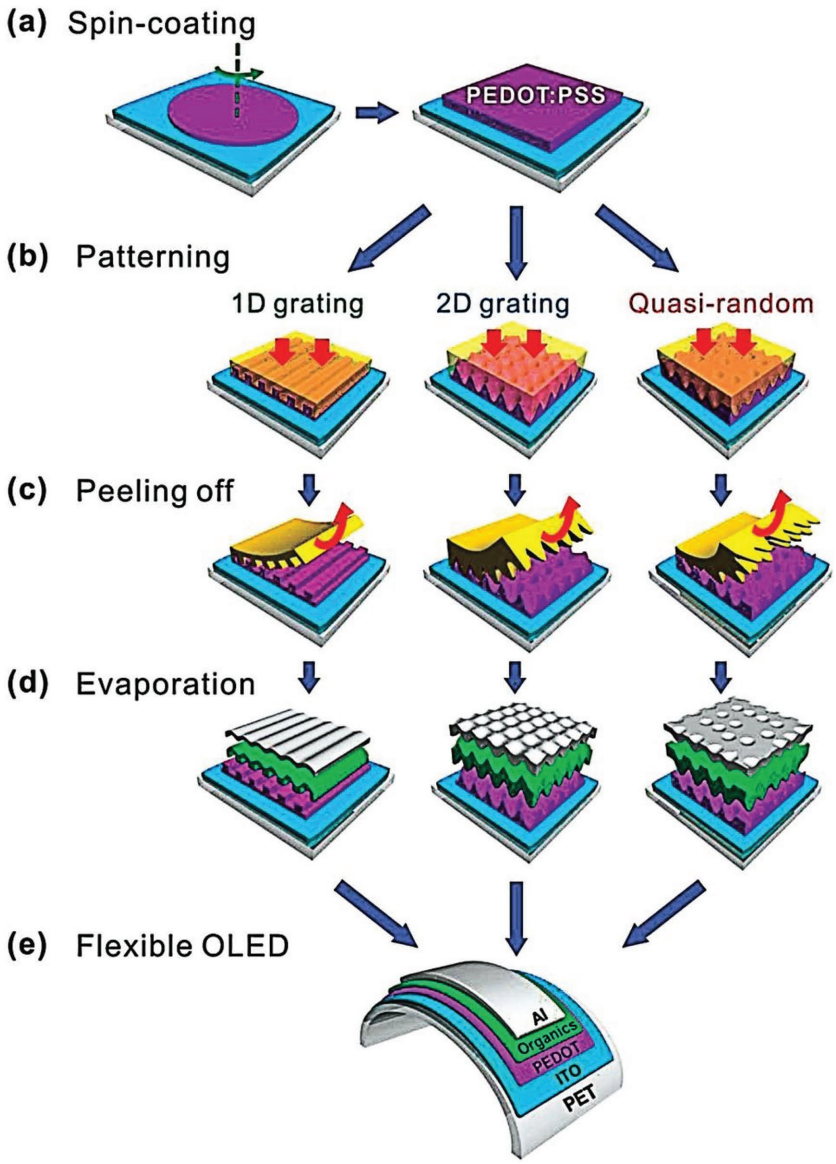

Figure 13. Schematic of the fabrication process of FOLEDs with three imprinted nanostructures. a) Spin-coating the PEDOT:PSS layer on flexible ITO-PET substrates. b) Patterning the PEDOT:PSS layers by soft nanoimprinting lithography with the PDMS molds of 1D grating, 2D grating, and quasirandom nanostructures, respectively. c) Peeling off the PDMS molds. d) Depositing the organic layers and metal electrode on top of the patterned substrates. e) Formation of the nanostructured FOLEDs. Reproduced with permission.[146] Copyright 2014, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

