

REVIEW

Stability of electroluminescent perovskite quantum dots light-emitting diode

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Abstract

Colloidal quantum dots (QDs) are considered to be a promising candidate for light-emitting diodes (LEDs) applications considering their wide gamut range, simple color tuning, compatibility with large-size and flexible substrates. Benefiting from the distinguished properties such as high photoluminescence quantum efficiency (PLQY), narrow emission linewidth, and low material cost, perovskite QDs have attracted wide attention from researchers. Despite tremendous progress has been made during the past several years, perovskite QDs-based LEDs (QLEDs) are still plagued by the instability arising from the operation of the perovskite QLEDs, which have not been studied systematically. In this review, the origin of the instability of perovskite QLEDs is attributed to the vulnerability of emission materials and constituent layers. Simultaneously, the effective strategies to improve the durability of perovskite QDs and QLEDs are summarized, such as component engineering, ligand passivation, fabrication of robust and uniform constituent layers, as well as adopt appropriate device structures and driving modes. Ultimately, we put forward insights on the future research direction and prospects for perovskite QLEDs in practical applications.

KEYWORDS

electroluminescence, nanocrystal, perovskite, quantum dots

1 | INTRODUCTION

Belonging to the core technologies in the contemporary information society, display technology can improve the convenience and quality of human life.^[1–3] As one of the major components of displays, light-emitting diodes (LEDs) are targeted to offer considerable resolution, high

purity and satisfactory efficiency. Despite organic LEDs (OLEDs) have been commercialized in recent years, it is inevitable that organic fluorophores have a wide emission spectrum, narrow color gamut and low color purity.^[4] Moreover, considering that vacuum-based thermal evaporation is the main way to fabricate OLEDs, it is difficult to achieve the cost-effective and large-area processing.

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Quantum dots (QDs) with the size of only a few nanometers have drawn research's attention considering their wide gamut range, convenient color tuning and the great compatibility with substrates.^[5,6]

Among these semiconductor nanocrystals, perovskite QDs emerging in recent years offer several advantages for using as components of LEDs.^[7–9] This direct-bandgap semiconductor material possesses high photoluminescence quantum yield (PLQY) as well as narrow PL spectra, which can be adjusted all over the visible spectral through straightforward halogen modification.^[10,11] Perovskite QD emitters offer the wider color gamut ($\approx 140\%$) than the National Television System Committee (NTSC) standard on a CIE chromaticity diagram. Furthermore, perovskite QDs have been proved to be synthesized and processed at room temperature easily.^[12,13] All of these fascinating features led to a rapid development of perovskite QDs-based LEDs (QLEDs), whose external quantum efficiencies (EQEs) is nearly approaching the respective values of OLEDs.^[14] However, due to the relatively poor stability of perovskite QD materials and devices, the commercialization of perovskite QLEDs are still challenging.^[15]

Not surprisingly, there have been considerable reviews describing the instability of perovskite in past few years, but they mainly focus on the perovskite QDs materials.^[16,17] In fact, the stability of the QLEDs depends on the structure of the device, the other constituent layer materials, and various external influence factors in addition to the emitting layer, which was not pointed out in detail in the previous reports. In this review, we will expound the factors affecting the stability of perovskite QLEDs such as the intrinsic instability of perovskites, inappropriate constituent layers and interface contact inducing inefficient and unbalanced carrier injection. At the same time, strategies reported for overcoming the instability of perovskite materials and devices are summarized, such as component engineering, ligand passivation, fabrication of robust and uniform constituent layers, as well as appropriate device structures and driving modes. On this basis, future research directions and an outlook of perovskite QLEDs for practical applications are prospected.

