

Operational and Spectral Stability of Perovskite Light-Emitting Diodes

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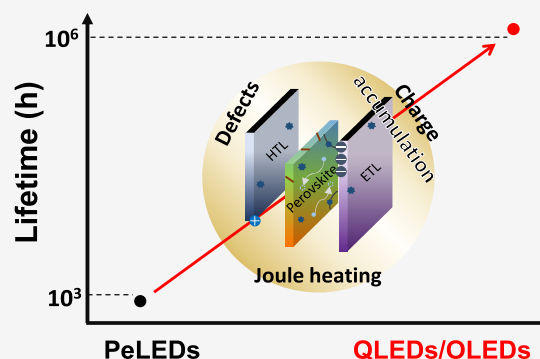
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ABSTRACT: As an emerging light source, perovskite light-emitting diodes (PeLEDs) based on metal halide perovskites have witnessed incredible progress in recent years, and device efficiency is considerably enhanced toward the level of practical applications. However, the stability of the PeLEDs is significantly behind that of commercial light sources. It is imminent to considerably improve the operational stability of the device. In this Review, we first analyze the factors that cause device degradation. Then, we describe the research progress in improving PeLEDs' operational stability via defect passivation, charge balance, and thermal management. Furthermore, the issue of spectral stability is presented. Finally, we discuss the future development directions for achieving high-performing and stable PeLEDs.



Since metal halide perovskites (MHPs) have been proven to be able to serve as the active layer of light-emitting diodes (LEDs) that emit bright light at room temperature through a solution-processing method,¹ researchers have started to investigate the use of MHP materials for light emission. In 2015, MHP quantum dots (QDs) were confirmed to show a high photoluminescence quantum yield (PLQY) and narrow emission full width at half-maximum (FWHM).^{2,3} The excellent optical performance and outstanding processability of the materials at low cost offer opportunities for its future application in display and lighting fields.⁴ Indeed, MHP materials have lived up to expectations. Researchers have boosted the device external quantum efficiency (EQE) of green and red metal halide perovskite LEDs (PeLEDs) from 0.1%¹ and 0.76%¹ to 20.3%⁵ and 20.7%⁶ in less than 4 years. At present, the highest EQEs of blue, green, and red PeLEDs are 12.3%,⁷ 23.4%,⁸ and 21.6%,⁹ respectively, and there is still huge potential for further improvement.¹⁰

Compared to the rapid increase in efficiency, the progress in stability of PeLEDs is still lagging behind. So far, the highest operational stability T50 (the time taken by the device to drop to 50% of its initial luminance or EQE) reported under the initial brightness of 100 cd/m² is 694 h for PeLEDs,¹¹ while those of organic LEDs (OLEDs) and quantum dot LEDs (QLEDs) have exceeded 1,000,000 h.¹² A summary of the operational stability of the PeLEDs is shown in Table 1 and Figure 1a. It is worth noting that a standard way to evaluate the stability of LEDs is based on the brightness rather than current. It will be good to provide such stability results of PeLEDs in the future. It can be seen that the lifetime of the red LEDs is longer than that of the

green ones, while the blue light is far behind the other colors (Figure 1a). This can be understood as wider band gap semiconductor LEDs need to operate at a higher biased voltage. High energy is more likely to cause device heating, which makes the device operate at a high temperature. In addition, the high injection barrier of the wide band gap material is causes the easy accumulation of charge carriers at the interface, which makes the device more prone to degradation.^{13–15} Meanwhile, as we know, MHP materials first showed great application potential in the field of photovoltaics.¹⁶ After more than 11 years of unremitting efforts, metal halide perovskite solar cells (PSCs) are now gradually approaching commercial application. The main obstacle that restricts their commercial use is the long-term stability of MHP materials. Compared with PSCs, PeLEDs work at a higher external bias. This puts higher requirements on the stability of the MHP materials, the rational selection of the transport layer, and the optimization of the device structure.

One advantage of MHPs is that the emission spectrum can be continuously adjusted in the visible light region by simply changing the composition of the halogen,^{17,18} which is conducive to future LED applications with tunable colors. However, the mixed halide MHP is prone to halide phase

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Table 1. Performance of RGB PeLEDs

material	device structure	EL peak (nm)	EQE _{max} (%)	lifetime (h) ^a	measurement conditions	ref
Red–Near-IR						
NFPI ₆ B	ITO/ZnO/PEIE/MHP/TFB/MoO _x /Au	763	11.7	2 ^{M,G}	At 10 mA/cm ² EQE = 0.5EQE _{max}	2016 ²⁶
NCPI ₆ Cl	ITO/ZnO/PEIE/MHP/TFB/MoO _x /Au	688	3.7	~5 ^{M,G}	At 10 mA/cm ² EQE = 0.5 EQE _{max}	2017 ²⁷
FPMAl-MAPbI ₃	ITO/Poly-TPD/MHP/TPBi/LiF/Al	749	7.9	>10 ^{M,G}	At 3 mA/cm ² EQE > EQE ₀	2017 ²⁸
Cs ₁₀ (MA _{0.17} FA _{0.83}) _(100-x) PbBr _{0.33} I _{0.67}	ITO/AZO/AZO:Cs/MHP/CuS-GaSnO/WO ₃ /Au	650	8.21	491 ^{M,E}	At 1.76 mA/cm ² 1000 cd/m ² decrease to 100 cd/m ²	2018 ²⁹
An-HI-CsPb(Br/I) ₃	ITO/PEDOT:PSS/Poly-TPD/MHP/TPBi/Liq/Al	645	14.1	3 ^{M,E}	At 1.25 mA/cm ² L ₀ = 100 cd/m ²	2018 ¹⁸
BA ₂ MA _{n-1} Pb _n I _{3n+1}	ITO/PEDOT:PSS/PCBM/MHP/TPD/BCP/TPBi/Al	744	0.4	>14 ^{M,E}	At 2 V	2018 ³⁰
Poly-HEMA-(NMA) ₂ (FA) _{n-1} Pb _n I _{3n+1}	ITO/MZO/PEIE/MHP/TFB:PFO/MoO _x /Au	790	20.1	46 ^{M,E}	At 0.1 mA/cm ²	2018 ³¹
PEO:BA ₂ Cs _{n-1} Pb _n I _{3n+1}	ITO/PEDOT:PSS/Poly-TPD/MHP/BCP/LiF/Al	680	6.23	4 ^{M,E}	At 3.5 V	2018 ³²
NF _{0.93} C _{0.07} PI ₇	ITO/ZnO/PEIE/MHP/TFB/MoO _x /Au	783	6.6	31 ^{M,G}	At 10 mA/cm ²	2018 ³³
SAVA-FAPbI ₃	ITO/ZnO/PEIE/MHP/TFB/MoO _x /Au	803	20.7	20 ^{M,E}	At 100 mA/cm ² EQE = 0.5 EQE _{max}	2018 ⁶
(PEAI) _x (CsI) _y (SnI ₂) _z	ITO/PVK/MHP/TMPyPb/LiF/Al	920	3	2 ^{M,G}	At 10 mA/cm ² EQE = 0.5 EQE _{max}	2019 ³⁴
PA ₂ CsPb ₂ I ₇	ITO/PEDOT:PSS/MHP/TPBi/LiF/Al	691	0.97	11 ^{M,G}	At 10 mA/cm ²	2019 ³⁵
ODEA-FAPbI ₃	ITO/ZnO/PEIE/MHP/TFB/MoO _x /Au	800	21.6	20 ^{M,G}	At 25 mA/cm ²	2019 ⁹
BA-FAPbI ₃	ITO/ZnO/PEIE/MHP/TFB/MoO _x /Au	804	14	23.7 ^{M,G}	At 100 mA/cm ²	2019 ³⁶
BA-MAPb(I/Br) ₃	ITO/Poly-TPD/MHP/Bphen/Liq/Ag	755	16.7	47 ^{M,G}	At 10 mA/cm ²	2019 ³⁷
BAB-FAPbI ₃	ITO/ZnO/PEIE/MHP/TFB/MoO ₃ /Al	776	5.2	100 ^{M,G}	At 25 mA/cm ² EQE = 0.5 EQE _{max}	2019 ³⁸
FA _{0.98} Cs _{0.02} PbI ₃ /(PEA) ₂ PbI ₄	ITO/ZnO/MHP/Poly-TPD/MoO ₃ /Ag	~800	7.7	~202.7 ^{C, G}	At constant voltage (initial radiance ≈ 10 W sr ⁻¹ m ⁻²)	2020 ³⁹
SAVA/Pb(SCN) ₂ -Cs _{0.17} FA _{0.83} PbI ₃	ITO/ZnO/PEIE/MHP/TFB/MoO _x /Au	780	15.01	11.3 ^{M,G}	At 100 mA/cm ²	2020 ¹³
DDS-FAPbI ₃	ITO/ZnO/PEIE/MHP/TFB/MoO _x /Au	802	17.3	100 ^{M,G}	At 20 mA/cm ²	2020 ⁴⁰
CsPbI ₃	ITO/PEDOT:PSS/PVK/MHP/ZnO/Ag	620	6.2	694 ^{C, E}	At 150 mA/cm ²	2020 ¹¹
PEA ₂ SnI ₄	ITO/PEDOT:PSS/MHP/TPBi/LiF/Al	632	5	15 ^{M,G}	20 cd/m ² decrease to 10 cd/m ²	2020 ⁴¹
SAVA/Pb(SCN) ₂ /xFAI:Cs _{0.17} FA _{0.83} PbI _{2.5} Br _{0.5}	ITO/ZnO/PEIE/MHP/TFB/MoO _x /Au	~756	17.4	20 ^{M,G}	At 10 mA/cm ²	2020 ⁴²
FPMATFA-FA _{0.33} Cs _{0.67} Pb(I _{0.7} Br _{0.3}) ₃	ITO/Poly-TPD/MHP/TPBi/LiF/Al	694	20.9	14 ^{M,G}	At 2.5 mA/cm ² L ₀ = 25 cd/m ²	2020 ⁴³
4-F-PMAI:CsPb I _{2.8} Br _{0.2}	ITO/Poly-TPD/MHP/TPBi/LiF/Al	689	18.6	20 ^{M,G}	At 5 mA/cm ² L ₀ = 100 cd/m ²	2020 ⁴⁴
GAI-(FA/Cs)PbI _x Br _{3-x}	ITO/ZnO/PEIE/MHP/TFB/MoO _x /Al	692	17.1	~9.38 ^{M,E}	At constant current density L ₀ = 1000 cd/m ²	2021 ⁴⁵
FA _{0.83} Cs _{0.17} PbI ₃	ITO/ZnO/PEIE/MHP/TFB/MoO _x /Au	789	17.5	130 ^{M,G}	At 100 mA/cm ²	2021 ⁴⁶
PMA-CsPbI ₃	ITO/PEDOT:PSS/Poly-TPD/MHP/TPBi/LiF/Al	689	17.8	317 ^{M,G}	At 30 mA/cm ²	2021 ⁴⁷
PAC-FAPbI ₃	ITO/ZnO/PEIE/MHP/TFB/MoO _x /Au	802	18.6	682 ^{M,G}	At 20 mA/cm ²	2021 ⁴⁸
Green						
MAPbBr ₃	ITO/TiO ₂ /EA/MHP/SPB-02T/MoO ₃ /Au	530	0.051	55 ^{M,E}	At 4.8 V L ₀ = 120 cd/m ²	2015 ⁴⁹
CsBr:CsPbBr ₃	ITO/PEDOT:PSS/MHP/B3PYMPM/CsCO ₃ /Al	524	0.15	15 ^{M,E}	At 66.67 mA/cm ² L > L ₀ = 100 cd/m ²	2016 ⁵⁰