effective, high quality, and high throughput. Many great efforts have been conducted.[142,143,146,149,150,152] As shown in **Figure 13**, SNIL has exhibited its general applicability, been used to fabricate nanostructures including periodic 1D, 2D grating and quasirandom nanostructures. Soft and hydrophobic PDMS was used as mold in favor of peeling off. Due to the conformality of evaporation, the nanostructures could be duplicated layer by layer from nanoimprinted patterns to top electrode. Xu et al. has demonstrated that SNIL could be used to fabricate a nanostructured DMD electrode on the PET substrate.[143] The white emission FOLEDs with the nanostructured DMD electrode enable the angle-independent and broadband outcoupling enhancement by minimizing the waveguide mode, microcavity effect, and surface plasmonic loss related to the metal electrode. The light extraction efficiency of this white FOLED is over 2.4 times that of a conventional device with ITO electrode. In case of integrating external structures, SNIL is employed to fabricate 2D tapered nanopillars array onto the photopolymer encapsulation film in our previous work (**Figure 14**).[152] The experimental and simulated results both demonstrate the enhancement of the light extraction by recovering the optical loss associated with the total reflection at the encapsulation film/air interface. Moreover, the hydrophobic property of the microstructured encapsulation film is helpful to prevent the FOLEDs from pollution by reducing the adhesion probability of dust particles and water droplets in practical applications.

On the other hand, nonstructural antireflection coating has been also used to improve the light extraction of FOLEDs. The mechanism of action of the antireflection coating layer is similar with the DMD electrodes. It enables reducing the light reflection by the combination of the destructive interference of the dielectric layers and the absorption by the thin metallic layers.[161,162] A nonpatterned low refractive index layer as host for the emitters and a nonpatterned high refractive index layer as substrate have demonstrated their effect on improving outcoupling of the WG modes in the ITO/organic layers. Wang et al. have reported a high-index Ta_2O_5 optical coupling layer combined with Au film electrode and MoO_3 on the plastic substrate (**Figure 15**).[162] Ta_2O_5 enables more light to emit from the device by reducing the total internal reflection and tuning the microcavity without altering the structure and thickness of the organic layers or influencing the electric characteristics of the device. A record of that time high external quantum efficiency (EQE) of ≈40% at a very high luminance of 10 000 cd m^{-2} has been acquired using for a green FOLED fabricated with this new electrode design.

4. Emerging Development of FOLEDs

As the development of material science and fabrication technique, some emerging materials and designs have been introduced into FOLEDs. They have promoted the improvements of FOLEDs comprehensively, from fundamental performances to fantastic designs. In this section, we will discuss the emerging FOLEDs including flexible organic/inorganic hybrid perovskite light-emitting devices (PeLEDs) and stretchable FOLEDs.