2 | ORIGINS OF THE INSTABILITY OF PEROVSKITE QLEDs

Quantum dots LEDs is one of the important fields in the research of perovskite LEDs. Different from the film one, perovskite QLEDs need to prepare colloidal QDs in solution firstly. And abundant ligands were introduced at this stage in order to stabilize the QDs with high specific surface area. Secondly, these perovskite QDs were used as emitting material and spin coated on the substrate during

the fabrication of LED. From this perspective, the instability of perovskite QLEDs can be categorized to the two aspects primarily: (i) The intrinsic instability of perovskites will lead to the decomposition of nanocrystals, which mainly from the ionicity of the perovskite and the weak interaction between the crystal and surface ligands. At the same time, external environment such as humidity, voltage, temperature and illumination will trigger the degradation of perovskite QDs and destroy the performance eventually. (ii) Fragile constituent layers and inappropriate interface contact can induce inefficient and unbalanced carrier injection, leading to considerable Joule heating and affecting long-term stability.^[18,19] In this section, we will expound the factors affecting the stability of perovskite QLEDs from the above aspects.

2.1 | Intrinsic instability of perovskite QDs

As the emitting material, perovskite QDs should be chemically stable. However, as shown in the previous reports, perovskite materials have serious problems about poor stability because of the weak ionic bond structure and dynamic ligand coordination.^[20–22] This will not only affect the luminescent properties of perovskite, but also greatly damage the stability, which will be discussed in detail as follows.

2.1.1 | Phase instability caused by mismatched ion radius

From the perspective of the emitting layer, the source of the instability of the perovskite QLED lies in the intrinsic chemical instability of perovskite. Recent studies have shown that perovskite nanocrystals have cubic, tetragonal, and orthorhombic crystal structures. Among them, cubic perovskite (ABX_3) exhibits higher symmetry and thermodynamic stability than other structures.^[23] Generally, Goldschmidt's tolerance factor was used in researches to rationalize the compositionally dependent formability of perovskites semi-quantitatively.^[24] The tolerance factor (t) can be calculated by the following formula:

$$t = \frac{R_A + R_X}{\sqrt{2}(R_B + R_X)}$$

In addition, the formability (μ) of the octahedra is determined by:

$$\mu = \frac{R_B}{R_X}$$

where R represents the effective radii of A, B, and X ions, respectively. There are several reports indicating that the cubic phase is stable when $0.813 \leq t \leq 1.107$ and $0.442 \leq \mu \leq 0.895$. Unfortunately, I ions are too large to meet the value of μ and cannot coordinate with lead effectively. Therefore, it is widely observed that the I-containing perovskite suffer from phase degradation to undesirable yellow orthorhombic phase at low temperature because of the weaker Pb-I bonding^[25–27] (Figure 1A). This phenomenon can also be observed in mixed halogen perovskites. With the introduction of I[−], the lattice constant of nanocrystals increases and the surface is reconstructed due to lattice distortion or strain^[28,29] (Figure 1B). Thereby, nanocrystals containing mixed halides of I and Br can be observed to degrade to the α -phase of CsPbBr₃ with residues of PbI₂ and CsI, resulting in the shift of spectrum and a rapid drop in PLQY. In this process, a large number of surface defects will be formed at the same time, which will act as the nonradiative recombination site and further deteriorate electrical and physical properties.^[30,31]

2.1.2 | Weak interaction between insulating ligands and QDs

Considering the higher surface-to-volume ratio and more sensitive surface states compared with nanocubes, it is challenging to synthesize and stabilize these zero-dimensional perovskite QDs.^[32,35,36] The surface defects causing non-radiative recombination of perovskite QDs are mainly composed of the uncoordinated atoms and dangling bonds. Unfortunately, massive defects are prone to regenerate during the purification and film forming process inevitably in spite of being passivated in the solution.^[37,38] These high density of defects will further evolve into the channels for ion migration under operational conditions, initiating and catalyzing device degradation and make the perovskite vulnerable to external stimuli.^[39–41] Furthermore, these defects on the interface between emission layer and carrier transport layer will reduce the carrier injection efficiency and radiation recombination, which will seriously affect the device performance.^[42,43]

