光学学报

柔性钙钛矿发光二极管研究进展

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摘要 钙钛矿发光二极管(PeLEDs)以其优异的光电性能和简单的制备工艺，成为柔性可穿戴电子产品及照明和显示领域的潜在候选者。然而，以高性能、高柔性和新兴应用为目标的柔性钙钛矿发光二极管(FPeLEDs)的发展仍面临许多挑战。本文主要介绍了近年来FPeLEDs领域的相关研究工作以及各功能层的优化方法，总结了目前该领域发展面临的主要问题，并对其未来的发展进行展望，有望为未来FPeLEDs在新兴领域中的开发应用提供系统的理解和有价值的启发。

关键词 钙钛矿；柔性钙钛矿发光二极管；柔性电子；高效；稳定性
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1 引 言

钙钛矿材料以其可溶液加工、吸收系数高、带隙可调、载流子扩散距离长等优点，在发光二极管、太阳能电池、激光器、光电探测器和图像传感器等光电子器件领域得到迅速发展。兼具有机和无机半导体优势的钙钛矿材料作为电致发光材料在发光二极管领域备受关注[1-3]。自2014年首个室温电致发光钙钛矿二极管(PeLEDs)问世以来，近红外、红光、绿光领域的外量子效率(EQE)均已超过了20%[4-9]。然而，传统刚性基板的PeLEDs无法满足现代社会对柔性显示及可穿戴电子设备的发展需求，因此开发出具有机械柔性、轻便可穿戴的柔性钙钛矿发光二极管(FPeLEDs)是目前亟需解决的问题[10]。

为了满足柔性器件的实际应用，FPeLEDs的每一层都需要具有良好的灵活性和稳定性。2015年，Kim等[11]通过使用柔性塑料基板代替传统的刚性玻璃基板，制备出第一个柔性器件，其EQE只达到0.125%，且最大弯曲半径为1.05 cm。然而需要高温溅射沉积的氧化铟锡(ITO)电极在反复弯曲或拉伸的情况下会严重退化或失效，这严重阻碍了柔性电子器件的发展，因此新型柔性电极材料应运而生，2017年，Seo等[12]开发出基于石墨烯阳极的杂化钙钛矿发光二极管，有效减少了激子淬灭，在聚对苯二甲酸乙二醇酯(PET)衬底上制备出的FPeLEDs实现了3.8%的EQE，且在7.5 mm的弯曲半径下循环弯曲1200次后仍能保持初始值的81%。尽管PeLEDs的发光性能有了巨大进步，但是钙钛矿材料在柔性衬底上较差的结晶性限制了其进一步发展，因此需要更有效的方法来同时实现高效、稳定和良好柔性的FPeLEDs。Zhao等[13]使用大体积有机卤化铵作为添加剂，器件EQE值达到13%，并且在1 mm的弯曲半径下弯曲10000次后，EQE仍然保持为初始EQE的80%，具有十分优异的柔性性能。2020年，Shen等[14]利用界面工程有效钝化缺陷，抑制非辐射复合损失，基于此制备出的绿光FPeLEDs的最高效率达到24.5%，在3 mm弯曲半径下循环弯曲1000次后仍能保持初始EQE的90%，具有相当优异的机械稳定性。

尽管FPeLEDs有着良好的应用前景，但是其EQE仍然落后于玻璃基底的刚性器件，这就限制了其在高性能可穿戴设备上的应用[15-18]。本文首先对卤化物钙钛矿的基本光电性能进行了简要介绍，并总结了柔性衬底和柔性电极的类型；然后，重点介绍了FPeLEDs的制膜工艺和性能优化方法，并概述了在界面和能级调控方面取得的主要研究进展；最后，展望了FPeLEDs未来的主要研究方向，为该领域的发展提出可行性建议。

2 柔性钙钛矿发光二极管

2.1 钙钛矿晶体结构和光电性能

最初的钙钛矿材料是指自然界中一种天然存在的矿石材料——钛酸钙($CaTiO_3$)，后来泛指和钛酸钙具有相似ABX_3结构的所有材料，其中A一般是有机阳离子[甲胺(MA^+，$CH_3NH_3^+$)、甲脒(FA^+，

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$CH(NH_2)_2^+$)或铯(Cs^+)]等;B一般是二价金属阳离子(Pb^{2+}、Sn^{2+})等;X为卤素阴离子(Cl^-、Br^-、I^-)[19]。钙钛矿的结构为金属阳离子B和邻近的卤素离子构成正八面体结构$[BX_6]^{4-}$,其中B位于正八面体结构的中心,而$[BX_6]^{4-}$以共顶点的方式连接,在三维空间内无限延伸,A位阳离子填充在八面体之间的空隙位置,其三维立方结构如图1(a)所示。

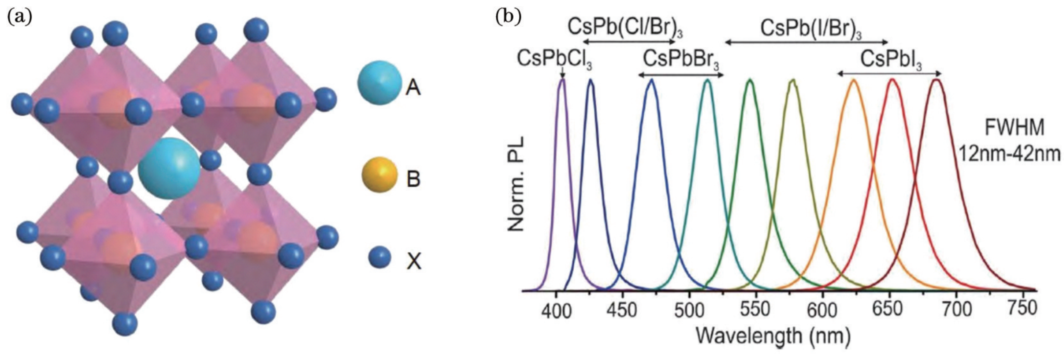

图1　钙钛矿晶体结构及发光光谱[21]。(a)钙钛矿理想晶体结构;(b)$CsPbX_3$纳米晶稳态光致发光光谱

Fig. 1　Crystal structure and emission spectra of perovskite[21]. (a) Ideal crystal structure of perovskites; (b) steady-state photoluminescence spectra of $CsPbX_3$ nanocrystals (NCs)

形成稳定的三维ABX_3钙钛矿晶体,A、B、X 3种离子的半径要满足容忍因子及八面体因子的取值范围,即

$$t=\frac{R_A+R_X}{\sqrt{2}\left(R_B+R_X\right)}, \tag{1}$$

$$\mu=\frac{R_B}{R_X}, \tag{2}$$

式中:容忍因子t要求在0.8~1范围内;八面体因子μ在0.4~0.9范围内;R_A、R_B、R_X分别为A、B、X离子的半径。

和传统的光电材料相比,钙钛矿作为一种直接带隙半导体材料,具有更加优异的光电性能。通过替换钙钛矿结构中的卤素阴离子,可以调整其禁带宽度,而阳离子如MA^+、FA^+和Cs^+的改变则可以将钙钛矿发射光谱扩大至近红外或者紫外区域[20],这有益于实现PeLEDs的全色彩显示。如图1(b)所示:通过改变钙钛矿中卤化物的组成和比例,纯无机钙钛矿$CsPbX_3$的发射光谱可在390~700 nm范围内连续调控[21];将A位阳离子替换为有机MA^+或FA^+离子,其发射波长范围可以拓展到近红外800 nm处;将B位金属阳离子替换为Sn^{2+},发光峰位可以进一步扩展到1050 nm[20]。因此,通过改变钙钛矿结构中A位或B位阳离子或调节X阴离子的比例,可以改变钙钛矿的禁带宽度,适用于发光二极管、太阳能电池、探测器等领域。

高质量钙钛矿薄膜可在低温下制备,并且具有较低的缺陷态密度和较高的载流子迁移率。以$MAPbI_3$钙钛矿为例,理论计算结果显示,其缺陷态密度为$5\times10^{16}\ cm^{-3}$,而单晶$MAPbI_3$钙钛矿的缺陷态密度更低,仅为10^9~$10^{10}\ cm^{-3}$[22]。钙钛矿材料在电荷转移方面具有双极性,既可以传输空穴也可以传输电子,且二者的传输相对平衡[23-24],其载流子迁移率可以达到0.1~10 $cm^2\cdot V^{-1}\cdot s^{-1}$[25],远优于OLEDs材料的载流子迁移率($10^{-5}$~$10^{-4}$ $cm^2\cdot V^{-1}\cdot s^{-1}$)[26]。

2.2　工作原理

PeLEDs是典型的电致发光器件,根据"三明治"结构中电子传输层(ETL)和空穴传输层(HTL)所处位置,可分为图2(a)所示的n-i-p型和p-i-n型。在外置电场的作用下,电子和空穴分别从两极注入,经过电子传输层和空穴传输层分别注入钙钛矿层[图2(b)],二者在库仑力的作用下形成激子(电子-空穴对)。随后激子进行辐射复合,释放出的能量被钙钛矿分子部分吸收,促使其从基态跃迁到激发态,而高能量的激发态不稳定,会再次释放能量回到基态,在此过程中能量以光子的形式释放,完成发光。由于钙钛矿材料具有良好的柔性和延展性,因此将传统刚性PeLEDs中的玻璃基板和电极替换为柔性材料,即可制得FPeLEDs器件[图2(c)]。柔性器件电荷传输层的选择对于器件性能至关重要,常用的电子传输层材料有ZnO、SnO_2、LiF、富勒烯等,常用的空穴传输层材料有MoO_3、NiO、聚(3-己基噻吩)(P3HT)、聚(苯乙烯磺酸盐)(PEDOT:PSS)等。