Table 1. continued

material	device structure	EL peak (nm)	EQE _{max} (%)	lifetime (h) ^a	measurement conditions	ref
Green						
Cs ₁₀ (MA _{0.17} FA _{0.83}) _(100-x) PbBr _{2.97} I _{0.03}	ITO/ZnO/MHP/NPD/MoO ₃ /Al	569	7.3	7.9 ^{M,E}	At 3 mA/cm ² 1335.7 cd/m ² decrease to 800.7 cd/m ²	2017 ⁵¹
CF-PEO:CsPbBr ₃	ITO/PEDOT:PSS/MHP/TPBi/LiF/Al	525	4.76	80 ^{M,E}	L ₀ = 1000 cd/m ² L = 82%L ₀	2017 ⁵²
CsPbBr ₃	Au/p-MgNiO/MHP/PMMA/n-MgZnO/n ⁺ -GaN	522	2.39	10 ^{M,w/o E}	At 10 V L = 80% L ₀	2017 ⁵³
Cs ₁₀ (MA _{0.17} FA _{0.83}) _(100-x) PbBr _{2.97} I _{0.03}	ITO/AZO/AZO:Cs/MHP/CuS-GaSnO/WO ₃ /Au	569	12.98	173 ^{M,E}	At 1.42 mA/cm ² 1000 cd/m ² decrease to 100 cd/m ²	2018 ²⁹
Tween 20:CsPbBr ₃	ITO/PEDOT:PSS/MHP/TPBi/LiF/Al	522	7.39	6 ^{M,w/o E}	At constant current density L ₀ = 100 cd/m ²	2018 ⁵⁴
CsPbBr ₃ /MABr	ITO/PEDOT:PSS/MHP/PMMA/B3PYMPM/LiF/Al	525	20.3	104.56 ^{C, G}	At 166.67 mA/cm ² L ₀ = 100 cd/m ²	2018 ⁵
CsPbBr ₃	Au/MgO/MHP/n-MgZnO/n ⁺ -GaN/c-Al ₂ O ₃	538	1.46	18 ^{M,E}	At 10 V L = 56.1%L ₀	2018 ⁵⁵
CsPbBr ₃	Au/NiO/MHP/MgZnO/Au NPs/n-ZnO NWs/In	518	4.63	60 ^{M,w/o E}	At 8 V L = 85.7%L ₀	2018 ⁵⁶
mPEG-CsPbBr ₃	ITO/m-PEDOT:PSS/MHP/Bphen/LiF/Ag	524	0.9	24 ^{M,E}	At constant current density L > L ₀ = 100 cd/m ²	2018 ⁵⁷
CsPbBr ₃	ITO/NiO/MHP/ZnO/Al	519	3.79	12 ^{M,w/o E}	At 8 V L = 81.8%L ₀	2018 ⁵⁸
PEA ₂ Cs _{n-1} Pb _n Br _{3n+1}	ITO/CQDs/MHP/TPBi/LiF/Al	514	13.8	~6.67 ^{M,G}	At 0.5 mA/cm ² L ₀ = 100 cd/m ²	2019 ⁵⁹
BM ₂ (Cs _{1-x} MA _x PbBr ₃) _{n-1} PbBr ₄	ITO/Poly-TPD/MHP/TPBi/Ca/Ag	516	5.2	24.8 ^{C, E}	At 100 mA/cm ² L ₀ = 100 cd/m ²	2019 ⁶⁰
TFA-CsPbBr ₃	ITO/PEDOT:PSS/MHP/TPBi/LiF/Al	518	10.5	250 ^{M,E}	L ₀ = 100 cd/m ²	2019 ⁶¹
(Cs _{0.83} Rb _{0.17}) _{0.95} K _{0.05} PbBr ₃	ITO/LiF/MHP/LiF/ZnS/ZnSe/Ag	519	11.05	255 ^{M,G}	L ₀ = 120 cd/m ²	2020 ⁶²
CsPbBr ₃	ITO/3AP-PEDOT:PSS/MHP/TPBi/LiF/Al	517	9.4	6.1 ^{M,E}	L ₀ = 100 cd/m ²	2020 ⁶³
CsPbBr ₃	ITO/PEDOT:PSS/PTAA/MHP/TPBi/PO-T2T/LiF/Al	520	21.63	180.1 ^{M,G}	At 6 mA/cm ² L ₀ = 100 cd/m ²	2020 ⁶⁴
CsPbBr ₃	ITO/PEDOT:PSS/PTAA/TSPO1/MHP/TSPO1/TPBi/LiF/Al	516	18.7	15.8 ^{M,G}	L ₀ = 100 cd/m ²	2020 ⁶⁵
Cyclam-PEA ₂ Cs _{n-1} Pb _n Br _{3n+1}	ITO/PVK-F4TCNQ/MHP/TPBi/LiF/Al	513	16.24	127 ^{C, G}	At 10 mA/cm ² L ₀ = 100 cd/m ²	2020 ⁶⁶
LiBr-PEA ₂ Cs _{n-1} Pb _n Br _{3n+1}	ITO/NiO _x /TFB/PVK/MHP/TPBi/LiF/Al	520	16.2	21 ^{M,E}	L ₀ = 100 cd/m ²	2020 ⁶⁷
PEG-PEA ₂ Cs _{n-1} Pb _n Br _{3n+1}	AgNWs/ZnO/ETA-PEDOT:PSS/MHP/TPBi/LiF/Al	514	17.1	~7.56 ^{M,E}	L ₀ = 100 cd/m ²	2020 ⁶⁸
MABr:CsPbBr ₃	ITO/PEDOT:PSS/MHP/TPBi/LiF/Al	530	16	~0.62 ^{M,w/o E}	At 4 V L _{max} = 10000 cd/m ²	2020 ⁶⁹
FA _{0.9} GA _{0.1} PbBr ₃	ITO/Buf-HIL/MHP/TBTB/TPBi/LiF/Al	531	23.4	2.2 ^{M,E}	L ₀ = 100 cd/m ²	2021 ⁸
CsPbBr ₃	ITO/PEDOT:PSS/MHP/B3PYMPM/Cs ₂ CO ₃ /Al	522	16.45	136 ^{C, G}	At 50 mA/cm ² L ₀ = 100 cd/m ²	2021 ⁷⁰
K ₂ S ₂ O ₈ -MBA-PEA ₂ Cs _{n-1} Pb _n Br _{3n+1}	ITO/PVK/F4TCNQ/MHP/TPBi/LiF/Al	516	16.8	208 ^{M,E}	At 0.2 mA/cm ² L ₀ = 100 cd/m ²	2021 ⁷¹
Blue						
Cs ₁₀ (MA _{0.17} FA _{0.83}) _(100-x) PbCl _{1.5} Br _{1.5}	ITO/ZnO/MHP/NPD/MoO ₃ /Al	475	1.7	2.5 ^{M,E}	At 3 mA/cm ² 948.5 cd/m ² decrease to 88.5 cd/m ²	2017 ⁵¹
Cs ₁₀ (MA _{0.17} FA _{0.83}) _(100-x) PbCl _{1.5} Br _{1.5}	ITO/AZO/AZO:Cs/MHP/CuS-GaSnO/WO ₃ /Au	475	2.58	16 ^{M,E}	At 2.02 mA/cm ² 1114 cd/m ² decrease to 0 cd/m ²	2018 ²⁹
CsPbCl _{0.9} Br _{2.1} + 100% PEABr	ITO/PEDOT:PSS/MHP/TPBi/LiF/Al	480	5.7	0.17 ^{M,G}	At 4.4 V	2019 ⁷²