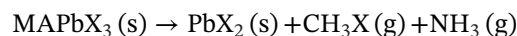
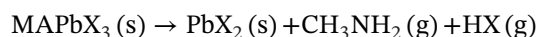
Actually, the generation of defects are related to the inevitable utilization of ligands during the synthesis of QDs.^[44] On the one hand, these ligands can saturate the dangling bonds, control the nucleation and growth kinetics of QDs, as well as isolate the QDs to avoid aggregation. On the other hand, the commonly used organic ligands (oleylamine and oleic acid) are proved to be dynamically bound to the nanocrystals, which can be detached from the surface and generate the high density of defects due to the proton exchange, leading to an increase in surface

trap states and the fatal effects in device application eventually^[32,45–47] (Figure 1C). Moreover, these induced defects in perovskite QDs will also act as triggers to accelerate the phase transition.^[48,49] It is worth mentioning that some reports indicate that the instability of perovskite QDs actually arises from oleylamine ligands because of oleylamine itself can accelerate the degradation of the nanocrystals by coordinating and dissolving the Pb-oleate from the surface.^[44,47,50] Another problem is the organic ligands with long chains can hinder sufficient carrier transport due to their insulating property. However, in order to ensure stability of nanocrystals, long-chain ligand molecules are usually required.^[51] Therefore, these surface ligands with poor electric conductivity will inevitably build an insulating barrier when perovskite QDs are assembled to form thin films, blocking the charge injection and resulting in inferior device performance.^[20,30,52]

2.1.3 | Accelerated degradation of the external environment

Considering the perovskite has ion crystal structure and weak binding energy with ligands, external stimuli such as thermal, voltage, atmosphere and illumination can trigger the damage to these materials, causing degradation of the device. In this section, we will enumerate how these factors affect the stability of the perovskite LEDs.

The perovskite QDs usually show the low thermal stability because of the fragile crystal structure.^[53] Therefore, with the accumulation of heating effect attributing to continuous bias voltage and operation time during the working conditions, nonradiative recombination rate increased significantly and the poor stability was observed.^[54,55] Considering the relatively low formation energies, these QDs cannot maintain their structure and generate a lot of defects. Accordingly, radiative recombination and carrier injection efficiency will be reduced simultaneously. Representatively, MAPbX₃ will suffer from the degradation under thermal stress by the following pathways:^[56,57]



It should be pointed out that such decomposition is irreversible due to the generation of volatile substances. What's worse, the product can further cause more serious damage to the device.^[58] The decay kinetics of CsPbBr₃ QDs under different temperatures was investigated by time-resolved PL measurement in a previous report.^[59] The results show that the exciton emission plays a dominant role at the low temperature (≤ 160 K). As the