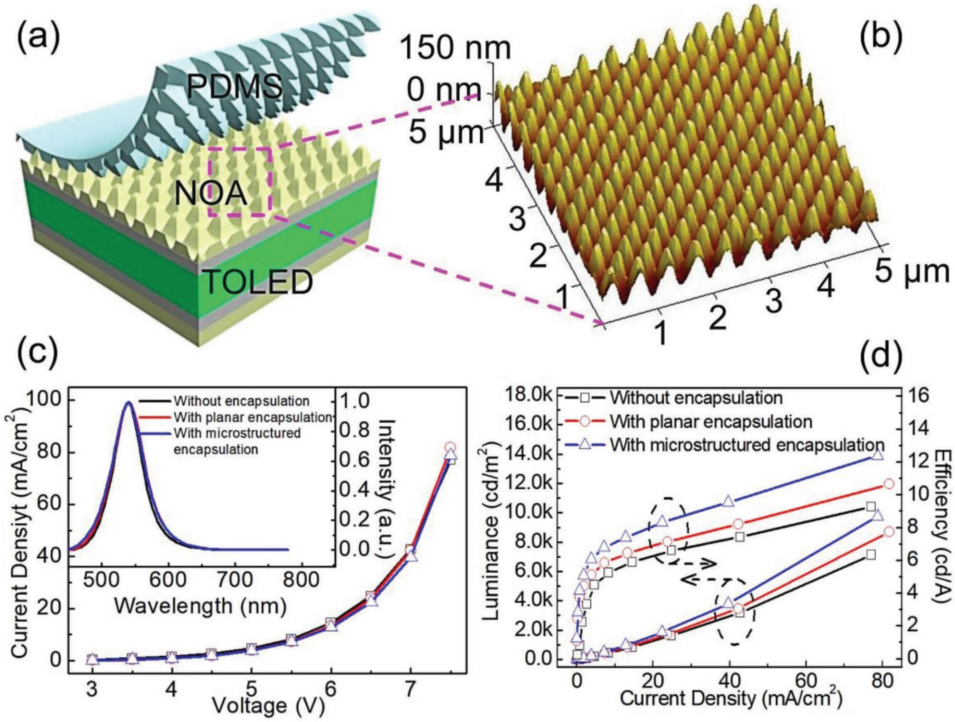

Figure 14. a) Schematic illustrations of TOLEDs with microstructured encapsulation process. b) AFM image of surface morphology for the microstructured encapsulation film. c) The current density–voltage, EL spectra (inset of (c)) and d) luminance–current density–efficiency characteristics of the TOLEDs with and without the encapsulation film. Reproduced with permission.[152] Copyright 2014, Elsevier B.V.

4.1. Flexible Perovskite Light-Emitting Devices

PeLEDs are emerging and promising devices due to their intrinsic advantage of superior low material cost, tunable band gap, color purity, and easy fabrication. Recently, the performance of PeLEDs has been improved remarkably resulting in an increasing number of efforts on them.[163–166] The compatibility with cost effective printing process makes perovskite suitable for flexible display devices. Bade et al. reported a composite film consisting of methylammonium lead tribromide (Br-Perovskite) and poly (ethylene oxide) (PEO) as the emitting layer, and AgNWs as the cathode both fabricated by printing (**Figure 16**a,b).[163] The printing process was carried out in the ambient air without any additive humidity control and it was found that printing the PEO/Perovskite composite film in ambient air could actually improve its luminescence properties. Moreover, the usage of CNT and AgNWs electrode allow for appreciable bending to a curvature radius of 5 mm without influencing the device properties. But so far, this flexible PeLED has lower performance compared with that on glass. In order to realize highly effcient flexible PeLEDs, Seo et al. have developed the PeLEDs using a graphene anode (Figure 16c,d).[165] They can achieve high efficiency, which the maximum current efficiency and external quantum efficiency are 18.0 cd A^{-1} and 3.8%, respectively, and are higher than those of PeLEDs with ITO anode. The mechanical robustness of the flexible PeLEDs

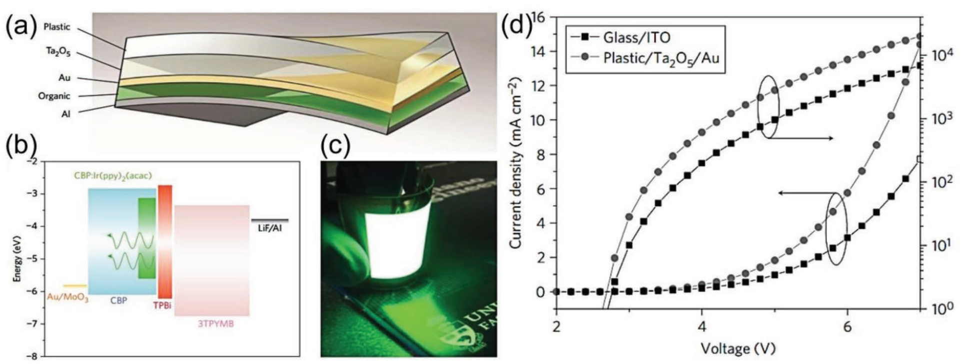

Figure 15. Schematic illustration of a) the optimized OLED structure and b) the energy level diagram on low-cost flexible plastic with metal electrodes. c) Photograph of a large-area FOLED (50 mm × 50 mm) working at high luminance (more than 5000 cd m^{-2}). d) Current–voltage and luminance–voltage characteristics of optimized OLEDs. Reproduced with permission.[162] Copyright 2011, Springer Nature.