Table 1. continued

material	device structure	EL peak (nm)	EQE _{max} (%)	lifetime (h) ^a	measurement conditions	ref
Blue						
PBABr ₇ (Cs _{0.7} FA _{0.3} PbBr ₃)	ITO/NiO _x /TFB/PVK/MHP/TPBi/LiF/Al	483	9.5	0.07 ^{M,G}	At 1 mA/cm ² L ₀ = 100 cd/m ²	2019 ⁷³
(P-PDA/PEA) ₂ Cs _{n-1} Pb _n Br _{3n+1}	ITO/PVK/PFI/MHP/3TPYMB/Liq/Al	465	2.6	0.23 ^{M,E}	At 0.35 mA/cm ² L ₀ = 100 cd/m ²	2019 ⁷⁴
(PEA) ₂ PbBr ₄	ITO/PEDOT:PSS/PVK/TAPC/MHP/TPBi/Ca/Ag	408.8	0.31	28 ^{M,E}	At 5 V EQE = 0.8EQE _{max}	2019 ⁷⁵
PEA _{0.2} Cs _{0.4} MA _{0.6} Pb _(1-y) Mn _y (Br/Cl) ₃	ITO/PEDOT:PSS/PFI/MHP/TPBi/LiF/Al	~495	0.58	0.4 ^{M,E}	At 3 mA/cm ²	2019 ⁷⁶
PEA ₂ (Rb _{0.6} Cs _{0.4}) ₂ Pb ₃ Br ₁₀	ITO/PEDOT:PSS/MHP/TmPyPB/LiF/Al	475	1.35	0.24 ^{M,G}	At 4.5 V	2019 ⁷⁷
CsPbBr ₃ :PEACl:2% YCl ₃	ITO/PEDOT:PSS/MHP/TPBi/LiF/Al	485	11	<1.83 ^{M,G}	At 3.2 V	2019 ⁷⁸
PEA ₂ FA ₂ Pb ₃ Br ₇ Cl ₃	ITO/MPS2-TBA/MHP/TPBi/LiF/Al	489	2.64	~0.28 ^{M,E}	At 0.5 mA/cm ²	2020 ⁷⁹
PEA ₂ (Cs _{1-x} EA _x PbBr ₃) ₂ PbBr ₄	ITO/m-PEDOT:PSS/MHP/TPBi/LiF/Al	488	12.1	<0.07 ^{M,G}	At 1.5 mA/cm ² L ₀ = 100 cd/m ²	2020 ⁸⁰
Na-PEA ₂ Cs _{n-1} Pb _n (Br/Cl) _{3n+1}	ITO/NiO _x /PTAA/PVK/MHP/TPBi/LiF/Al	488	11.7	<0.16 ^{M,w/o E}	L ₀ = 100 cd/m ²	2020 ⁸¹
PEA ₂ Cs _{1.6} MA _{0.4} Pb ₃ Br _{10-x} Cl _x :DPOCl	ITO/m-PEDOT:PSS/Poly-TPD/MHP/TPBi/LiF/Al	479	5.2	1.5 ^{M,E}	L ₀ = 100 cd/m ²	2020 ²²
(PEA/PA) ₂ (CsPbBr ₃) _{n-1} PbBr ₄	ITO/NiO _x /MHP/TPBi/LiF/Al	488	7.51	1.1 ^{M,E}	At 1.5 mA/cm ²	2020 ⁸²
(Cs/Rb/FA/PEA/K)Pb(Cl/Br) ₃	ITO/LiF/MHP/LiF/Bphen/LiF/Al	484	2.01	5 ^{M,G}	At 25 mA/cm ²	2020 ⁸³
(PEA/NPA) ₂ (CsPbBr ₃) _{n-1} PbBr ₄	ITO/PVK/MHP/PO-T2T/Liq/Al	485	2.62	~0.07 ^{M,G}	At 3.4 V L ₀ = 100 cd/m ²	2020 ⁸⁴
(Cs/FA/p-F-PEA)Pb(Cl/Br) ₃	ITO/m-PEDOT:PSS/MHP/TPBi/LiF/Al	469	4.14	~0.23 ^{M,E}	At 1 mA/cm ²	2020 ⁸⁵
(PEA/PA/ABA) ₂ (CsPbBr ₃) _{n-1} PbBr ₄	ITO/PVK/MHP/TPBi/LiF/Al	486	10.11	~1.36 ^{M,E}	At 0.3 mA/cm ²	2020 ⁸⁶
Zn-CsPbBr ₃	ITO/PEDOT:PSS/PTAA/MHP/TPBi/LiF/Al	479	12.3	~0.33 ^{M,E}	90 cd/m ² decrease to 45 cd/m ²	2020 ⁷
Tween-TPPB-CsPb(Br _{0.65} Cl _{0.35}) ₃	ITO/PEDOT:PSS/MHP/TmPyPB/LiF/Al	482	4.1	~0.68 ^{M,G}	At 3 mA/cm ²	2020 ²³
DPPA/PEA-CsPbClBr ₂	ITO/PEDOT:PSS/TFB/MHP/TPBi/LiF/Al	473	8.8	~0.11 ^{M,G}	At 1.95 mA/cm ² L ₀ = 100 cd/m ²	2020 ⁸⁷
BI ₂ PbBr ₄	ITO/PEDOT:PSS/Poly-TPD/MHP/TPBi/LiF/Al	445	3.08	3.5 ^{M,G}	At 5 V	2021 ⁸⁸
TTDDA-Rb-CsPb(Br/Cl) ₃	ITO/NiO _x /PVK/PVP/MHP/TPBi/LiF/Al	477	11	~0.05 ^{M,G}	At 1.34 mA/cm ² 102 cd/m ² decrease to 51 cd/m ²	2021 ⁸⁹
CsPbBr ₃	ITO/PEDOT:PSS/PVK/MHP/ZnO/Ag	470	4.7	12 ^{M,G}	At 12.5 mA/cm ² 102 cd/m ² decrease to 51 cd/m ²	2021 ⁹⁰

^aSuperscripts: C, calculated value; M, measured value; G, test in glovebox; E (or w/o E), test in air with (or without) encapsulation.

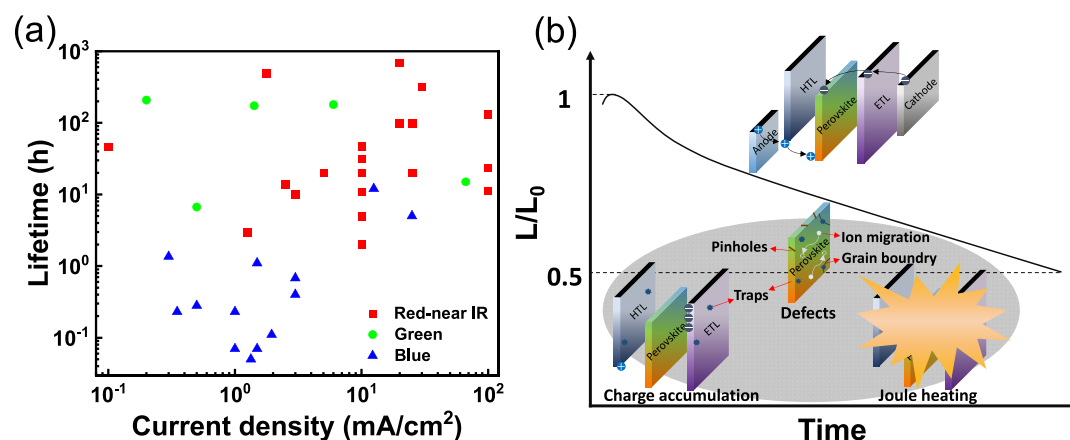


Figure 1. (a) Lifetime of RGB PeLEDs tested at different constant current density (the data were extracted from Table 1). (b) Schematic illustration of the instability of the PeLEDs.

separation, resulting in instability of the emission spectrum.¹⁹ Under external forward bias, the phenomenon of “halide segregation” will not only cause the color purity of the device to deteriorate but also introduce more defect sites, which will adversely affect the efficiency and stability of the PeLEDs.^{20,21} Recently, a series of strategies have been proposed to address the spectral stability issue of Br/Cl mixed and Br/I mixed PeLEDs, including halide insertion–immobilization,²² halide homogenization,²³ and ligand engineering.²⁴ The results showed that the electroluminescence (EL) exhibited good spectral stability under different applied voltages and continuous current bias. Overall, the long-term spectral stability of the mixed-halide PeLEDs needs to be further investigated and improved.

Overall, the long-term spectral stability of the mixed-halide PeLEDs needs to be further investigated and improved.

Here, we first analyze the factors that affect the operational stability of PeLEDs in terms of the three aspects of defects, charge accumulation, and Joule heating. Then we review the research progress in these three areas. Furthermore, we discuss the spectral stability, particularly for the mixed halide MHPs which suffer from phase separation,²⁵ and thus have concerns about their spectral stability.²⁰ In addition, we summarize the recent breakthroughs in device stability and provide an outlook on promising ways to improve the stabilities in the future.