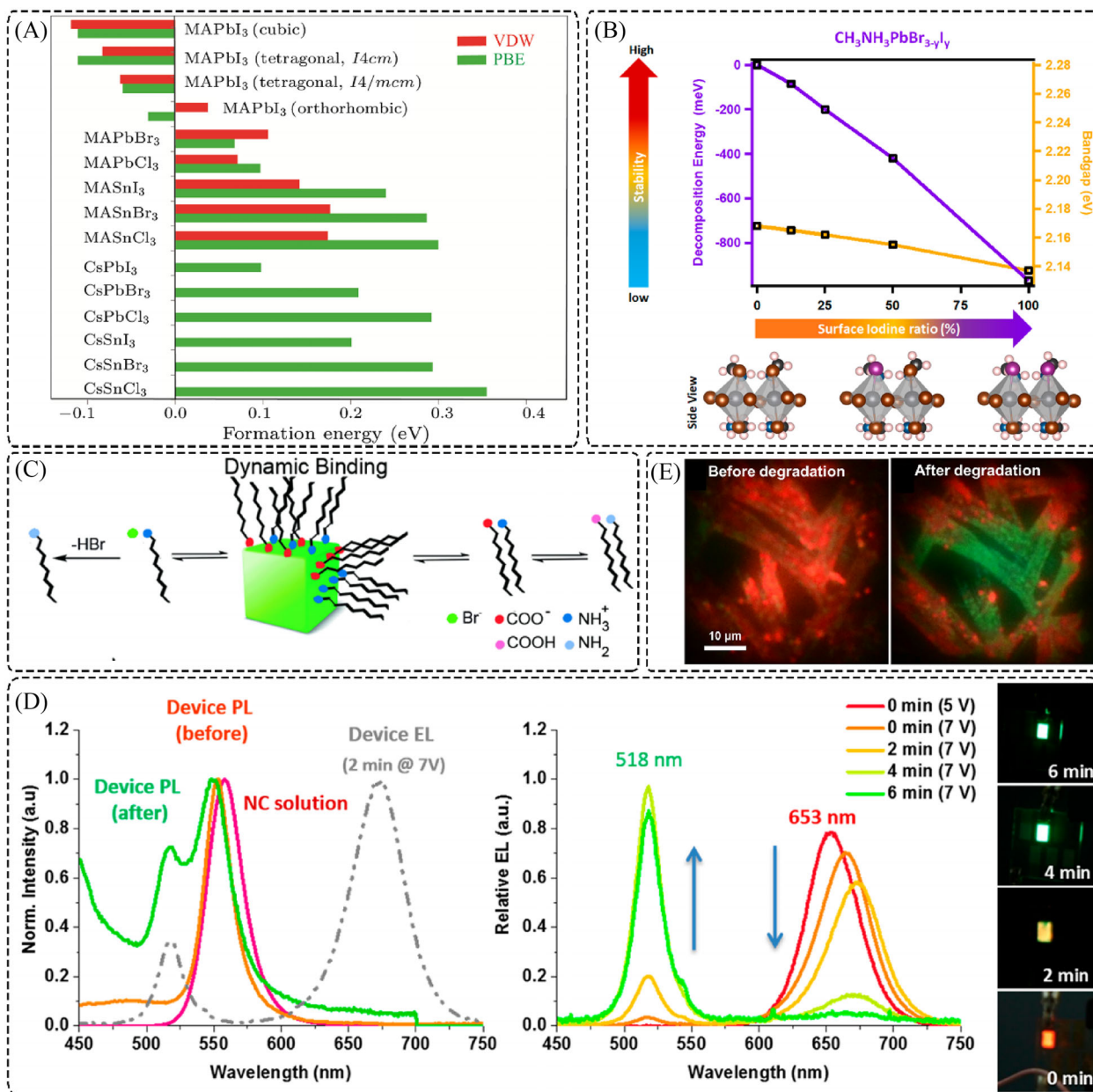


FIGURE 1 A, The calculated energy cost of the phase separation of ABX₃ (A = CH₃NH₃, Cs, B = Pb, Sn, X = I, Br, Cl) in their orthorhombic structure. For CH₃NH₃PbI₃, the results of all the three structures are plotted. For the organic-inorganic hybrid perovskites, the results from both the vdW-TS and PBE functionals are plotted. Positive number indicates that the compound is stable at T = 0 K. Reproduced with permission.^[27] Copyright 2018, IOP. B, Stability versus band gap change in mixed perovskite. Decomposition energy difference between CH₃NH₃PbBr_{3-y}I_y and pristine perovskite determined by DFT calculations. The calculations were performed on a six layer slab with only the top layer being substituted. Bottom panels show the atomic structure of the pristine, half-mixed, and completely substituted top surface of the perovskite. Color code: Br (brown), I (purple), Cl (green), C (black), N (blue), H (light pink). Reproduced with permission.^[25] Copyright 2019, American Chemical Society. C, Depiction of Conventional Ligand Capping of Perovskite Nanocrystals Using Long-Chain Molecules with Single Head Groups, In the Ionized Form (OA⁻ or Br⁻, OLAH⁺). Reproduced with permission.^[32] Copyright 2018, American Chemical Society. D, Photoluminescence spectrum (left chart) of a solution prior to thin film formation and of the finished ITO/PEDOT:PSS/pTPD/perovskite nanocrystals/TPBi/Al device before (orange line) and after (green line) device operation. The electroluminescence from the device after operation at 7 V for 2 minutes is included for reference (dashed gray line). After the initial PL measurements, the voltage for the LED was raised to the turn-on voltage of ~5 V until EL was observed and then switched to 7 V to ensure observation of ionic separation. Electroluminescence spectra (top center chart) and photos (top right) of the same LED were taken at the turn-on voltage and at 7 V and then at 2 minutes intervals until the spectrum had shifted completely. Reproduced with permission.^[33] Copyright 2017, American Chemical Society. E, PL image prior to the degradation of a MAPbI₃ bulk sample excited at 458 nm captured by a commercial color camera where the pure MAPbI₃ structures are seen as red. Another image taken of the same region after the degradation where the green emission, corresponding to PbI₂ emission, is observed. Reproduced with permission.^[34] Copyright 2016, American Chemical Society