在FPeLEDs中,器件的柔性通常可以通过测量器件在外力作用下的弯曲半径(R)来衡量[图2(d)]。具体而言,在相同的循环弯曲次数下,如果器件的弯曲半径越小,性能参数变化越小,说明器件的机械稳定性越优异。

3　柔性钙钛矿发光二极管的研究进展

自从第一个LED器件问世以来,提高器件性能一直是研究人员致力解决的问题,并为此进行了广泛的研究探索[27-31]。本节总结了对柔性钙钛矿发光二极管中各功能层的研究进展,包括柔性基板、柔性电极、钙

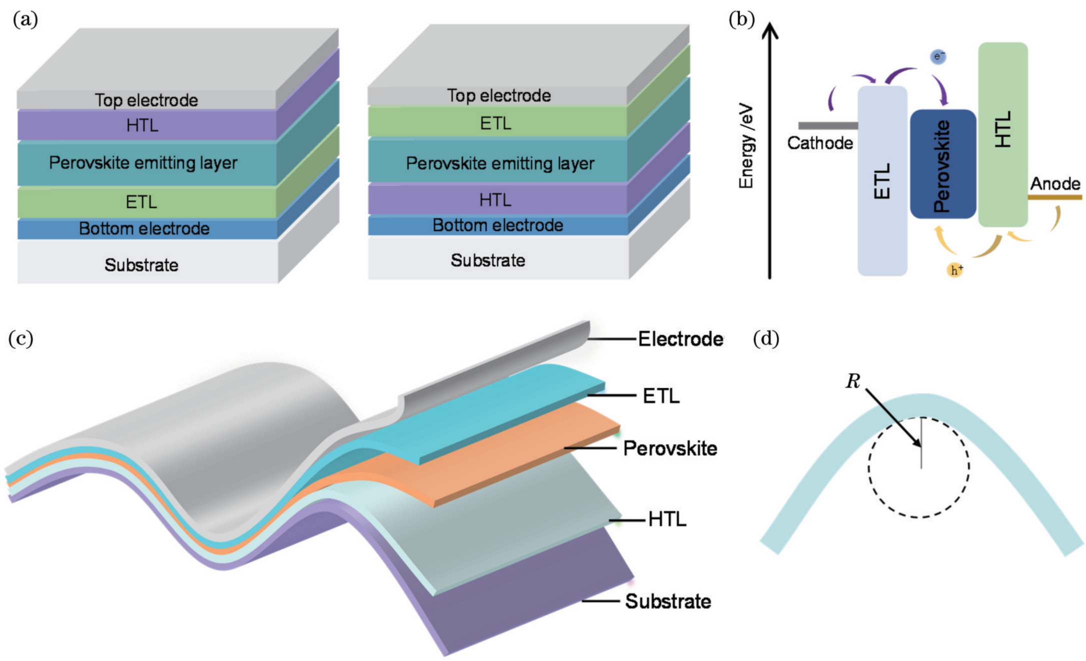

图 2 器件结构及工作原理。(a)常见 PeLEDs 的器件结构；(b)PeLEDs 工作原理；(c)FPeLEDs 结构示意图；(d)FPeLEDs 在弯曲状态下的示意图

Fig. 2 Device structure and working principle. (a) Device structure of PeLEDs; (b) working principle of PeLEDs; (c) device structure of FPeLEDs; (d) schematic of FPeLEDs in bending state

钛矿发光层和界面工程等方面的研究内容。

3.1 柔性基板

柔性基板的选择对柔性器件的光伏性能起着关键作用。薄膜在基板上的沉积会受到其化学性能和力学性能的影响，因此适用于 FPeLEDs 的基板必须具有优异的柔韧性、高透过率和良好的稳定性。目前，柔性器件的制备中主要采用透明聚合物、金属箔、柔性玻璃等作为柔性基板[32-38]。

透明聚合物基板具有柔韧性好、透光性强、质量轻等优点，被广泛应用于柔性器件中[39]。常用的透明聚合物基板的性能如表 1 所示。其中，PET 和聚萘二甲酸乙二醇酯(PEN)常被用作柔性发光二极管器件的基底材料，并在其表面沉积 ITO 作为透明导电电极。它们具有低成本、高透光性和耐溶剂等优点。目前，采用 PET 基板制备的柔性绿色发光器件已经达到 24.5% 的最高效率[14]。然而，柔性聚合物基板在高于玻璃转变温度(T_g)的环境下会出现变形和薄层电阻增大的问题[图 3(a)]，这将直接导致器件退化甚至失效[40]。例如，高质量的 ITO 沉积需要在较高的温度(300～400 ℃)下进行直流溅射，而柔性聚合物基板的 T_g 常常低于 150 ℃，这可能导致沉积的 ITO 呈非晶态或破坏基底[41]。此外，透明电极中存在大量的缺陷和电荷陷阱，不利于高效率发光器件的制备[42]。为此，研究人员选择具有耐高温性能的柔性聚合物聚酰亚胺(PI)作为基板，以获得高质量的透明电极[13, 43]，然而 PI 的成本问题限制了其商业化进程。

柔性器件常常处于受到外应力的变形状态，柔性衬底和脆性涂层之间存在模量差异，导致各层间剪切力分布不均，从而产生微裂纹甚至贯穿纹，造成效率出现不同程度的衰减。因此，对衬底的形变恢复性和有效释放应变能力的表征也十分重要。Li 等[44]通过湿化学方法成功地将银纳米线(Ag NWs)嵌入柔性 PI 衬底，增强了柔性透明基板的光学性能和电学性能[图 3(b)]，基于此制备出的柔性钙钛矿量子点发光二极管显示出非常好的机械拉伸稳定性，即使经过 1000 次连续拉伸-释放循环，亮度仅下降了 25%，电流效率(CE)则比初始值高出 25%，这与超薄薄膜在屈曲过程中应力的有效释放密切相关。Qian 等[33]采用 PI/Ag NWs 复合策略，显著地提升了基板复合电极的机械柔性，在不同弯曲应变的实验中，PI/Ag NWs 电极的电阻保持在 20 $\Omega\cdot sq^{-1}$ 左右[图 3(c)]，说明引入银纳米线可提升基板的形变恢复性能和释放应变的能力。

为了应对塑料基材的环境污染问题，研究人员正在开发可生物降解和生物相容的新型柔性基板。Jin 等[46]制备了一种兼具机械坚固和生物友好特性的甲壳质纳米纤维纸质基板，这种纳米纤维纸在整个可见光范围内具有优异的透射性[550 nm，91.7%，图 3(d)]和良好的热稳定性。此外，塑料基材本身的抗湿性和抗氧性较差，可能导致器件寿命较短。云母是一种天然的透明结晶材料，具有透过率高、水氧阻隔性能好、热膨胀系数小、表面光滑、耐热性好等特点。同时，云母可以被剥离成具有高透明度和可弯曲性的微纳米级柔性片[39]，因此被认为是一种非常适合替代柔性聚合物基板的材质。

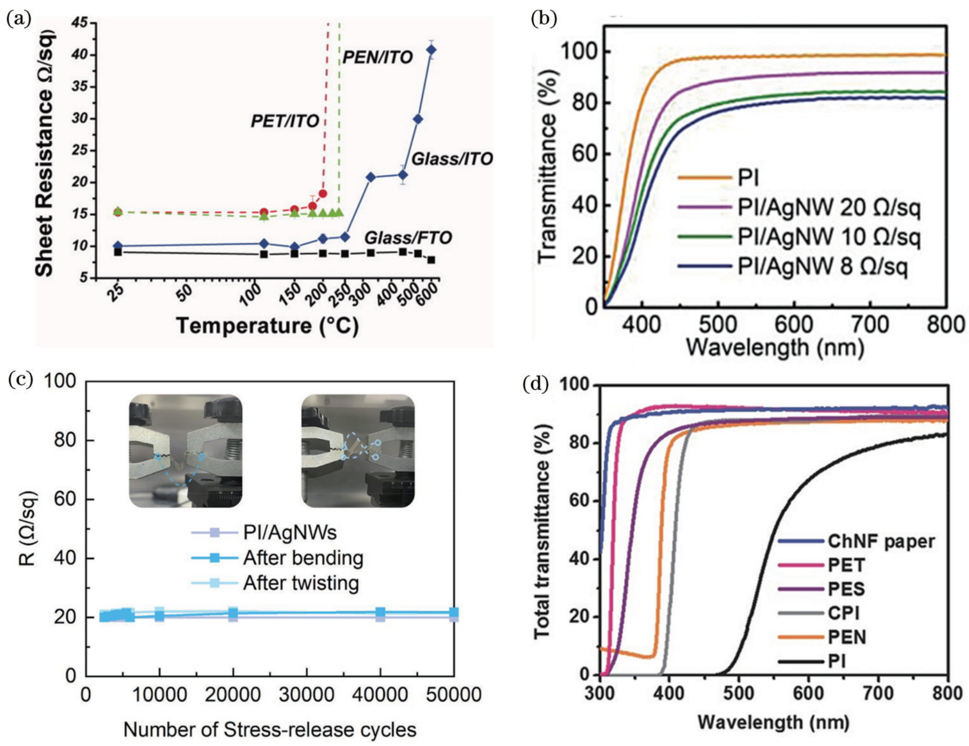

图3 柔性基板的性能。(a)柔性基板和玻璃基板在不同温度下热处理 30 min 后的薄层电阻[40];(b)片状电阻为 20、10、8 Ω·sq^{-1}的纯PI膜和PI/Ag NWs复合膜的透射光谱[44];(c)PI/Ag NWs电极的薄片电阻和弯曲应力的函数关系,插图为电极在特定曲率半径处弯曲的照片[33];(d)甲壳质纳米纤维纸与其他聚合物的透射光谱对比[46]