■ ORIGINS OF DEVICE DEGRADATION

MHPs can be tuned from a bulk material to a nanoscale dimension material (i.e., quantum dots (QDs)) by reducing the physical dimension of the MHP. Also, MHP can change from three-dimensional (3D) to quasi-two-dimensional (quasi-2D) or two-dimensional (2D) MHP by reducing the structural dimensionality. 3D MHP and QDs are generally represented by the formula ABX_3 , where A-site (e.g., cesium, methylammonium (MA), and formamidinium (FA)) and B-site (e.g., lead and tin) are cations with different sizes, as well as X-site (e.g., chlorine, bromine, and iodine) is a halogen anion. Quasi-2D or 2D MHP structure generally have the formula $C_mA_{n-1}B_nX_{3n+1}$, where C-site is generally a large-sized monovalent ($m = 2$) or divalent ($m = 1$) cation.^{89–91}

As shown in Figure 1b, in the device stability test, under constant current, the brightness or EQE of PeLEDs is often observed an overshoot in the initial stage and followed by reduction.^{64,71,92} Zao et al.⁹² explained the phenomenon through experiments that the overshoot is caused by the migration of excess mobile ions to fill up the local defects, and the descending process was due to the migration of ions that make up the crystal lattice, which caused more defects.⁹³ The stability of the PeLEDs are mainly affected by (i) the defects in MHP layer,^{92,23,38,40,43,44,52,66,67,74–77,82,83,86,89,94–103} and interface,^{59,65,79,85,104–107} (ii) the charge accumulation,^{37,64,108–114} and (iii) Joule heating.^{13–15,115,116} Due to the small defect formation energy of MHP with soft-lattice, defect sites generally exist in the MHP film.¹¹⁷ For 3D and quasi-2D or 2D MHP, there are two main classes of defects will be formed in a polycrystalline film during the crystallization process of the precursor into the thin film. One is the point defects which include vacancies, interstitial defects, and anti-site defects. The other is the structural defects including dislocations and grain boundaries. Complex defects including Schottky defects and

Frankel defects will also exist.¹¹⁷ For MHP QDs, different from the core–shell structure of traditional QDs (i.e., CdSe/ZnS), the chemical groups of the defect sites on the surface of MHP QDs are simply bonded with the chemical groups of the surface organic ligands. The dynamic nature of the ligand binding, the low formation energy, and the high ionicity of the bonding lead to the instability of MHP QDs.¹¹⁸ The aggregation of the residual insulated organic compounds and the QDs in highly concentrated dispersion or during film formation lead to poor electrical conductivity and low PLQY of MHP QD films.^{119,120} In the QD films, due to the shedding of weakly interacting ligands, a lot of defects exist at the boundaries of QD particles. Overall, the defects exist in the MHP crystal interior, crystal surface, and grain boundaries.^{121,122} When electrons are injected, these defect sites will serve as non-radiative recombination centers. Meanwhile, the electron energy will be converted into heat energy, which will lead to the decomposition of the MHP.¹¹⁶

The ion migration caused by the electric field is considered to be the main reason that affects the stability of the PeLEDs.^{92,121,123} The activation energy (E_a) of ion migration is an important factor to determine whether ion migration occurs. For MHP, the value of E_a is low. Theoretical and experimental results show that the E_a values of halide ions are lower than those of A- and B-site cations,^{71,124–126} in the range of 0.1–0.6 eV. Therefore, during external forward bias, it is easy to induce the migration of halide ions, which will then cause the redistribution of defect sites,¹²⁷ thereby causing photoluminescence (PL) quenching. In addition, in the mixed halide MHP, the migration of halide can also cause the instability of the emission spectrum.¹²⁸ It is worth noting that although a series of strategies have been used to increase the E_a (e.g., introducing A- or B-site cation doping to enhance the bonding strength,^{129,130} preparing the 2D or quasi-2D structure to use organic cations between layers to hinder ion migration,^{131,132} and adopting cross-linking method to form a strong passivation layer around the crystal grains to hinder ion migration⁷¹), their values do not exceed 1 eV. Considering the higher bias voltage of PeLEDs, the boost is far from enough. This may be the main reason why the operational stability of PeLEDs is difficult to increase substantially. We refer the reader to references discussing ion migration.¹³³ In the future, more in-depth research work is needed to increase the E_a .

The role of the charge transport layer is very important in the entire device.¹³⁴ The difference in the mobility of the hole and electron transport layer, as well as the energy level matching with the active layer, which greatly affects the balance of carrier injection. Unbalanced carrier injection will cause the accumulation of carriers at the interface and luminescence quenching.¹³⁵ Meanwhile, the defect sites in the interface also seriously affect the performance of the PeLEDs. For example, oxygen vacancies in metal oxides will cause the PL quenching of MHP film.¹⁰⁷ Joule heat will accelerate the ion migration of the MHP layer and cause the degradation of the interface layer.¹⁴ It is also considered to be the main factor causing the device efficiency roll-off.¹⁵

■ STRATEGIES TO IMPROVE OPERATIONAL STABILITY OF PeLEDs

Adjusting the Goldschmidt tolerance factor (TF) has been regarded as an effective strategy to improve the structural stability of MHP materials and avoid the introduction of defect sites due to the lattice deformation.^{136,137} Tuning the

Reducing Defects

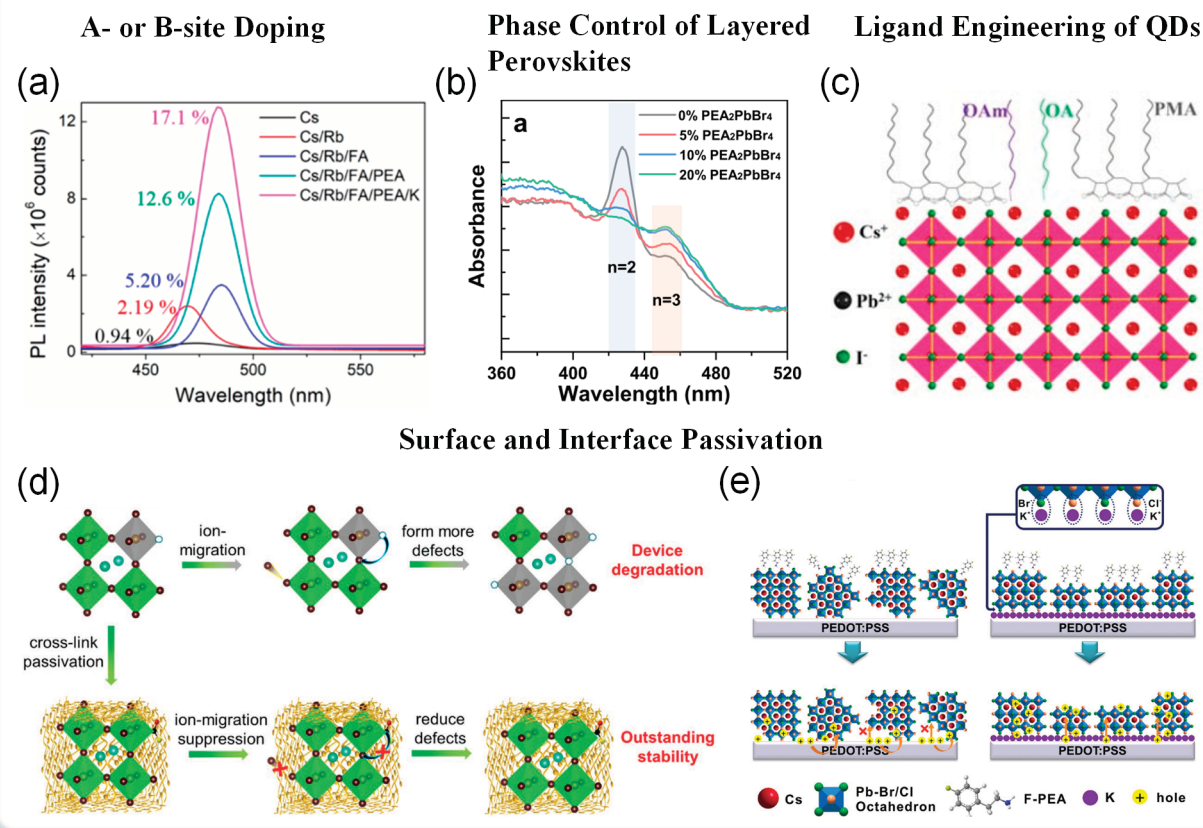


Figure 2. (a) PL spectra and the PLQY of cocktail multi-cation $\text{APb}(\text{Cl}/\text{Br})_3$ perovskite film. Reproduced with permission from ref 83. Copyright 2020 American Chemical Society. (b) UV–visible absorption spectra of $\text{PEA}_2\text{PbBr}_4$ incorporated perovskite films. Reproduced with permission from ref 82. Copyright 2020 American Chemical Society. (c) Schematic diagram of the PMA incorporated CsPbI_3 NCs (OA = oleic acid, OAm = oleylamine). Reproduced with permission from ref 47. Copyright 2021 Wiley-VCH. (d) Mechanism diagram of the improvement of device operational stability by cross-link passivation. Reproduced with permission from ref 71. Copyright 2021 Wiley-VCH. (e) Schematic illustration of the K^+ -guided grain growth and hole transport of perovskite films. Reproduced with permission from ref 85. Copyright 2021 Wiley-VCH.

composition of the A-site cation is a common way to change TF. As shown in Figure 2a, Yuan et al.⁸³ employed rubidium, FA, phenylethylammonium (PEA), and potassium cations into the $\text{CsPb}(\text{Cl}/\text{Br})_3$ MHP, the PLQY of the quasi-2D film increased from 0.94% to 17.1%. They tested the operational stability at a constant current density of 25 mA/cm^2 . The Cs/Rb/FA/PEA/K-based device exhibited a lifetime T50 of 5 h, while the Cs/Rb-based lifetime was less than 0.34 h. Their results showed that each introduction of a cation would cause the PLQY of the film to increase, which points out the possible direction for further optimizing the performance of the current single cation device. Partial substitution of Pb^{2+} ions has also been confirmed to enhance the formation energy of MHP films, thereby significantly reducing the degradation.¹³⁸ For MHP QDs, introducing different ions (i.e., Mn^{2+} , Ce^{3+} , Sn^{4+} , Cu^{2+} , and La^{3+}) into the host of the MHP lattice have been proven as the effective mean to effectively improve the stability and PLQY.^{99,139–142} For example, Zhang et al.⁹⁹ doped Cu^{2+} into CsPbBrI_2 QDs to prepare red PeLEDs. When 3.7 mol% Cu^{2+} ions were doped, the absolute value of the formation energy (ΔE_{form}) of MHP increased from 13.122 to 13.161 eV. The doping of Cu^{2+} ions caused a slight lattice contraction, and hence stabilized the cubic phase of these QDs. Combining Cu doping and halogen-rich synthesis, the PLQY of QDs reached 94.8%.

The operational stability of the device has also been significantly improved.