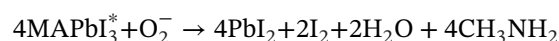
temperature rises above 180 K, the emission decay is recombination of initially generated excitons with light absorption and bimolecular radiative recombination or exciton recombination involving the surface states.^[60] However, τ_{ave} decreased significantly with the further increase of temperature (>343 K), which corresponds to the loss of ligands from the surface of CsPbBr₃ QDs at high temperature.^[61,62] It is worth mentioning that thermal is not only harmful to the device. There are multiple studies have shown a certain increase in fluorescence at the beginning of test. It could be attributed to annealing effect induced by Joule heating.^[54,63]

Mixed halide can achieve effective adjustment of perovskite emission wavelength.^[3,7,9] However, the EL spectrum usually shows an obvious shift as the voltage increases, accompanied by the appearance of multiple emission peaks^[33] (Figure 1D). This phenomenon is a result of the ion migration induced by an electrical field.^[64–68] After the test, these multiple emissions can still be observed, indicating the separation of halide is irreversible and the halide anions will redistribute among the nanocrystals.^[33] Furthermore, there are lots of evidences showing that the degradation of perovskite is induced by organic cation migration to some extent.^[69,70] The serious ions migration under high working voltage could destroy the lattice of perovskites, generate surface defects and form charge-accumulated interfaces, leading to the poor performance of QLEDs.^[71–73]

Atmospheric gases (e.g., oxygen and water) can adversely affect perovskite devices. Although perovskite QLEDs are usually prepared in an anhydrous and oxygen-free glove box filled with nitrogen in order to maintain their performance, they will inevitably be affected by the working environment in actual use. Due to the ionic properties of perovskite, polar solvents such as DMF, ethanol or water have been proved to destroy perovskite.^[34,74] These polar molecules can easily intrude into the emitting layer and diffuse rapidly, resulting in protonation and desorption of the capping ligands.^[75] With the presence of the external stimuli mentioned above, this process will even be accelerated.^[76] Furthermore, moisture can also damage the charge-transport materials such as PCBM and PEDOT:PSS, as will be discussed later.