Fig. 3 Performance of flexible substrates. (a) Variation of thin layer resistance of flexible substrate and glass substrate after 30 min heat treatment at different temperatures[40]; (b) transmittance spectra of neat PI film and PI/Ag NWs composite films with sheet resistance of 20, 10, and 8 Ω·sq^{-1}[44]; (c) functional relationship between sheet resistance and bending stress of PI/Ag NWs electrodes, inset shows the photograph of electrode bent at a specific radius of curvature[33]; (d) transmission spectra of chitin nanofiber paper compared to other polymers[46]

表1 常用柔性聚合物基板的性能参数[45]

Table 1 Parameters of commonly used flexible polymer substrates[45]

Type	T_g /℃	Melting temperature (T_m) /℃	Density /(g·cm^{-3})	Modulus /MPa	Work temperature /℃	CTE /(10^{-6} ℃$^{-1}$)	Water absorption /%	Solvent resistance	Dimensional stability
PEN	120-155	269	1.36	$(0.1\text{-}0.5)\times10^3$	−155	20	0.3-0.4	Good	Good
PET	70-110	115-258	1.39	$(2\text{-}4.1)\times10^3$	−50-150	15-33	0.4-0.6	Good	Good
PI	155-270	250-452	1.36-1.43	2.5×10^3	~400	8-20	1.3-3.0	Good	Fair
PC	145	115-160	1.20-1.22	$(2\text{-}2.6)\times10^3$	−40-130	75	0.16-0.35	Poor	Fair
PDMS	−125	—	1.03	1	−45-200	310	>0.1	Poor	Good
TPU	80	180	1.18	7	130	153	0.2	Good	Good

与透明聚合物基板相比,金属基板如钛、铜箔和不锈钢箔具有更大的加工温度窗口,但是金属基板的表面粗糙度较高,导致后续溅射的电极覆盖度不高,从而产生严重的漏电问题[47]。此外,在热应力或光照下,金属基板中的卤素离子和金属原子会扩散,严重损坏器件的性能[48]。柔性玻璃具有高功率质量比的特点,也被用作柔性基板材料,然而该材料的曲率半径仅为厘米级,且成本较高[49],不适合大面积推广使用。

3.2 柔性电极

为了实现高性能FPeLEDs器件,选择适当的柔性电极至关重要。传统刚性器件中常用的ITO电极需要高温沉积,与柔性基板的要求相悖[50-51]。因此,研究人员开发了多种替代ITO的柔性电极材料,包括机械柔性好的金属电极、碳电极和导电聚合物电极等。

3.2.1 金属电极

金属薄膜(如Au、Ag、Al)具有导电性好、机械柔性优和反射率高等优点,因此在柔性电极中得到广泛应用。然而,金属电极和柔性基底之间的表面能不匹配、润湿性差等都会导致金属薄膜在沉积过程中呈岛状生长,导致表面粗糙度和漏电电流增加[52]。为了解

决这些问题，Liu等[53]制备了厚度为7 nm的超薄Au电极，并将其用于高性能FPeLEDs。他们发现，沉积在不同衬底上的Au薄膜的表面形态如图4(a)所示，相比之下，沉积在MoO_3/SU-8改性玻璃衬底上的Au薄膜的表面形貌和连续性得到明显改善。这主要是因为SU-8层和Au原子之间存在强共价键，而MoO_3的添加进一步抑制了表面金属原子的扩散。与基于ITO的器件相比，基于超薄Au薄膜的器件CE提高了10%，并在经过1000次弯曲循环后仍然保持良好的机械柔性。Zhang等[54]报道了一种由无润湿层制备光滑Ag薄膜作为柔性透明导体(FTC)的新方法[图4(b)]。他们通过Ni和Ag的共沉积获得了具有低光学损耗和强热稳定性的Ag薄膜电极，平均可见光透过率达到80%。与基于ITO电极的器件相比，所制备FPLEDs的CE提高了30%。经过1200次0.25 inch(1 inch=2.54 cm)弯曲半径下的循环弯曲后，该器件的最大EQE达到4.14%，并保持在初始值的70%。

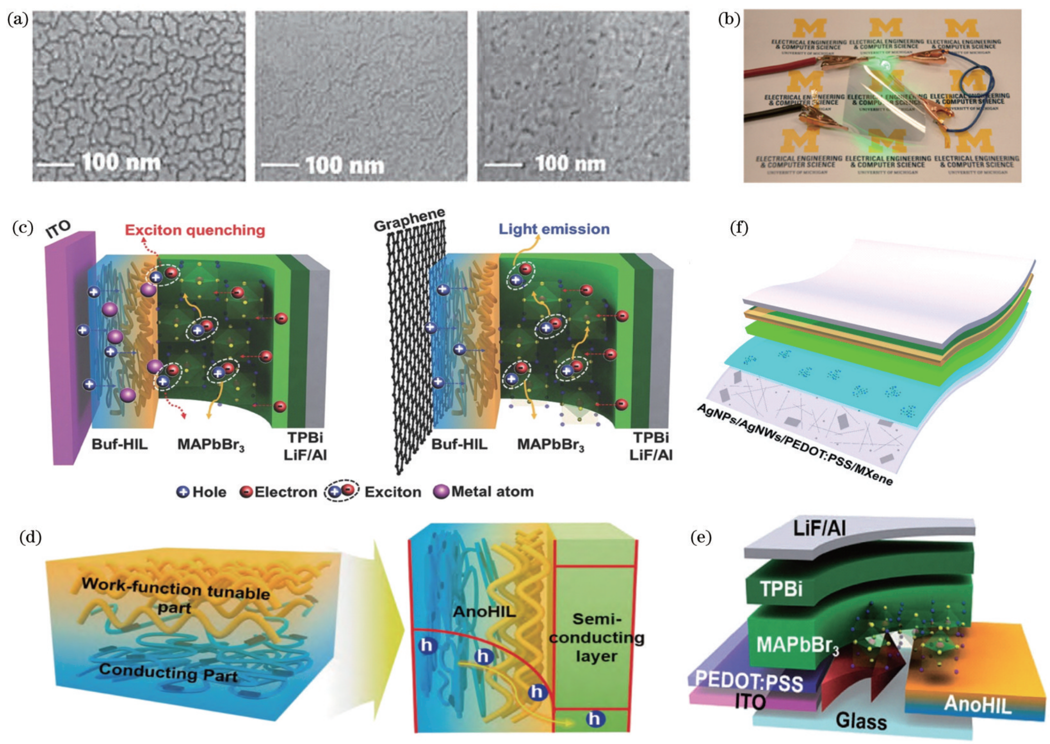

图4 基于不同柔性电极材料的薄膜及器件。(a)沉积在玻璃、SU-8改性玻璃和MoO_3/SU-8改性玻璃衬底上的7 nm超薄Au膜的SEM图像[53]；(b)基于Ni掺杂的大面积Ag-FTC在绿光LEDs中的应用[54]；(c)ITO基PeLEDs中In和Sn原子导致的激子淬灭的示意图，以及石墨烯基PeLEDs对荧光淬灭的避免[12]；(d)双功能Ano-HTL和从Ano-HTL到相邻半导体层的空穴注入过程示意图[66]；(e)器件结构图[66]；(f)基于复合电极的器件结构示意图[32]

Fig. 4 Thin films and devices based on various flexible electrode materials. (a) SEM images of surface morphologies of the 7 nm ultrathin Au films deposited on bare glass, SU-8 modified glass, and MoO_3/SU-8 modified glass substrates[53]; (b) picture of a large-area Ni-doped Ag-FTC being utilized in a circuit that lights up a green LEDs[54]; (c) schematic of exciton quenching resulting from In and Sn atoms in ITO-based PeLEDs and circumvention of this process in graphene-based PeLEDs[12]; (d) schematic of difunctional Ano-HTL and hole injection process from Ano-HTL into an overlaying semiconducting layer[66]; (e) device structure of simplified PeLEDs[66]; (f) schematic of device structure based on composite electrodes[32]

与金属薄膜相比，金属纳米线电极透过率更高，机械柔韧性更好，可使用溶液法进行大规模制备[55-56]，但是其较差的稳定性和较高的接触电阻限制了器件性能的提升。为了解决这些问题，Kang等[57]使用溶液电镀技术制备Ag-Ni核壳型电极，不仅避免了Ag NWs电极和钙钛矿材料间的有害反应，减少了缺陷态的数量，还增加了Ag NWs电极的功函数，促进了空穴的有效注入，基于此获得了EQE为9.67%的高效$FAPbBr_3$-FPeLEDs。