Compared with 3D MHP, considerable research has been conducted on layered (2D or quasi-2D) MHP which show better stability,¹³⁷ faster radiative recombination rates,¹⁴³ and less susceptibility to ion migration.¹⁴⁴ While the multi-phase layered film will lead to wide emission bandwidth, which reduces the color purity. Meanwhile, random and wide phase distribution is prone to cause inefficient internal energy transfer.^{81,82,145} Tam et al.¹⁴⁶ systematically studied the influence of the length of the alkyl chain in phenyl-alkyl spacer cations on the phase distribution. They employed benzylammonium (BZA) with one carbon atom, PEA with two carbon atoms, and phenylpropylammonium (PPA) with three carbon atoms to prepare quasi-2D films. The lattice-parameters obtained from the computational relaxation were in good agreement with the diffraction reflection of 002 at $d = 26.84 \text{ \AA}$. The aromatic rings of PPA showed non-parallel arrangement, indicating a weakened π – π interaction and enhanced $\text{CH}\cdots\pi$ interactions, which could suppress the formation of the $n = 1$ phase. Mixed C-site cations are proven to effectively control phase purity. As shown in Figure 2b, Choy et al.⁸² introduced PEA cations into $\text{PA}_2(\text{CsPbBr}_3)_{n-1}\text{PbBr}_4$ MHP (PA = propylamine), the first-principles total energy (TE) calculations results showed that the

$n = 2$ phase is thermodynamically unstable after introducing PEA, and $n = 3$ phase is more favorable to form in the film.⁸² The transient absorption (TA) results indicated the dominant $n = 3$ phase after treatment. The PLQY of the treated film increased from 39.5% to 49.6%, and the sky blue PeLEDs showed long operational lifetime of 3961 s at a constant current density of 1.5 mA/cm². In addition, Wang et al.¹⁴⁷ proposed to introduce a chelating agent to help control the growth kinetics of quantum wells (QWs) and thus limit the n value, stabilizing the formed phase. They employed γ -aminobutyric acid (GABA) to coordinate with Pb and destabilized the Pb sites to inhibit the growth of thicker layers. The TA results showed that the GABA-treated films exhibited a narrow and stable distribution of QWs. The GABA-treated films exhibited higher PLQY of 82% than the control film (40%). The GABA-treated device showed a T50 of 150 s at an initial luminance of 100 cd/m², which is an order of magnitude larger than the control device.

As mentioned earlier, the selection and regulation of surface ligands play a crucial role in the optoelectronic properties of MHP QDs.^{148,149} Traditional ligands (such as oleic acid (OA) and oleylamine (OAm)) have weak binding energy to QDs, which cannot guarantee the long-term stability of QDs. Baek et al.¹⁵⁰ used the trioctylphosphine-oxide (TOPO) and zinc halide (ZnX₂) as surface ligands to solve this problem. TOPO, as an L-type ligand, is easily combined with Zn ions to form a TOPO-Zn complex with Lewis acid properties, which could effectively passivate the halide surface. Meanwhile, the X-ray photoelectron spectroscopy (XPS) analysis confirmed that Zn would react with oxygen (O) on the surface of the QDs, thereby effectively reducing the surface oxygen (such as the Pb–O bond in the crystal lattice) and improving the overall thermal stability (at 100 °C) of QDs by at least 10 times.¹⁵⁰ The operational lifetime of green PeLEDs was increased from 1.5 to 13.5 min at a constant current density of 1 mA/cm². Bi et al.⁹⁰ provided a different idea to deal with the surface defects of QDs. They used the strong acidity of hydrobromic acid (HBr) to protonate the original ligand OAm and desorb the OA, removing the defective [PbBr₆]^{4−} octahedrons on the surface of small sized (~4 nm) CsPbBr₃ QDs, then used the didodecylamine (DDAM) and PEA to coordinate with the uncoordinated sites of QDs. PEA was used because it is with a benzene ring structure which helped to improve the carrier transport between adjacent QDs.⁹⁰ The treated QDs showed ultra-low defect density and 97% of PLQY. The prepared blue PeLED exhibited a brightness of 3850 cd/m² at 470 nm and an EQE of 4.7%, and the stability T50 reached 12 h at a constant current density of 12.5 mA/cm². This is the current record of the operational lifetime of QDs-based blue PeLEDs. Moreover, Choy et al.⁴⁷ introduced poly(maleic anhydride-*alt*-1-octadecene) (PMA) into the precursor for the synthesis of CsPbI₃ QDs (Figure 2c), and the synthesized CsPbI₃ showed excellent stability. It was explained that the PMA reacted with PbI₂ to regulate the crystallization kinetics of the MHP and was adsorbed on the surface of the QDs through the Pb–O bond. The chemically cross-linked PMA could increase the formation energy of defects and reduce the occurrence of deep-level defects. The red light device showed an efficiency of 17.8%. More importantly, the operational stability has reached 317 h at a constant current density of 30 mA/cm².

The existence of defects at the surface of MHP grains and the interface between MHP and the charge transport layer seriously affects the efficiency and stability of the device. For eliminating the surface defects of MHP, various methods including changing the type and composition of ligands on the crystal surface,^{82,86}

introducing inorganic salts,¹⁵¹ and embedding crystals in polymers³¹ have proven to be effective. For example, Wang et al.⁶¹ used cesium trifluoroacetate (CsTFA) instead of cesium bromide (CsBr) as the cesium source to prepare the 3D MHP film by a one-step solution method without anti-solvent treatment. Experimental results showed that the prepared film was typically flat and dense, with a grain size of 60 nm, while the control sample has more voids and a grain size of 300 nm.⁶¹ They reported that the interaction between TFA[−] anion and Pb²⁺ cation could greatly increase the crystallization rate of MHP. Meanwhile, TFA[−] anions passivated the grain boundaries to form a larger band gap, which made electrons and holes be confined inside the crystal without being trapped by defects at the grain boundaries. The final device exhibited an EQE of 10.5% and a lifetime T50 of 250 h at an initial luminance of 100 cd/m². Han et al.⁶⁶ introduced a new Lewis base cyclam to form a strong interaction with uncoordinated Pb. Meanwhile, they stated that the cyclam could also promote charge transport in a delocalized system. Therefore, the efficiency of the prepared quasi-2D MHP-based device was increased from 1.2% to 16.24%, and the stability T50 reached 127 h at an initial brightness of 100 cd/m². In addition, they proposed a methylene-bis-acrylamide (MBA) cross-linking strategy to reduce the surface defects and suppress ion migration in the quasi-2D MHP film (Figure 2d).⁷¹ The passivation of the monomer is metastable. Therefore, the authors first introduced MBA to fill the vacancies at the grain boundary and then added potassium persulfate (K₂S₂O₈) to initiate the cross-linking reaction of MBA monomers; the cross-linked network formation can immobilize the monomer. As a result, the binding energy acylamide on MHP crystals increased from 0.54 to 0.92 eV, and the E_a of ion migration increased from 0.21 to 0.5 eV. The T50 of PeLEDs therefore increased significantly from 2.1 to 208 h at a constant current density of 0.2 mA/cm².

The rational selection of the interface layer material can reduce interface defects, regulate the crystal growth of the MHP, and prevent the migration of ions in the MHP film to the transport layer or to the electrode, thereby avoiding unnecessary chemical reactions and affecting the operational stability of the device.¹⁵² For example, Xu et al.⁶⁵ introduced the strategy of bilateral passivation, evaporating phosphine oxide molecule–diphenylphosphine oxide-4-(triphenylsilyl)phenyl (TSPO1) layers on the upper and lower surfaces of the MHP. The P=O from TSPO1 can interacted with uncoordinated Pb, the density of states (DOS) results showed that the trap states in QD films after passivating by TSPO1 was greatly weakened. They reported that the PLQY of the film increased from 43% to 79% after introducing the double-sided organic layer. The stability of the device was greatly improved from 0.8 to 15.8 h at an initial brightness of 100 cd/m². Shen et al.⁸⁵ dissolved K₂SO₄ powder in a PEDOT:PSS solution to prepare a hole transport layer as the substrate, and K⁺ ions were distributed on the surface of PEDOT:PSS. In the process of MHP nucleation and crystallization, K⁺ ions acted as a provider of nucleation seeds and induced the directional growth of MHP to form well-packed crystal, which facilitated unhindered injection of holes (Figure 2e).⁸⁵ The T50 of the K⁺-modified device was 14 min at a constant current density of 1.0 mA/cm², while the reference device was only 4 min.

Interface engineering is critical for the balance of carrier injection to avoid carrier quenching due to the accumulation of charges at the interface (such as Auger recombination).¹⁵³ Fang et al.⁶⁴ first employed the fluorescent dye DCJTb into the