Maintaining the photostability of perovskite QLEDs in their practical application is another troubled problem^[34] (Figure 1E). This could be attributed to the two following aspects: (i) Under photo illumination, the weak ligands will desorb from the surface, leading to the formation of trap sites and affecting the performance of device.^[77] (ii) The surface of perovskite will inevitably adhere to some oxygen. When perovskite is photoexcited, a fast oxygen diffusion is observed accompanied by photo-induced formation of superoxide species.^[78] According to the

following reaction:



These super-oxides will trigger the oxidation and decomposition of perovskite.^[79]

2.2 | Instability of constituent layers

Generally, LEDs consist of emission layer, charge transport layers, and electrodes. Therefore, except for the instability of the QDs, the poor performance of perovskite QLEDs originate from these constituent layers and their interfaces contact. The inherent instability of the constituent layer materials and the ineffective contact among them will generate defects, degrade charge-transport layers, induce unbalanced injection and corrode electrodes. Furthermore, diffusion of metallic species from electrodes can react with other materials and accelerate the degradation of perovskite QLEDs.

2.2.1 | Rough morphology of the emission layer

Many literatures have demonstrated that the morphology of the emission layer determines the performance of device.^[80–82] Improper processing methods will bring about the clustering of nanocrystal and lead to the rough thin film, which deteriorates not only the optical performance but also long-term stability of the device^[83] (Figure 2A). On the one hand, this may result in a turn-on voltage (V_{on}) much higher than the band gap energy of the nanocrystals, suggesting that poor contact between the functional layers and the carrier injection from charge-transporting layers (CTL) into nanocrystals was inefficient. On the other hand, the invalid and excessive carrier caused by the inhomogeneous film will increase the operating temperature of the whole device, which in turn accelerates the degradation of QLEDs.^[63,84]

2.2.2 | Vulnerable carrier transport materials

In general, the standard perovskite LEDs includes anode, cathode, electron transport layer, hole transport layer (HTL) and light emitting layer. The favorable carrier transport layers not only contribute to the well-balanced charge transport, but also beneficial to achieve high device stability.

Some conventional carrier transport materials, such as poly(3,4-ethylenedioxythiophen) polystyrenesulfonate