3.2.2 碳电极

与金属电极相比，石墨烯和碳纳米管(CNT)等碳电极具有更高的透过率和载流子迁移率，以及良好的机械柔性，是柔性器件中透明阳极的良好选择。尽管单个CNT的导电性极好，但是多个CNTs间的势垒会导致薄膜的导电性差且薄层电阻高，同时较差的表面形态也是亟需解决的问题。研究人员尝试了多种方法，包括添加钝化层、化学后处理、掺杂等。Cai等[58]将多壁纳米管(MWCNTs)嵌入聚二甲基硅氧烷(PDMS)中，以制备具有高透明度和优异导电性的可

拉伸柔性电极。他们首先将SWCNT膜浸入硝酸溶液中以引入亲水基团并去除杂质，利用氧等离子体处理PDMS以增强表面亲水性；然后，将处理后的SWCNT薄膜铺到PDMS表面，并使用蒸馏水压平，以获得具有良好电学性能的SWCNT薄膜电极。

通常，石墨烯可以通过机械剥离、液相剥离、化学气相沉积（CVD）等方法制备。在Ni、Cu或Pt等衬底上生长高质量、大规模且连续的石墨烯，随后将其从衬底上转移到靶衬底是常用的制备方法[59]。Seo等[12]首次使用石墨烯作为阳极替代ITO，制备出高效的FPeLEDs，避免了传统ITO层中In和Sn原子扩散导致的淬灭位点的形成［图4(c)］，显著提高了发光层的光致发光（PL）强度和寿命，以7.5 mm弯曲半径进行1200次循环弯曲测试后，电流保持为初始值的81%，而基于ITO电极的器件则完全失效。但是，在石墨烯的制备和转移过程中，会引入一些缺陷，如重叠边界、针孔、裂纹，导致透过率和电导率降低。为了提高石墨烯层的薄膜质量，Kang等[60]使用氯化金（$AuCl_3$-CH_3NO_2）和硝酸作为掺杂剂，获得43 $\Omega \cdot sq^{-1}$的低电阻和89%高透过率的高质量石墨烯膜，并且提高了其机械稳定性，该石墨烯膜更适合在柔性器件中应用。

3.2.3 导电聚合物

除了金属和碳电极，具有良好导电性和柔性的聚合物也是理想的柔性电极材料[61]，常用的有聚乙炔、聚苯胺、PEDOT：PSS、P3HT及其衍生物[62-64]。在PEDOT：PSS中，可以通过掺入二甲基亚砜（DMSO）、N-甲基吡咯烷酮（NMP）和离子液体等溶剂来增强其导电性。Vosgueritchian等[65]使用DMSO和氟表面活性剂（Zonyl）来改善PEDOT：PSS薄膜的导电性，改性后PEDOT：PSS能够轻松沉积在各种疏水基底上，并且能够承受超过5000次的拉伸循环。Jeong等[66]开发了一种新型阳极材料（Ano-HTL），该材料兼具阳极和空穴传输层的功能，将其应用于DMSO作为导电增强剂，四氟乙烯-全氟-3,6-二氧杂-4-甲基-7-辛烯磺酸共聚物（PFSA）作为功函调节剂的器件中，可获得EQE达到8.66%、最大CE为42 $cd \cdot A^{-1}$的高性能器件。图4(d)展示了具有双重功能的Ano-HTL示意图和从Ano-HTL阳极到相邻半导体层的空穴注入过程，图4(e)为简化后的器件结构示意图。

3.2.4 复合电极

为了充分利用各种电极的优点，复合电极应运而生。Lee等[67]通过使用PEDOT：PSS和Ag NWs复合电极，并进行表面H_2SO_4处理，制备出高效的FPeLEDs。经H_2SO_4处理后的复合电极薄膜的粗糙度最低，这是因为PEDOT：PSS填充了Ag NWs的间隙，并降低了表面张力，有利于从玻璃上剥离。基于复合电极的FPeLEDs具有更低的启亮电压和更高的效率，CE（最大值为17.9 $cd \cdot A^{-1}$）也明显高于使用PEN/ITO电极的器件（最大值为9.48 $cd \cdot A^{-1}$）。然而，目前柔性复合电极的整体性能仍落后于ITO。为此，Cao等[32]提出一种基于混合尺寸（0D-1D-2D-3D）复合电极的高效FPeLEDs，如图4(f)所示。该复合电极由0D Ag纳米颗粒（Ag NPs）/1D Ag NWs/2D MXene/3D PEDOT：PSS组成。引入的Ag NPs能够有效填充Ag NWs之间的空隙，在MXene和PEDOT：PSS的协同作用下，获得更加光滑均匀的表面形貌。同时，高导热性的MXene使PEDOT：PSS发生相变，进而增强了材料的导电能力并提高了散热效果。基于此复合电极制备的FPeLEDs获得了49 807 $cd \cdot m^{-2}$的最高亮度和16.5%的最大效率。经过500次弯曲半径为1 cm的循环弯曲后，其EQE保持为初始值的90%，表现出良好的机械稳定性。

3.3 钙钛矿发光层

钙钛矿层的薄膜质量对于器件性能至关重要。良好的薄膜形貌和均匀的晶粒尺寸是实现高性能器件的先决条件。首先，钙钛矿薄膜的制备方法直接影响其形貌和结晶质量，因此研究人员开发了多种成膜方法来制备高质量发光层。此外，添加剂工程、界面工程等也被广泛应用于钙钛矿发光层的优化中，以提升钙钛矿薄膜的质量与机械柔性[68]。

3.3.1 薄膜制备工艺

旋涂法是实验室常用的薄膜制备方法，它是将溶液或悬浮液涂覆在基底上，通过离心力使液体均匀分布在基底表面，并通过旋转过程中的溶剂挥发或固化来形成薄膜[69]。Kim等[11]使用旋涂法，在柔性PET基底上制备出首个FPeLEDs器件，效率仅为0.125%，且最大弯曲半径为1.05 cm。旋涂法操作简单，但高质量薄膜的获得需要优化制备条件，包括溶剂选择[70-72]、退火温度[73-74]、溶液浓度[75-76]和前体溶液组成[77-78]等。例如，Cho等[79]采用纳米钉扎（NCP）技术，通过使用高挥发性的氯仿作为溶剂在旋涂过程中洗去DMSO，进而缩短溶剂挥发时间，减小$MAPbBr_3$的晶粒尺寸（平均值为99.7 nm），提高薄膜覆盖率。基于此方法制备的FPeLEDs即使在高弯曲状态下也表现出良好的运行稳定性。此外，Jung等[34]从薄膜退火角度出发，提出一种闪光退火（FLA）法来实现钙钛矿的超快重结晶，如图5(a)所示。旋涂制备的钙钛矿薄膜通常呈现立方体形态，具有较大的粗糙度和大量空隙，而经过FLA处理后的薄膜更加光滑致密，晶粒尺寸减小（约38 nm），有助于辐射复合。经过FLA处理后的FPeLEDs的电流效率和亮度分别提高了252%和409%。然而，简单旋涂的方法不适合大面积均匀钙钛矿薄膜的制备，更无法满足大规模商业化发展的需求。

双源热蒸发也可实现柔性钙钛矿薄膜的制备，它是在高真空环境下使用两个源同时将钙钛矿前驱体材料（$PbBr_2$和CsBr）沉积到基板上。通过调控不同前驱

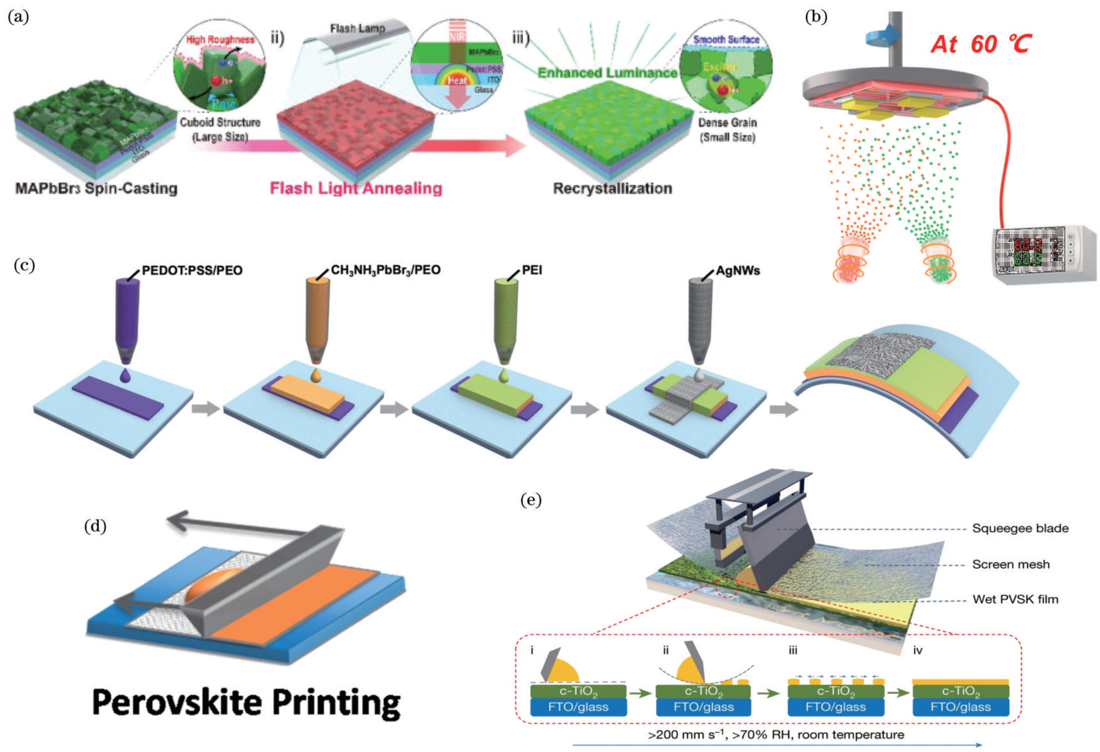

图 5 柔性钙钛矿薄膜制备方法。(a)FLA 方法示意图[34];(b)$CsPbBr_3$发光层双源热蒸发结合原位动态热结晶过程示意图[81];(c)全喷墨打印 FPeLEDs过程示意图[82];(d)钙钛矿薄膜刮涂制备过程示意图[83];(e)丝网印刷沉积钙钛矿薄膜示意图[84]