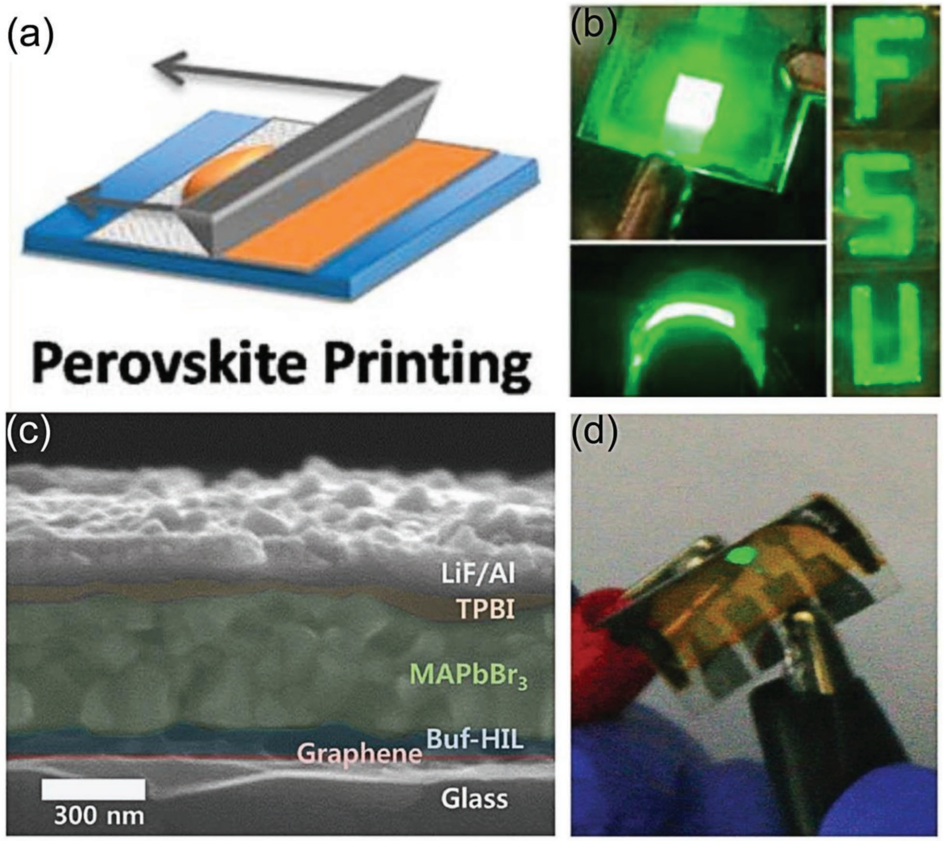

Figure 16. a) Schematic illustrating the fabrication of printed PeLEDs. b) The photographs of the operating PeLEDs. c) Cross-section SEM image and d) a photograph of PeLEDs with graphene anode. (a,b) Reproduced with permission.[163] Copyright 2015, American Chemical Society. (c,d) Reproduced with permission.[165] Copyright 2017, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

with graphene anode is also superior. The current density of flexible PeLEDs just decreased slightly to ≈81% of initial current density after 1200 bending cycles.

4.2. Stretchable Organic Light-Emitting Devices

Stretchable OLEDs have been considered as an alternative technology for the next generation displays.[21,58,164,167–170] Stretchable display enables foldable and expandable screens for wearable display because it conforms to any surface configuration and is mechanically stable to fatigue strain. It is also a biocompatible light source for epidermal or vivo medical devices. As early as ten years ago, Sekitani et al. have already reported a stretchable OLED active-matrix display using the elastic printable conductors.[169] They fabricated the elastic printable conductors which comprised uniformly dispersed SWNTs within a fluorinated rubber. However, this stretchable active-matrix display is constituted with the rigid light-emitting units, which themselves are not stretchable. To realize intrinsical stretchable OLEDs, all the constituent materials of OLEDs should be elastic and stretchable. Liang et al. reported a light-emitting device with a polymer electroluminescent layer sandwiched between a pair of novel elastomeric transparent composite electrodes.[58] The composite electrodes are based on the thin AgNWs networks embedded in the surface of PUA matrix. This device could be stretched at 30% linear strain repeatedly for 1000 cycles of stretching and releasing, and tolerate stretching up to a maximum strain of 120%. But it has a drawback insufficient efficiency. Recently, the efficiency of stretchable OLEDs has been improved drastically. Yin et al. developed a buckling process with laser-programmable technique to achieve stretchable OLEDs with the ordered-buckling (**Figure 17**).[168] The OLEDs were fabricated on smooth and ultrathin polymer film with a thickness of around 10 µm. It is independent of fabricating ordered bucking which are used for stretch by bending and flattening. So the high efficiency could be obtained and maintained repeated stretch–release cycles were conducted. The maximum efficiencies of the device be acquired to 72.5 and 70 cd A^{-1} at 0% and 70% strain, respectively. In addition, it can sustain 100% strain while exhibiting just a little degradation in performance over 15 000 cycles of stretch–release.