Balancing Charge Injection

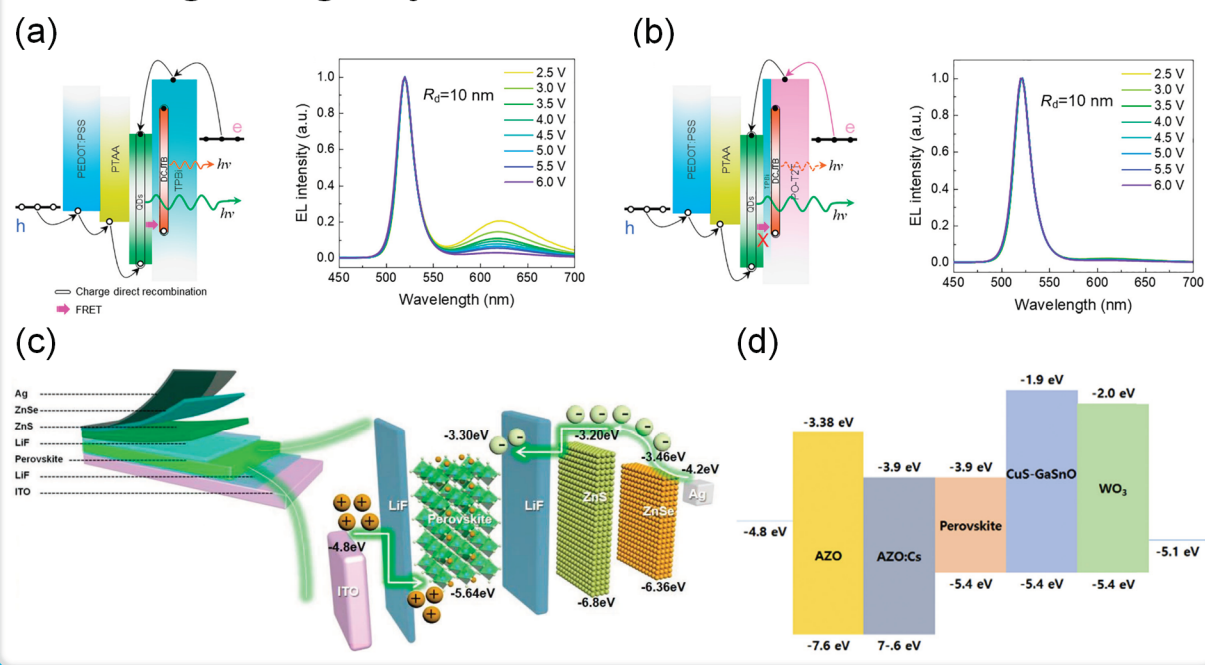


Figure 3. (a) Illustration of the control PeLED device structure with TPBi as ETL and the electroluminescence spectra. (b) Illustration of PO-T2T-modified PeLED device structure and the electroluminescence spectra. Panels a and b reproduced with permission from ref 64. Copyright 2021 Elsevier. (c) Device structure and cross-sectional SEM image of all-inorganic PeLED. Reproduced with permission from ref 62. Copyright 2020 Wiley-VCH. (d) Energy band diagram of the PLEDs. Reproduced with permission from ref 29. Copyright 2018 Elsevier.

electron transport layer TPBi to determine whether more holes or electrons were injected. The control device exhibited red emission at 620 nm originating from direct exciton recombination of DCJTb (Figure 3a), indicated that more holes were injected into the QDs and TPBi. By introducing POT2T, which had higher electron mobility than TPBi, to replace part of TPBi, the result showed that there was no emission peak of DCJTb, which referred that the balance of carrier injection was realized (Figure 3b). The T50 of the PeLEDs increased from 9.5 to 180.1 h at an initial brightness of 100 cd/m². Such a large improvement also verifies the importance of charge injection balance for improving the operational stability of the device. In addition to adjusting the charge transport layer material, changing the device structure is also an effective method. Wu et al. proposed the structure of “insulator–perovskite–insulator” (IPI) early on.¹⁵⁴ A thin LiF layer was evaporated on the lower and upper surfaces of the MHP film. This structure was proven to significantly prevent ion migration between the MHP and the transport layer. Meanwhile, it effectively suppressed the quenching of excitons at the interface. In the latest work, they introduced inorganic electron transport layers ZnS and ZnSe to replace the previously used 4,7-diphenyl-1,10-phenanthroline (Bphen) (Figure 3c),⁶² which further reduced the damage of Joule heat to the transport layer. The prepared device showed an efficiency of 11.05% and a stability of 255 h at an initial luminance of 120 cd/m². This is the current stability record of green light PeLEDs. In fact, the strategy of adopting an inorganic transport layer and a double-layer achieved good results in 2018. Yusoff et al.²⁹ spin-coated double-layer inorganic electron transport layers (cesium-doped aluminum–zinc oxide nanoparticles) and hole transport layers (copper sulfide–gallium–tin oxide and tungsten trioxide nanoparticles) on both sides of the

MHP layer (Figure 3d). The efficiency roll-off was small for the prepared multi-color device, and the brightness of the red light PeLEDs was reduced from 1000 to 100 cd/m² in 491 h.

During PeLED operation, because of the various defects of the entire device and the limitation of the optical structure, it is challenging to completely convert all injected electrons into photons for light emission. Part of the electric energy is converted into heat energy. The Joule heat has a profound effect on the long-term stability of the device. Moreover, MHP materials are currently regarded as promising thermoelectric materials because of their phonon glass properties and high carrier mobility.^{155,156} Experimental and simulation results show that the thermal conductivity of MHP is between 10^{−1} and 10⁰ W m^{−1} K^{−1}, which is 1–2 orders of magnitude lower than that of the traditional inorganic semiconductors.^{157,158} This means that heat is difficult to diffuse from the inside of the device during operation. Therefore, the effective thermal management is necessary for PeLEDs. Zou et al.¹⁴ found that significant Joule heat was generated during PeLED operation. The surface temperature of the device reached 70 °C under high voltage. They observed the degradation process of the electron transport layer 1,3,5-tris(*N*-phenylbenzimidazol-2-yl) (TPBi). The degradation of the interface caused a chemical reaction between the MHP layer and the metal Al electrode, which made the MHP layer show obvious PL quenching. Zhao et al.¹⁰¹ reduced the thickness of the emitting layer to suppress the optical waveguide and improve the optical outcoupling efficiency as well as reduce the Joule heat generated in the device. As shown in Figure 4a,b, when the active layer film thickness was 80 nm, the surface temperature of the device rose from 30 °C after 60 s operation to 32.5 °C after 120 s. For the case that the film thickness was 40 nm, the temperature was only 28 °C without any clear change

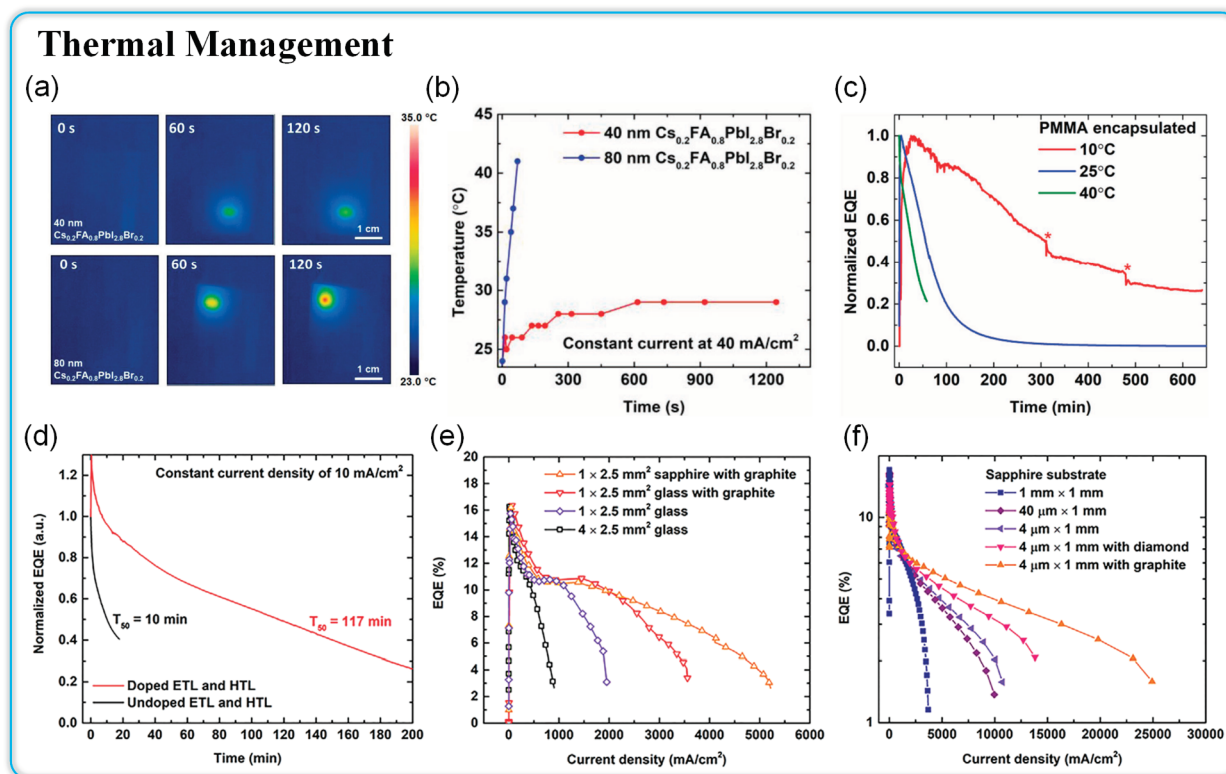


Figure 4. (a) Temperature analysis of perovskite LEDs during operation. (b) Junction temperature extracted from a $\text{Cs}_{0.2}\text{FA}_{0.8}\text{PbI}_{2.8}\text{Br}_{0.2}$ -based PeLED. (c) Operational stability of perovskite LEDs based on 35 nm MAPbI_3 thin films working at various temperatures. Panels a–c reproduced with permission from ref 101. Copyright 2019 Wiley-VCH. (d) Lifetime test of perovskite LEDs at a constant current density of 10 mA/cm^2 . (e) EQE–current density curves of perovskite LEDs on glass or sapphire substrates, with or without a graphite heat spreader and copper heat sink. (f) EQE–current density curves of perovskite LEDs on sapphire substrates, with or without a graphite or diamond heat spreader and copper heat sink. Panels d–f reproduced with permission from ref 15. Copyright 2020 Wiley-VCH.

during operation. Furthermore, they found that the ambient temperature had a considerable influence on the stability of the device as shown in Figure 4c. When the ambient temperature was 10 °C, it took 150 min for the device EQE to decay to 80% of the initial value at a constant current density of 10 mA/cm^2 , which was 40 times the lifetime at 40 °C. After finding these phenomena, they used a series of thermal management methods to improve the operational lifetime of the device.¹⁵ First, they doped the transport layer for improving the charge transport performance and the stability of the device increased from 10 to 117 min at a constant current density of 10 mA/cm^2 (Figure 4d). They believed the doping would reduce the generation of Joule heat. In order to enhance the heat dissipation, they replaced the glass with sapphire and placed a graphite heat spreader and a copper heat sink on the Ag electrode. Their results showed that the thermally managed device could operate at a current density of more than 5000 mA/cm^2 . Besides, the efficiency roll-off significantly reduced at high current density with an EQE of 10% being maintained at 2000 mA/cm^2 (Figure 4e). The device achieved a maximum radiance of 1323 $\text{W sr}^{-1} \text{m}^{-2}$ at the current density of 3.7 A/cm^2 . In addition, the thermal management strategy showed good heat dissipation on small-area device that a maximum radiance of 2555 $\text{W sr}^{-1} \text{m}^{-2}$ at the current density of 25 A/cm^2 was reported in 4 $\mu\text{m} \times 1 \text{ mm}$ small area device as shown in Figure 4f.