Fig. 5 Preparation methods of flexible perovskite thin films. (a) Schematic of FLA method [34]; (b) schematic of $CsPbBr_3$ layer thermal vacuum co-evaporation deposition process in conjunction with *in situ* dynamic thermal crystallization[81]; (c) schematic of fabrication processes of all-inkjet-printed FPeLEDs[82]; (d) schematic of preparation process of perovskite thin film by scraping[83]; (e) schematic of screen-printing sedimentary perovskite membrane[84]

体材料的沉积条件,如温度、沉积速率和时间等,获得具有不同Cs和Pb物质的量比与厚度的薄膜。Li等[80]使用该方法制备出FPeLEDs,其电流效率可达到4.16 cd·A^{-1},EQE达到1.37%。在5 mm弯曲半径下进行100次循环弯曲后,亮度仍能保持初始值的80%,具有良好的抗弯曲性能。然而,热蒸发制备的钙钛矿薄膜的均匀性和结晶性有待进一步提高。Chen等[81]将连续低温热退火与热蒸发相结合[图5(b)],通过调节衬底温度和沉积速率来调控薄膜质量,成功制备出亮度约为10000 cd·m^{-2}、最佳电流密度为400 mA·cm^{-2}的高性能大面积(70 mm×20 mm) FPeLEDs。尽管双源共蒸发法可以获得相对均匀平整的钙钛矿薄膜,但是需要对薄膜成分进行精确控制,这在实践中具有一定的挑战性。此外,该方法需要在高真空环境下进行,这增加了设备成本,并限制了其在大规模制备中的应用。因此,针对热蒸发法的改进和发展仍然是必要的,以满足未来钙钛矿材料在FPeLEDs应用中的需求。

喷墨打印可以在柔性基底上直接制备钙钛矿薄膜,并通过对油墨消耗和图案的精确控制,实现高分辨率的图案化,展示了其在柔性器件领域的潜力。Zhao等[82]报道了一种全喷墨打印的FPeLEDs[图5(c)],他们在弹性基底上制出新型四层结构,包括底部电极、钙钛矿发光层、缓冲层和顶部电极。该喷墨打印的FPeLEDs在性能上表现出色,最大亮度为10227 cd·m^{-2},最大电流效率为2.01 cd·A^{-1},且该器件在2.5 mm弯曲半径下仍能保持优异的稳定性。然而,喷墨打印仍面临一些挑战,如对墨水稳定性、材料兼容性和印刷分辨率的改进等。通过进一步的技术改进,喷墨打印有望成为推动FPeLEDs产业化发展的重要工艺之一。

刮涂法是一种将溶液涂覆在衬底上,并使用刮刀均匀刮平形成薄膜的方法。Bade等[83]在柔性聚合物/碳纳米管基底上刮涂由钙钛矿和PEO组成的复合薄膜[图5(d)],并成功制备出最大EQE为0.14%的柔性器件,在5 mm弯曲半径下仍能正常发光。刮涂法可用于柔性大面积钙钛矿薄膜的制备,具有灵活性强的特点,但其在薄膜均匀性和厚度控制方面具有一定的挑战。相比之下,丝网印刷法在薄膜质量调控方面更具有优势。它是将油墨置于丝网镂孔板上,用刮刀将油墨刮过丝网,使油墨通过镂孔传递到衬底上,形成所需的薄膜结构。丝网印刷工艺对油墨的黏度要求高,而用于溶解钙钛矿前驱体的传统有机溶剂,如DMF、DMSO等的黏度通常较低,难以满足丝网印刷的需求。为解决这一问题,Chen等[84]提出使用黏度高且黏度可调的离子液体MAAc来替代传统有机溶剂,

通过调控温度和前驱体组成等参数，实现了基于钙钛矿薄膜的丝网印刷工艺[图 5(e)]。基于丝网印刷工艺在薄膜多元化、生产成本和效率等方面的优势，将其应用于FPeLEDs具有良好的前景。

3.3.2 薄膜性能优化

钙钛矿薄膜通常具有致密的多晶结构，其高脆性限制了其在FPeLEDs中的应用。为了克服这一问题，研究人员致力于通过调控薄膜形貌和结构来提高钙钛矿薄膜的柔性性能。

1)调控晶粒尺寸

晶粒尺寸是钙钛矿薄膜柔性的关键影响因素之一，较小的晶粒尺寸可以提高薄膜的形变能力，降低应力集中程度，减慢裂纹传播速度。然而，晶粒尺寸的减小必然会导致晶界处缺陷态密度增加，从而影响载流子的传输和辐射复合过程，降低器件的发光。因此，在设计和制备柔性薄膜时，需要综合考虑机械柔性和光电性能之间的平衡，而引入添加剂是常用的调控策略。Zhao等[13]发现不同烷基链长的大体积有机铵卤化物添加剂可以显著降低钙钛矿晶粒尺寸[10.5~12.4 nm vs (70±18) nm]。同时，他们还研究了烷基链长度对钙钛矿薄膜光致发光量子产率(PLQY)和柔性性能的影响[图 6(a)]，最终选择4-氟苄基碘化铵(FPMAI)添加剂成功改善了钙钛矿层的光电性能和机械柔性，实现了高效稳定的FPeLEDs。这些器件的最大EQE达到了13%，在弯曲半径为2 mm的10000次弯曲循环后也没有发生退化[图 6(b)]。Cheng等[85]