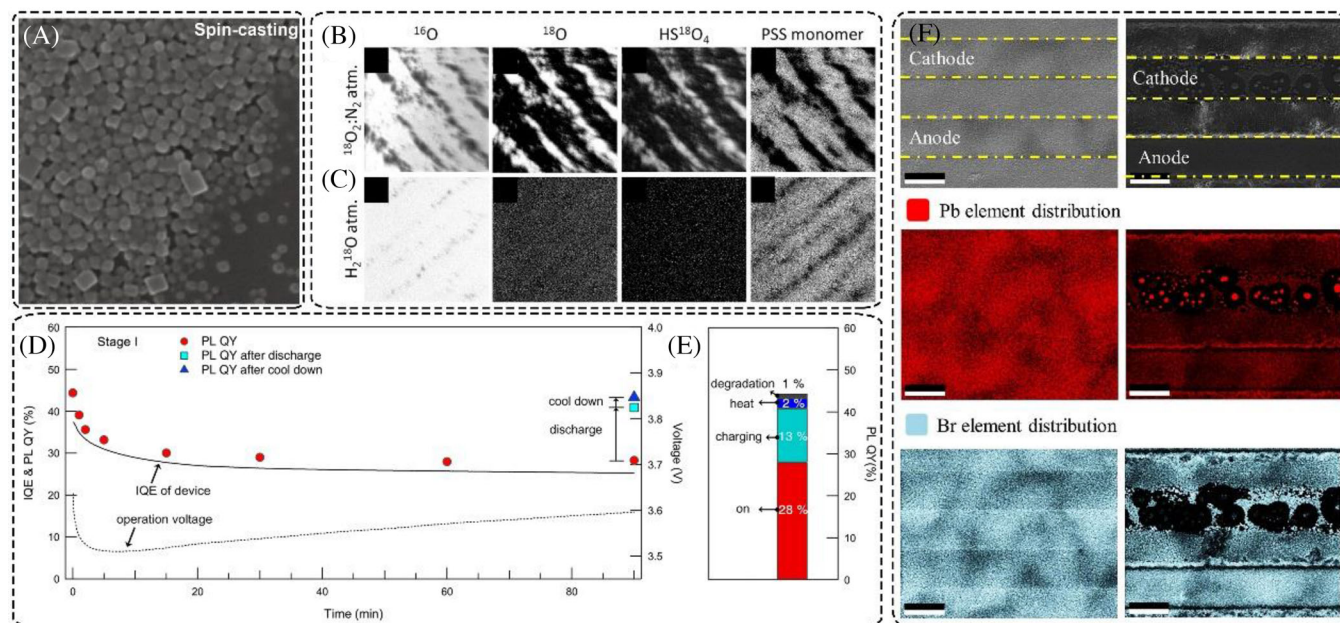


FIGURE 2 A, CsPbBr₃ nanocrystal film prepared by spin-casting methods without PMMA. Reproduced with permission.^[83] Copyright 2017, John Wiley and Sons. B, TOF-SIMS ion images (500 × 500 μm²) of PEDOT:PSS surfaces from partial devices with the composition ITO-ZnO-P3HT:PCBM-PEDOT:PSS (no Ag electrode). The devices were illuminated (AM1.5, 330 W m⁻², 65 ± 2 °C) without encapsulation for 3 weeks in a dry ¹⁸O₂:N₂ (1 atm) atmosphere or (C) in an oxygen free H₂¹⁸O:N₂ (65 ± 2% rh) atmosphere. Black corresponds to 0% signal, grey to some signal, and white to 100% signal. Reproduced with permission.^[85] Copyright 2010, American Chemical Society. D, Operation time-dependent traces of IQE (black solid line) and the operation voltage (black broken line) of the QLED employing CdSe (r = 2.0 nm)/Zn_{0.5}Cd_{0.5}S (h = 6.3 nm) QDs under continuing operation at a current density of 30 mA cm⁻² and PL QY (red circle) of the QD emissive film in the corresponding device. PL QYs of the QD emissive layer in 90 minutes-operated QLED after applying reverse voltage (-7 V, cyan square) and additional cooling for 1 hour (blue triangle) are overlaid for comparison. E, Contributions of charging, heat, and the permanent degradation of QDs to the reduction of luminescence efficiency of the QD emissive layer in a 90 minutes-operated QLED. Reproduced with permission.^[86] Copyright 2018, American Chemical Society. F, EDX elements mapping profile of Pb, Br after 0V, 5V bias. And the corresponding image of Pb and Br distribution, respectively. (The scale bar is 20 μm). Reproduced with permission.^[87] Copyright 2018, Springer Nature

(PEDOT:PSS),^[22,88,89] 2,2',7,7'-tetrakis(N,N-pdimethoxyphenylamino)-9,9'-spirofluorene,^[55] [6-6]-phenyl C61 butyric acid methyl ester (PCBM),^[90,91] and 1,3,5- tris(2-N-phenylbenzimidazolyl) benzene (TPBi),^[92] have been frequently employed in previous reports, which facilitate the injection of charge carriers to perovskite active layer. However, these organic materials are proven to interact with water and oxygen inevitably, resulting in significant device degradation^[85] (Figure 2B and 2C). For instance, the PCBM itself can be degraded through adsorbing H₂O and O₂. These will increase the resistivity and shift the HOMO-LUMO energy levels of PCBM, deteriorating device performance seriously. Besides, the PCBM layer is prone to metal diffusion, which increases the possibility of direct contact between the perovskite and electrode.^[93] Unfortunately, such process is irreversible and will induces the decomposition of perovskite LEDs eventually. Analogously, PEDOT:PSS has been widely used as the HTL of perovskite QLEDs because their solution processability and appropriate energy levels.