5. Conclusion and Outlook

In this review, we have discussed recent developments in FOLEDs including the substrates, electrodes, fabrication, and encapsulation technologies. The merits and demerits of various flexible substrates have been summarized. In terms of the

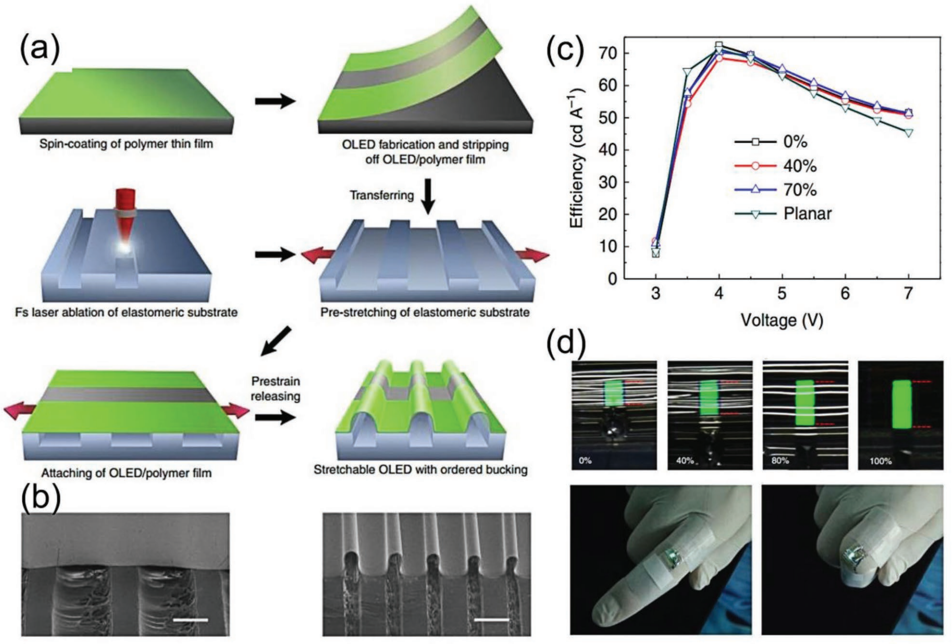

Figure 17. a) Schematic illustration of the fabrication process of stretchable OLEDs. b) Tilted 45o-view SEM images of the stretchable OLEDs corresponding to the prestretching elastomeric substrate (left) and releasing the prestrain to obtain the ordered buckles (right). c) Current efficiency–voltage characteristics of stretchable and planar OLEDs. d) Photographs of the stretchable OLEDs based on a 200% prestretched substrate at 5 V with strain values of 0%, 40%, 80%, and 100% (upper) and fixed on a straight and bent finger joint (bottom). Reproduced under the terms of a Creative Commons Attribution 4.0 International License.[168] Copyright 2016, Da Yin, Jing Feng and Hong-Bo Sun.

electrodes, from conventional electrodes such as thin metal film, conductive polymer, and DMD multilayers electrodes to emerging electrodes such as graphene, CNTs, AgNWs, and their composite have been all introduced. As FOLEDs are compatible with roll-to-roll techniques, we emphasized low cost printing and coating techniques, for an instance of inkjet printing, gravure printing, screen printing, and slot-die coating. Considering the performance of FOLEDs, stability and efficiency play a decisive role in the success of the practical application. Therefore, the encapsulation technology and efficiency improvement strategies including surface morphology manipulation, energy level modification of electrode/organic interface, and light extraction have been always hotspots in FOLEDs. Moreover, perovskite as a rapidly increasing and promising material has been also discussed. To keep pace with the development of next expandable and foldable screens, an increasing number of research on stretchable OLEDs have been conducted and we also presented these in this review.

As the aforementioned improvements of FOLEDs, they have become most promising display technique as well as the emerging light panels. However, the continual progress in efficiency and lifetime and in addition of reducing cost through efficient industrial manufacturing techniques should be carried out persistently. FOLEDs will surely bring a new fantastic display and light world to our daily life.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

flexible, light emitting devices, organic materials

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