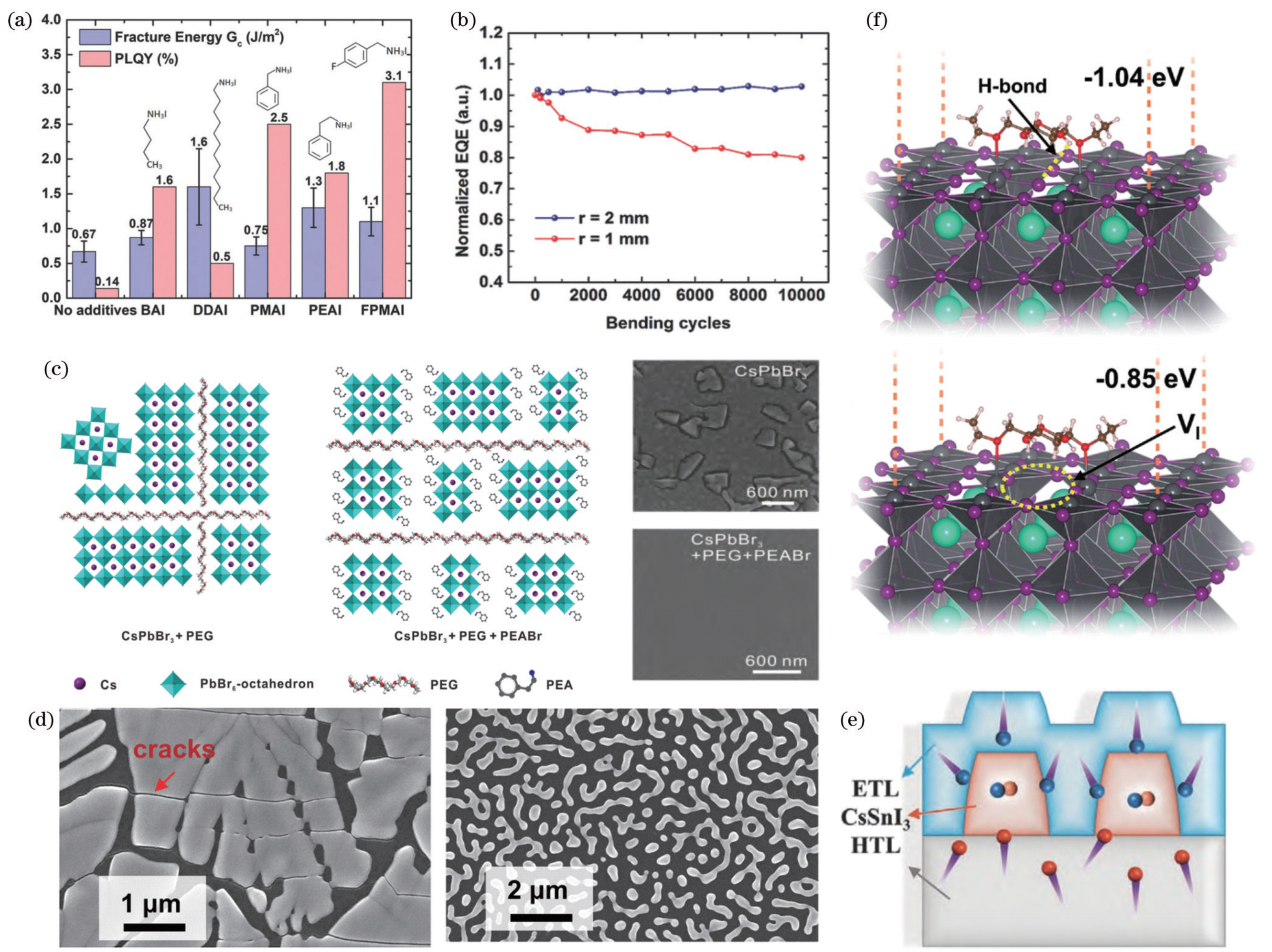

图6 FPeLEDs中钙钛矿多晶膜的性能优化。(a)标件及引入不同添加剂的钙钛矿薄膜的断裂能和PLQY[13]；(b)含有FPMAI添加剂的柔性器件在不同弯曲半径下的EQE随弯曲次数的变化[13]；(c)两种添加剂(PEABr和PEG)协同调控钙钛矿薄膜形貌原理及SEM图谱[85]；(d)2000次弯曲循环后，钙钛矿薄膜的SEM图谱[36]；(e)钙钛矿薄膜树枝状形貌中载流子的传输、复合过程示意图[36]；(f)EC和$CsPbI_3$相互作用示意图[35]

Fig. 6 Optimization of perovskite films in FPeLEDs. (a) Fracture energy and PLQY of perovskite films with or without various additives[13]; (b) normalized EQE versus bending cycles at bending radii of 1 mm and 2 mm for the FPeLEDs with FPMAI additives[13]; (c) principle diagram and SEM diagram of PEABr and PEG synergistic regulation of perovskite morphology[85]; (d) SEM images of dense and dendritic $CsSnI_3$ films after 2000 bending cycles[36]; (e) schematic of charge carriers transport and recombination process in dendritic $CsSnI_3$ film[36]; (f) schematic of the interaction between EC and $CsPbI_3$[35]

采用双添加剂[苯乙基溴化铵(PEABr)和聚乙二醇(PEG)]协同策略，其中PEABr用于减小钙钛矿晶粒尺寸(<15 nm)，PEG用于钝化薄膜缺陷[图6(c)]。基于此，他们成功制备出CE和EQE分别达到31.0 cd·A^{-1}和10.1%的绿光FPeLEDs。

2)构筑薄膜微纳结构

微纳结构的引入能够减轻在器件弯曲过程中可能发生的裂纹和元素扩散等问题，从而提高器件的柔性性能。Lu等[36]制备的$CsSnI_3$钙钛矿呈现出独特的树状结构。如图6(d)所示，在2000次弯曲循环后钙钛矿薄膜的形态几乎没有发生变化，显示出其在柔性器件制备方面的巨大潜力。由此制备的FPeLEDs在50次弯曲循环后，EQE仅衰减了6.6%，大面积(15 mm×6 mm)柔性器件在弯曲状态下仍能保持均匀明亮的发光状态。同时，这种枝状结构能够增大其和电子传输层的接触面积，显著增强电子的注入，增大辐射复合的概率[图6(e)]。

3)物理分散和化学交联

聚甲基丙烯酸甲酯(PMMA)是一种稳定的透明材料，具有优异的透光性和良好的可加工性能，Chen等[86]将其应用到活性层的制备中，获得了具有良好的柔性和稳定性的钙钛矿/PMMA复合薄膜，其能在保持良好发光特性的同时承受多次弯曲和折叠(4000次弯曲循环后，荧光强度仍能保持初始值的50%)。Sun等[35]将低成本高柔性的绿色生物质材料——乙基纤维素(由含有OH^-和乙醚基团的六元杂环构成的长链聚合物)添加到$CsPbI_3$纳米晶体中。乙基纤维素中OH^-和钙钛矿中的I^-形成氢键以及Pb-O之间配位键的协同作用，使其成为相邻钙钛矿卤化物八面体之间的交联剂[图6(f)]，使得钙钛矿薄膜的缺陷态密度明显降低，PLQY从63%增加到87%，抗变形能力和机械稳定性明显增强。基于此制备的FPeLEDs器件具有1674 cd·m^{-2}的最大亮度和12.1%的最大EQE值，以3 mm弯曲半径重复弯曲1000个循环，其效率几乎不变，即使弯曲到1 mm的超小半径，该器件仍展现出十分优异的柔性性能。

量子点(QDs)策略也是提高器件柔性的重要方法，因为传统的钙钛矿发光层存在薄膜厚和晶粒尺寸较大的问题，使其柔性受限，而量子点的小尺寸和稳定性使其在薄膜机械变形过程中仍能保持良好性能。Zhao等[87]使用$CH_3NH_3PbBr_3$量子点制备的发光层在2.5 mm的弯曲半径下进行10次弯曲循环后，仅有<10 nm的细小裂纹出现，而相同基板上的多晶钙钛矿层则出现了不可恢复的微米级裂纹。基于此制得的绿光FPeLEDs分别获得10.4 cd·A^{-1}的CE和2.6%的EQE，在5.5 mm、4 mm和2.5 mm弯曲半径下，器件的亮度从最初的184 cd·m^{-2}，分别下降了15%、16%和20%，恢复平面形状时的亮度为178 cd·m^{-2}，为初始值的97%，展现出十分优异的机械稳定性。同时，在4 mm弯曲半径下弯曲1000次，器件的性能没有发生退化。

除了提高钙钛矿层的机械柔性，赋予钙钛矿层在缺陷/损伤形成后的自修复能力，也是减缓由钙钛矿层弯曲变形导致性能退化的有效方法。Zang等[88]将三甲氧基硅烷(TFPTMS)掺入钙钛矿前驱体，含氟硅烷水解缩合形成三维弹性交联网络，可逆的水解缩合反应和氟化烷基链的存在赋予钙钛矿薄膜优异的机械柔性和自修复能力[图7(a)]。硅氧键与钙钛矿配位，进一步钝化铅离子空位缺陷，显著抑制缺陷诱导的非辐射复合，使得钙钛矿薄膜的PLQY和载流子寿命均明显提升。基于TFPTMS制备的柔性器件最大亮度为11353 cd·m^{-2}，EQE高达16.2%。如图7(b)所示：标件在5 mm弯曲半径下进行500次弯曲循环后，EQE就下降为初始值的10%，并在1000次弯曲循环后就不能被点亮；添加了TFPTMS的器件即使在1000次弯曲循环后仍可以保持初始EQE的75%。Qian等[33]受到穿山甲鳞片的启发，提出一种仿生结构设计，采用二苯基甲烷二异氰酸酯聚氨酯(MDI-PU)聚合物来提升钙钛矿在柔性基板上的结晶性，并赋予器件良好的自修复性能。MDI-PU中携带的氢键软链段(聚醚和聚酯)提供弹性和自修复能力，硬质段(MDI)用于巩固聚合物网络。掺入钙钛矿前体溶液的MDI-PU通过氨基甲酸酯连接脆性钙钛矿晶体，以提高钙钛矿薄膜的柔韧性和弯曲性[图7(c)]，并且通过钝化晶界处的空位缺陷来调节成核并优化钙钛矿薄膜的结晶度。更为重要的是，MDI-PU聚合物本身的分子内/分子间氢键，在加热条件下可以修复反复弯曲过程中诱发的裂纹，实现有效自修复。如图7(d)、(e)所示，基于此制备的天蓝光FPeLEDs实现了13.5%的高效率并表现出高抗弯曲应变能力，在重复弯曲和扭转2000次后分别保持其初始效率的87.8%和80.7%。

3.4 界面工程和能级调控

常用的解决FPeLEDs中电荷注入和传输不平衡问题的方法包括引入缓冲层、对电荷传输层和电极进行掺杂或后处理。这些方法可以减少界面非辐射复合的损失，改善两个结构层之间的能带偏移，从而提高FPeLEDs的效率和稳定性[89-91]。例如，Lee等[92]利用共轭高分子电解质PFN作为电子传输层(SPW-111)和Ag NWs电极之间的界面层，降低了电子的注入势垒，所制备的柔性器件如图8(a)所示。由于Ag NWs电极的功函数较高(4.82 eV)，基于该电极制备的器件的电子注入效率低，无法满足高性能器件的制备需求。PFN同时具有的π共轭有机骨架和离子官能团在PFN/Ag NWs层的形成过程中，疏水性聚合物主链优先位于有机SPW-111层的一侧，而负离子位于亲水性Ag NWs的一侧，形成自发的偶极极化，能够有效降低接触层之间的能垒。由此制得的器件在2 mm弯曲半径下循环弯曲400次，仍能保持初始亮度的80%。