STRATEGIES TO IMPROVE SPECTRAL STABILITY OF PeLEDs

In order to achieve ultra-high-definition displays, the three primary colors of red, green, and blue need to meet the requirements of the newly defined International Telecommunication Union (ITU) Recommendation BT 2020 (Rec. 2020) standard. For PeLEDs, green light shall be able to meet this requirement. Currently, the pure Br-based MHP (i.e., 3D, 2D, and 0D MHPs) emission peak demonstrates the Commission Internationale de l'Eclairage (CIE) coordinates of (0.170, 0.797) (corresponding to 532 nm).^{5,159} While the CIE coordinates of pure blue and pure red are (0.131, 0.046) (corresponding to 467 nm) and (0.708, 0.292) (corresponding to 630 nm), respectively, there is room for blue and red light to meet this standard through pure Cl- and I-based low-dimensional MHPs. As mentioned earlier, MHPs can easily adjust the emission spectrum by mixing halogens, but due to the phenomenon of halide phase separation, this method faces the issue of spectral stability. This issue has been reported in the early stage in the field of PSCs.¹⁶⁰ Recent studies have shown that the diffusion of interstitial defects and vacancies played a vital role in the process of halogen separation.^{128,161} The movement of halide ions in MHP materials was considered to occur mainly through the nearest-neighbor hopping mechanism,¹⁶¹ that is, halide ions move from one vacancy site in the lattice to another nearby ones. Besides the method of mixing halogens in MHP, pure blue and pure red light can also be achieved through the quantum confinement effect of low-

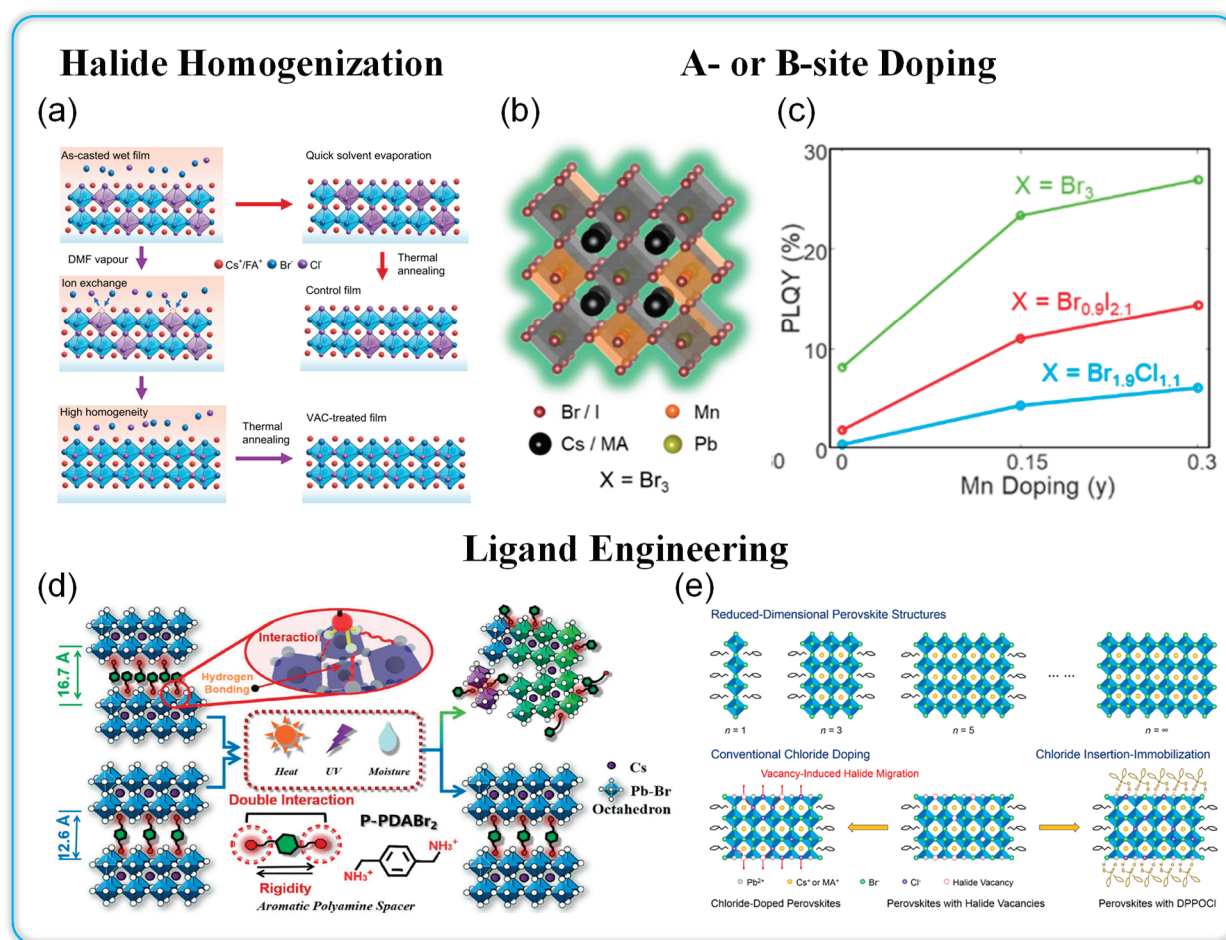


Figure 5. (a) Schematic illustration of the proposed mechanism for halide redistribution. Reproduced with permission from ref 89. Copyright 2021 Springer Nature. (b) Mn-doped perovskite crystal structure. (c) PLQY measurements with Mn doping. Panels b and c reproduced with permission from ref 76. Copyright 2019 American Chemical Society. (d) Schematic diagram of structure and alteration of 2D perovskite with different spacer. Reproduced with permission from ref 74. Copyright 2019 Wiley-VCH. (e) Perovskite structures and mechanism of the chloride insertion-immobilization process. Reproduced with permission from ref 22. Copyright 2020 American Chemical Society.

dimensional (2D or quasi-2D MHPs and QDs) pure halogen MHP.^{7,162} However, it is still difficult to accurately control the emission wavelength and prepare high-efficiency devices. Here, we mainly discuss the case of mixed halide MHP.

For 3D MHP, Cheng et al.²³ found that the phase separation after film formation could be suppressed when the halogen in the precursor solution was uniformly mixed before the film is prepared. When the precursor was not treated, the emission spectrum of the device prepared by this film was very unstable under electrical stress. When polyoxyethylene sorbitan monolaurate (Tween) and tetraphenylphosphonium bromide (TPPB) were separately introduced into the precursor solution, the phase separation in the film was significantly improved. As a cationic surfactant, TPPB was considered to effectively promote the exchange of halide ions to establish a chemical balance. That is, to exchange part of the Cl (or Br) in the Cl-rich (or Br-rich) complex. Tween could form a weak interaction with Cs ions, allowing Br ions in the precursor solution to be more mobile, thereby promoting more uniform mixing of Cl and Br ions.²³ The degree of phase separation was the lowest when both additives were introduced. There was no change in the emission peak of the prepared device spectrum. First-principle thermodynamic calculations were used to analyze how the uniform mixing of halogens affected the stability of the material.

The results showed that a uniform phase distribution corresponded to a uniform strain field distribution, which could reduce the strain energy. Besides, a more balanced charge distribution could reduce the Coulomb interaction within the MHP, resulting in a smaller energy gain.²³ Similarly, Karsson et al.⁸⁹ used vapor-assisted crystallization to mitigate the heterogeneity of the components in the mixed Br/Cl 3D MHP film. The proposed mechanism for halide redistribution is shown in Figure 5a. Due to the nonequilibrium grain growth, there are Cl-rich solid phases and Br-rich liquid phases in the spin-coated film. If the sample is treated with thermal annealing, the components of the MHP would be quickly fixed due to the volatilization of the solvent, resulting in uneven halide distribution. In contrast, in an environment with DMF vapor, the liquid Br-rich phase can present for a long time. Due to the chemical potential difference between the Cl-rich phase and the Br-rich phase, the halide exchange process will occur spontaneously and eventually reach a chemical equilibrium, forming a uniform halide distribution. This strategy has also proven to be as effective as quasi-2D MHP.⁸⁹

While inhibiting the formation of defects could effectively hinder phase separation and thus improve the spectral stability,^{163,164} the A- or B-site doping is an efficient way to improve the structural stability and reduce defects of MHP. For

quasi-2D MHP, Gangishetty et al.⁷⁶ doped Mn cation into the $\text{PEA}_{0.2}\text{Cs}_{0.4}\text{MA}_{0.6}\text{PbBr}_{0.9}\text{I}_{2.1}$, $\text{PEA}_{0.2}\text{Cs}_{0.4}\text{MA}_{0.6}\text{PbBr}_3$, and $\text{PEA}_{0.2}\text{Cs}_{0.4}\text{MA}_{0.6}\text{Pb}(\text{Br}/\text{Cl})_3$ quasi-2D MHP (Figure 5b), they found that the morphology of the films has changed significantly. Without Mn doping, the crystal grain size in the film was large and there were large gaps between the grains, while the crystal grain size of the film became smaller and the film became denser after Mn doping. Their results showed that with Mn doping, the PLQY of the films increased accordingly (Figure 5c), the devices showed excellent operational stability, with the sky-blue devices lasting for 20 min and red devices over 5 h with strong spectral stability at a constant current density of 3 mA/cm². In addition to quasi-2D MHP, this A- or B-site doping strategy is equally effective for MHP QDs.^{165,166} Zhang et al.¹⁶⁵ proposed a N–H...X strategy to achieve spectral stable blue PeLEDs. They doped FA cations or guanidine (GA) cations with amino groups into the $\text{CsPb}(\text{Br}/\text{Cl})_3$ QDs, through the formation of N–H...X hydrogen bonds between the amino groups and X-sites to increase the energy barrier of halide migration, thereby preventing the shift of the spectrum. The results of temperature-dependent conductivity measurements showed that the E_a of ion migration increased from 0.42 eV (without doping) to 0.49 (GA) and 0.52 eV (FA).