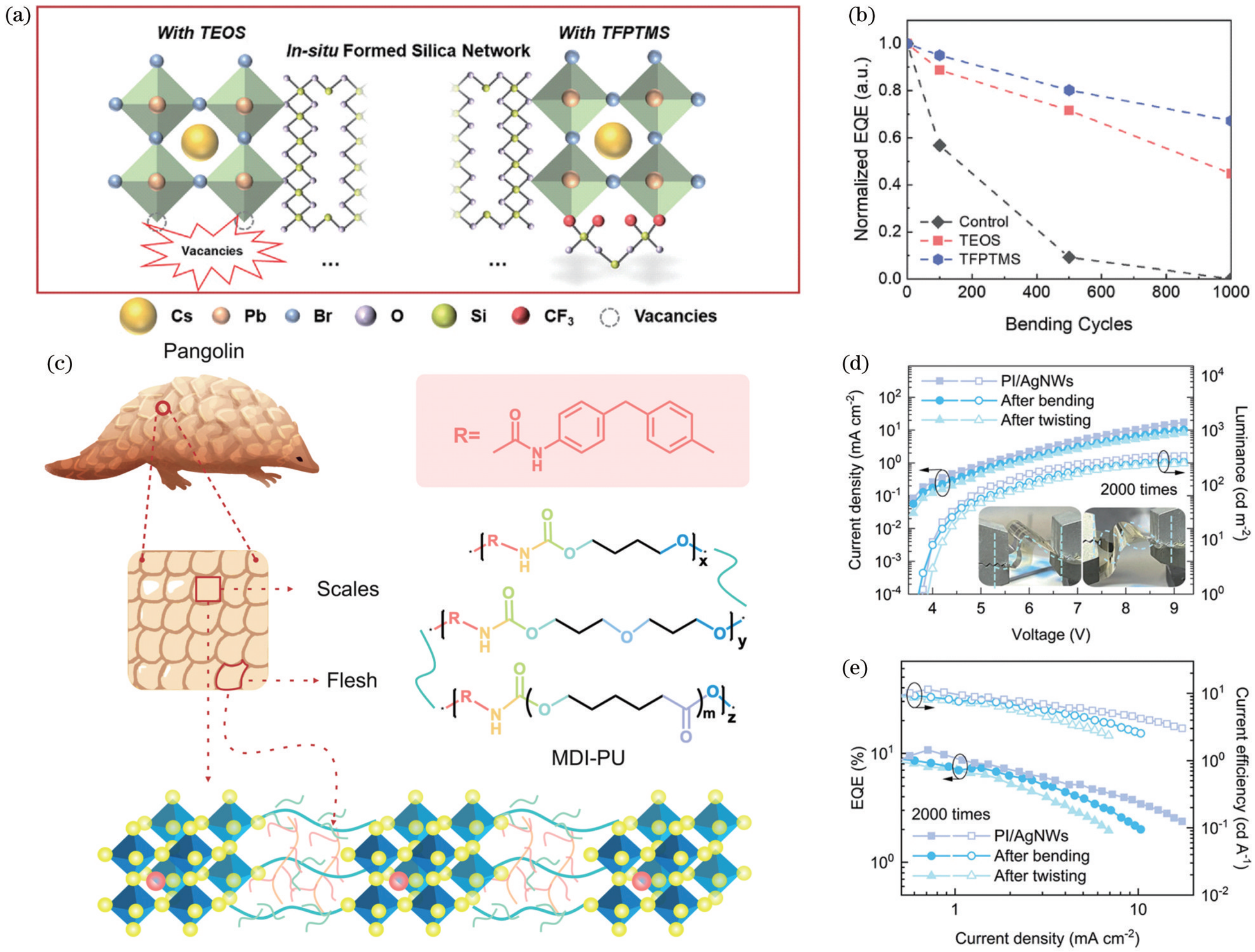

图7 FPeLEDs中钙钛矿薄膜自修复。(a)形成的SiO_2网络以及和钙钛矿相互作用示意图[88];(b)5 mm弯曲半径下不同循环次数的EQE退化图[88];(c)MDI-PU化学结构及钙钛矿晶体和MDI-PU间相互作用示意图[33];(d)电流-电压-亮度曲线,插图为弯曲实验和扭曲实验图片[33];(e)2000次弯曲和扭曲实验前后的EQE-电流曲线图[33]

Fig. 7 Self-healing of perovskite thin films in FPeLEDs. (a) Schematic of formed silica network (left, TEOS; right, TFPTMS) and the interaction with perovskite[88]; (b) degradation diagram of EQE under different cycles with a bending radius of 5 mm[88]; (c) schematic of chemical structure of MDI-PU and interaction between perovskite crystals and MDI-PU[33]; (d) *J*-*V*-*L* characteristics with insets showing the photographs of bending test and twisting test setups[33]; (e) EQE-*J* diagram before and after 2000 bending and twisting experiments[33]

为了抑制发光淬灭并促进PEDOT:PSS/钙钛矿界面处的空穴注入,Lee等[67]使用不同浓度的不导电氟表面活性剂Zonyl FS-300 (Zonyl)处理PEDOT:PSS,以改变空穴传输层的能级。基于PEDOT:PSS空穴传输层的FPeLEDs在不同Zonyl添加浓度下的能带图和*J*—*V*曲线如图8(b)所示。Zonyl的加入降低了空穴传输层和EML之间的能垒,增强了空穴注入,并减少了PEDOT:PSS/钙钛矿界面处的激子淬灭。该方法将器件的CE最大值从16.02 cd·A^{-1}提升到33.18 cd·A^{-1},即使在2.5 mm弯曲半径下循环弯曲1000次,器件效率仍保持不变。此外,Kim等[93]使用异丙醇(IPA)和聚(4-苯乙烯磺酸钠)(PSS-Na),对PEDOT:PSS溶液进行溶剂改性和掺杂剂改性,将空穴传输层的功函数从5.18 eV增大到5.46 eV,降低了PEDOT:PSS和钙钛矿间的能垒,促进了空穴传输,沉积在改性后的空穴层上的钙钛矿薄膜表现出更高的结晶度和更少的发光淬灭,由此制备的柔性器件的最大CE和EQE分别为25.13 cd·A^{-1}和5.91%。

LED发光效率主要受内量子效率和光提取效率两个因素的影响。在PeLEDs中产生的光子并非全部都能发射到自由空间,而是可能会被发射层、电极和衬底所捕获[94-96]。因此,提高光耦合效率成为进一步提高器件性能的关键因素[97]。提升光耦合效率的衍射光栅、低折射率网格和表面粗化等方法已在其他类型的LEDs器件中被广泛使用[98-100]。Shen等[14]提出一种基于合理界面工程的柔性薄膜结构,实现了高效光子的产生和增强的光输出耦合效率。图8(c)展示了FPeLEDs的制备过程,即使用PET基板代替玻璃基板,柔性透明电极由Ag NWs和图案化ZnO组成,ZnO的使用可以避免Ag NWs和钙钛矿层直接接触,提高

器件的运行稳定性。从图8(d)可以看出，图案化器件的最大EQE为24.5%，大约是平面器件的1.4倍，并且展现出良好的重现性。这种效率提升正是由光的输出耦合效率增大导致的。

4 总结与展望

从钙钛矿材料出发，全面介绍了FPeLEDs的器件结构和工作原理，讨论了柔性基板、电极、发光层和界面能级调控等对器件柔性、稳定性和效率等方面的影响，总结了对各功能层性能提升所采用的优化策略和相关研究进展。得益于钙钛矿材料优异的光电特性，FPeLEDs在可穿戴和照明显示领域有着巨大的发展空间，如在不平整或弧形墙体上，硬质LED面临着无法使用或成本增大的难题，而FPeLEDs可以在完美解决这个问题的同时降低生产成本，并可完美适配室外景观项目、艺术造型等特殊应用场景。此外，