For 2D/quasi-2D MHP and QDs, ligand engineering is very important for phase stability and defects passivation. For quasi-2D MHP, organic ammonium cations play a vital role in phase stability.^{80,84,147} Yuan et al.⁷⁴ introduced a new aromatic polyammonium bromide [1,4-bis(aminomethyl)benzene bromide (P-PDABr₂)] with double functional amino terminals to prepare quasi-2D MHP (Figure 5d). Density functional theory (DFT) simulation results show that, compared with PEA with a single amino group, the absolute value of the formation energy of MHP with the pure $n = 3$ phase has increased from 5.01 to 8.32 eV. P-PDABr₂ exhibited a stronger binding force with PbBr_6^- and pinned the domain phase distribution, thereby improving spectral stability. The treatment of halogen vacancies on the surface is also critical for quasi-2D MHP. Ma et al.²² proposed a chloride insertion–immobilization strategy to mitigate the halide segregation (Figure 5e). They first prepared a pure bromine MHP precursor solution. During the spin-coating process, chloroform solvent dissolved with organic chlorides was introduced for anti-solvent treatment. After the organic chloride reacted with a small amount of water, it inserted and *in situ* immobilized the chloride ions on the MHP crystal surface; thus the vacancies on the crystal surface were saturated and hydrogen bonds were formed.²² The prepared PeLEDs based on this strategy exhibited an EQE of 5.2% at 479 nm, an operational lifetime of 1.5 h at an initial luminance of 100 cd/m². Notably, the EL spectrum did not shift during the entire operational lifetime test.

For perovskite QDs, the selection and regulation of surface ligands will affect the spectral stability of the device.^{167–169} Halide vacancies are the predominant defect species of CsPbX_3 QDs.¹⁷⁰ Unlike Br and I, the absence of Cl ions can cause deep-level defects.¹⁷¹ Therefore, the passivation of Cl vacancies is particularly important. Zeng et al.¹⁶⁷ used organic pseudohalide (*n*-dodecylammonium thiocyanate (DAT)) to passivate the Cl vacancies in the $\text{CsPb}(\text{Br}/\text{Cl})_3$ QDs. The treated QDs dispersion exhibited near-unity PLQY and the EL spectrum of the assembled PeLEDs only slightly shifted under different bias voltages. In addition to the Br/Cl mixed case, the Br/I mixed perovskite also suffered from the issue of phase separation. Very recently, Yasser et al.²⁴ used multidentate ligands ethylene-

diaminetetraacetic acid (EDTA) and reduced L-glutathione to post-processed the synthesized $\text{MAPb}(\text{I}_x\text{Br}_{1-x})_3$ QDs, which suppressed the phase separation under an electric field and improved the stability of the EL spectrum. The EDTA and glutamine ligands bind strongly to Pb. In the process of introducing these two ligands for ligand exchange, the uncoordinated Pb on the surface of the nanocrystal would be removed. Also, the ligand had a strong affinity with the nanocrystal surface, which improved the formation energy of the iodine Frankel defect, thereby inhibited the formation of this defect. Moreover, the intermolecular hydrogen bonding of the carboxylic groups of the two ligands made the molecular framework adsorbed on the surface become more stable.²⁴

CONCLUSIONS AND OUTLOOK

In summary, we reviewed the latest research progress on stability of PeLEDs. It can be seen that the spectral stability and operational stability of multi-color PeLEDs were greatly improved in the past several years. The key strategies for operational stability can be summarized into three aspects:

- (1) Reduce defects. A smooth, and dense film can effectively avoid the electric leakage and defects, and the morphology of the film can be greatly improved by adjusting the composition of the precursor (i.e., CsTFA,⁶¹ PEG,¹⁷² 18-crown-6¹⁷³), anti-solvent treatment (i.e., chloroform,⁵² ethyl acetate⁷³), and preparation methods (i.e., electric-field-deposition (EFD) method⁷⁵). The defects in perovskite films can be minimized by adjusting the composition of ligands on the crystal surface (i.e., cyclam,⁶⁶), introducing inorganic salts (i.e., ZnBr_2 ,¹⁵¹ lithium halide⁶⁷), and embedding crystals in polymers (i.e., poly-HEMA,³¹ PMA⁴⁷). The stability of perovskite materials can be significantly improved by metal doping (i.e., Rb,⁸³ Mn⁷⁶).
- (2) Interface engineering. The selection and structure optimization of the interface layer material (i.e., TSPO1,⁶⁵ LiF,¹⁵⁴ ZnS,⁶² ETPTA,¹⁷⁴ TBTB,⁸ PAC⁴⁸) is crucial to the balance of carrier injection, interface defects, and the crystallization of perovskite films.
- (3) Thermal management. Joule heat has a profound effect on the long-term stability of the device, and it can be reduced by regulating the thickness of the emitting layer,¹⁰¹ doping the transport layer, and replacing the glass with sapphire, a graphite heat spreader, and a copper heat sink.¹⁵

The spectral stability can be significantly improved by three strategies:

- (1) Halide homogenization. The uniform distribution of multiple halogens in the mixed-halide film seems to be able to effectively improve the spectral stability. Methods to achieve uniform distribution of halogens include the addition of organic molecules (i.e., Tween,²³ TPPB²³) and vapor-assisted crystallization (i.e., DMF vapor⁸⁹).
- (2) Metal-doping and alloying. Metal doping (i.e., Rb,⁷⁷ Ni,¹⁷⁵ Mn¹⁶⁶) can effectively improve the structure stability of the perovskite and reduce defects, which is beneficial to improve the spectral stability.
- (3) Ligand engineering. The phase stability of the quasi-2D perovskite can be improved by adjusting the C-site ligand (i.e., P-PDABr₂⁷⁴). Defects play an important role in the process of ion migration, and appropriate selection of

ligands (i.e., EDTA,²⁴ KI¹⁷⁶) can effectively reduce defects.

Although some progress has been made in operational stability of PeLEDs, compared with relatively mature OLEDs and QLEDs, there is still a gap of 2–3 orders of magnitude in operational stability. Different from organic materials and inorganic QD materials, the ionic crystal characteristics of MHP make it prone to ion migration under high bias voltage. As discussed above, although defects can be reduced through various strategies and the E_a can be improved, significant ion migration can still be measured after long-term bias voltages.⁷¹ Some current work has tried to reduce the defects of MHPs and the interface between the MHP and carrier transport layer to an extremely low level. As a result, the devices exhibit a high EQE.^{8,174,177} However, more efforts shall be put on the operational stability. From the recent cross-linking method⁷¹ and the polymer ligand incorporation⁴⁷ for improving the lifetime to dual multidentate ligands incorporation²⁴ for solving the phase separation, we would suggest that for PeLEDs, when performing defect passivation, besides considering the strong binding force between the passivating molecule and the chemical groups of the defect sites, the steric hindrance should also be considered to prevent ion migration. Furthermore, the issue of ion migration for MHP materials with extremely low defects has not yet been fully understood. To reach the level of

To reach the level of practical application, it is necessary to further understand the impact of ion migration on light-emitting devices, and more research efforts on increasing E_a and block ion migration are needed.

practical application, it is necessary to further understand the impact of ion migration on light-emitting devices, and more research efforts on increasing E_a and block ion migration are needed. In addition, it is worth noting that the test conditions of various reports are different when performing operational stability tests. This brings difficulties to make comparison of stability. Under different test conditions, there is a huge difference in the life span. As mentioned in the work of Choy et al.,⁸² when the device was operated under a constant current density of 1.5 mA/cm², the device exhibited an impressive lifetime of 3961 s, while in the case of 10 and 20 mA/cm², the lifetime were 296 and 174 s, respectively. In order to have more fair comparison on device operating stability, it is important to report performances with clear and detailed descriptions of test conditions (i.e., the value of constant voltage or current and the value of initial brightness or EQE) and develop test standards.¹⁷⁸ Some works test the spectral stability for a short time, and whether the issue of spectral stability has been solved still needs to be verified under more stringent test conditions.

There are several promising development directions to improve the operational and spectral stability of perovskite LEDs. As we know, various defects from atomic vacancies to dangling bonds exist on perovskite surface,¹²² ligand engineering plays an important role in passivating defects. Recently, ligands with strong electron withdrawing groups have been proposed to passivate and achieve defect-free perovskite, such as sulfonates,¹⁷⁹ fluorinated carboxylates, and alkylphosphonates.¹⁷¹ Multidentate ligands (i.e., cyclam,⁶⁶ EDTA,²⁴ PAC⁴⁸) also show

great potential in improving device efficiency and operational and spectral stability. Although there is still no report of a wide band gap material with a well-matched lattice constant, the perovskite core–shell structure showed prolonged the stability of the material.^{180,181} It is beneficial to suppress the Auger recombination at high current density, and to realize low efficiency roll-off LEDs by choosing the proper shell material. In addition, the electronic structure of the shell materials and the optimal lattice match between the core and shell should be carefully investigated. For example, Zhang et al. suggested CsPbI₃/Mg_xZn_{1-x}Te core/thick shell QDs, which may simultaneously offer balanced charge injection, enhanced emission, and suppressed ion migration, thus improving the operational and spectral stability.¹⁸² Overall, the core–shell structure of perovskites requires future design, synthesis, and testing the feasibility and effectiveness.

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