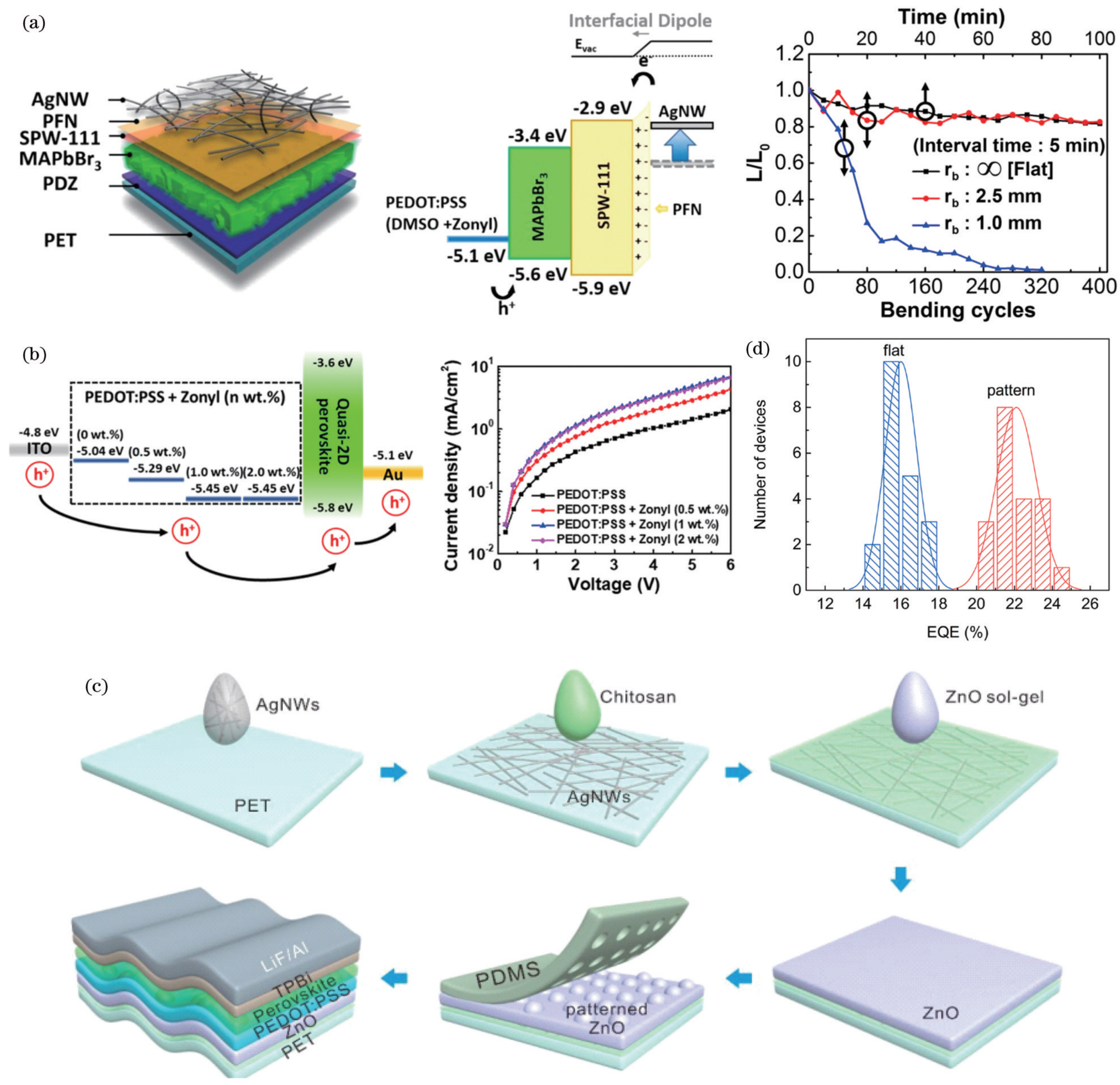

图8 FPeLEDs中的界面工程和能级调控。(a)PFN作界面层的FPeLEDs器件结构、能级结构和不同弯曲半径和弯曲次数下的器件亮度[92]；(b)PEDOT:PSS中添加不同浓度Zonyl的FPeLEDs的能级结构和电流-电压曲线[67]；(c)基于图案化ZnO的柔性透明电极制备过程[14]；(d)平面和图案化柔性透明电极的FPeLEDs的EQE分布统计图[14]

Fig. 8 Interface engineering and energy level regulation in FPeLEDs. (a) FPeLEDs device structure, energy level structure, and device brightness under different bending radii and bending times using PFN as interface layer[92]; (b) energy level structure and *J*-*V* curve of FPeLEDs with different concentrations of Zonyl added to PEDOT : PSS[67]; (c) preparation process of flexible transparent electrodes based on patterned ZnO[14]; (d) EQE distribution statistics of FPeLEDs with planar and patterned flexible transparent electrodes[14]

FPeLEDs的优势激发了许多显示以外的新应用，如可将其制成可穿戴的健康监测传感器、穿戴式体外除颤仪等。

FPeLEDs的实际应用仍存在许多挑战，需进行深入研究。首先是弯曲状态下FPeLEDs性能衰退机制的研究。柔性器件在弯曲和扭曲等变形状态下可能出现薄膜形貌受损、载流子传输损失和发光效率降低等问题。深入研究这些衰退机制有助于理解器件在弯曲状态下的行为，为器件结构设计提供指导，以实现更小弯曲半径下优异的器件性能。此外，需进一步优化FPeLEDs中的各功能层薄膜的设计和制备。这涉及材料选择、涂覆技术、薄膜微纳结构构筑、表/界面处理等方面的研究和创新，目标是在满足柔性要求的同时保持良好的光电特性。各功能之间的黏附性也是一个重要因素，需要探索界面改性、交联技术等方法，以实现膜层之间良好的界面接触。通过以上研究，可以在各功能层薄膜中实现良好的柔性和光电特性之间的平衡，从而提高柔性器件的性能和可靠性，推动其在可穿戴技术、智能电子等领域的实际应用。

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Progress on Flexible Perovskite Light-Emitting Diodes

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Abstract

Significance Metal halide perovskite materials have rapidly developed in optoelectronic devices such as light-emitting diodes, solar cells, lasers, photodetectors, and image sensors due to their advantages of solution processability, high absorption coefficient, tunable bandgap, and long carrier diffusion distance. As a promising electroluminescent material, perovskite materials combining organic and inorganic semiconductor advantages have attracted significant attention in light-emitting diodes (LEDs). Since the first room-temperature perovskite light-emitting diode (PeLED) was introduced in 2014, the external quantum efficiencies (EQE) in the near-infrared, red, and green light regions have exceeded 20%. However, traditional rigid substrate PeLEDs cannot meet the growing demand for flexible display and wearable electronic devices, with emphasis on the need for flexible perovskite LEDs (FPeLEDs).

For practical applications of flexible devices, each layer of FPeLEDs requires sound flexibility and stability, including substrates, electrodes, emitting layers, and interface layers. In 2014, Kim *et al.* achieved the first flexible device with a flexible plastic substrate instead of a rigid glass substrate to realize an EQE of 0.125% with a maximum bending radius of 1.05 cm. Over the years, significant research progress has been made for FPeLEDs. However, their EQE still lags behind rigid glass-based devices, limiting their application in high-performance wearable devices. Therefore, summarizing existing research is necessary to identify challenges and future directions for FPeLEDs' development.

Progress Suitable substrates for FPeLEDs must exhibit excellent flexibility, high transmittance, and sound stability. Various transparent polymer substrates are reported (Table 1). Among them, polyethylene terephthalate (PET) and polyethylene naphthalate (PEN) are commonly employed. However, they suffer from deformation and increased resistance at high temperatures. Flexible polyimide (PI) substrates with high-temperature resistance have been explored with cost limitations. Enhancements in mechanical flexibility and strain release have been achieved by incorporating silver nanowires (Ag NWs) into PI substrates. Additionally, biodegradable substrates and mica with high transparency and flexibility are being developed as alternatives.

Traditional indium tin oxide (ITO) electrodes adopted in rigid devices are not compatible with flexible substrates due to high-temperature deposition requirements. As alternatives, metal electrodes, carbon electrodes, and conductive polymers have been explored (Fig. 4). Metals (such as metal films and metal nanowires) are widely utilized as flexible electrodes in flexible optoelectronic devices due to their high conductivity and good mechanical flexibility. Carbon electrodes, including graphene and carbon nanotubes (CNTs), provide high transparency, carrier mobility, and flexibility. Strategies like passivation layers, chemical post-treatment, and doping have been employed to enhance the conductivity and surface morphology of carbon electrodes. Conductive polymers like PEDOT: PSS are ideal electrode materials due to their conductivity and flexibility. Incorporating solvents or additives can further enhance their conductivity. Composite electrodes that combine different materials have also been developed to realize improved performance compared with single-component electrodes.

Perovskite emissive layers play a critical role in device performance, and their film quality is of utmost importance. Achieving high-performance devices requires well-formed films with uniform grain size. Various deposition methods have been developed to prepare flexible perovskite films, including spin coating, dual-source thermal evaporation, inkjet printing, blade coating, and screen printing, each with its advantages and challenges (Fig. 5). Meanwhile, perovskite thin films typically have dense polycrystalline structure, which limits their flexibility and application in FPeLEDs. To this end, researchers have focused on improving the flexibility of perovskite films through grain size control, micro/nanostructure construction, and physical dispersion/chemical cross-linking (Fig. 6). Additionally, the quantum dot strategies and incorporating self-healing properties in perovskite layers are also discussed.

To address charge injection and transport imbalances in FPeLEDs, methods such as introducing buffer layers,

doping, and post-processing of charge transport layers and electrodes are commonly adopted. These approaches reduce non-radiative recombination losses and improve energy level alignment between layers to enhance FPeLEDs' efficiency and stability. Lee *et al*. employed the conjugated polymer electrolyte PFN as an interface layer between the electron transport layer (SPW-111) and Ag NWs electrode, lowering the electron injection barrier. Their flexible devices maintained 80% initial brightness after 400 bending cycles with a 2 mm radius. Lee *et al*. modified the hole transport layer by Zonyl FS-300 to enhance hole injection and reduce emission quenching at the PEDOT:PSS/perovskite interface. These modifications increased the device's efficiency and maintained it even after 1000 bending cycles with a 2.5 mm radius. In FPeLEDs, not all the generated photons are emitted into free space but captured by emission layers, electrodes, and substrates. Therefore, improving the outcoupling efficiency is a key factor for further improving device performance. Shen *et al*. achieved high-efficiency photon generation and improved light output coupling efficiency by utilizing rational interface engineering and patterned ZnO in a flexible thin-film structure, resulting in devices with an EQE approximately 1.4 times higher than planar devices (Fig. 8).

Conclusions and Prospects We discuss the influence of flexible substrates, electrodes, perovskite emissive layers, and interface energy level alignment on the flexibility, stability, and efficiency of FPeLEDs. We summarize strategies for optimizing the performance of each functional layer. FPeLEDs show significant potential in wearable and display lighting applications, overcoming the limitations of rigid PeLEDs. However, challenges remain, such as studying performance degradation during bending, optimizing thin film design and fabrication, and improving interlayer adhesion. Addressing these challenges will enhance the performance and reliability of FPeLEDs, and realize their practical applications in various fields.

Key words perovskite; flexible perovskite light-emitting diodes; flexible electronics; high efficiency; stability