However, the intrinsic hygroscopic and acidic nature of PEDOT:PSS tends to corrode indium tin oxide (ITO) electrodes, reducing device stability.^[85,94–96] In addition, due to the insufficient electron blocking ability of PEDOT:PSS, carrier quenching will occur inevitably at the interface.^[97]

Another problem caused by the transport layer is the unbalanced injection of charge carriers. It is reported that efficiency of device decreased rapidly at the high current density, exhibiting the poor stability. This phenomenon, also known as efficiency roll-off, has been attributed to the thermal degradation of perovskite and unbalanced charge carrier injection at large current density.^[98–100] The possible causes include heat degradation, enhanced Auger recombination, carrier overflow at high carrier population, and carrier delocalization effect.^[10,101–104] Considering the faster electron mobility in the device than the hole, this unbalanced charge injection will bring about charge accumulation at the perovskite interface, generating considerable Joule heating and degrading the device performance^[86,105,106] (Figure 2D and 2E).

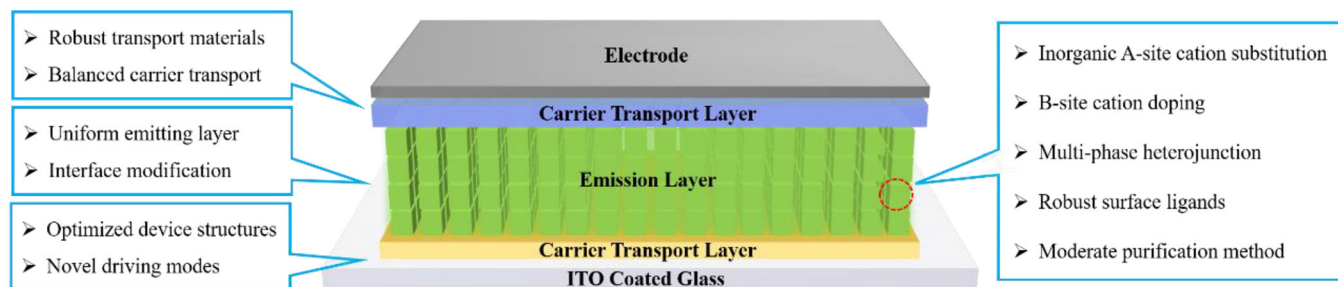
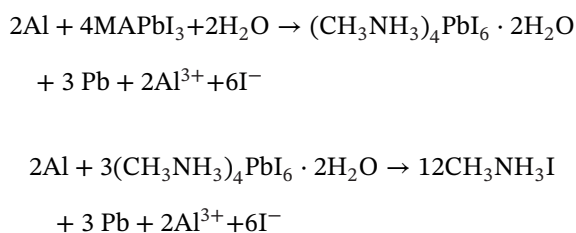


FIGURE 3 Summary of methods to improve the stability of perovskite QDs LEDs

2.2.3 | Electrode corrosion

ITO, the commonly used electrode, has been shown to affect device stability because of the releasing of In and Sn atoms under the etching of PEDOT:PSS.^[107] These metallic species can diffuse into emitting layers, and will serve as a luminescence-quenching center to accelerate the degradation of the perovskite LEDs. Furthermore, metal electrodes used in device can also be corroded by ions migrated from the perovskite^[87] (Figure 2F). For instance, I^- anions released from emission layer will corrode Ag and create AgI.^[108] This insulating components will generate deep traps or block efficient charge injection and extraction, destroying perovskite QLEDs.^[109] Furthermore, Pb^{2+} cations can react with metal electrodes under the acceleration of moisture through the following redox reactions:^[110]



3 | IMPROVING THE STABILITY OF PEROVSKITE QLEDs

In this section, we review the strategies reported recently for overcoming the instability of perovskite QDs and perovskite QLEDs. The reported operational lifetime of devices is summarized in the Table 1. As illustrated from the table, most of the devices are stable in a few minutes to hours, and only a few works improve the stability to dozens of hours. It is worth mentioning that despite the summarized studies have different measurement conditions for operational lifetime, it cannot prevent us from inferring which are effective methods to improve

stability. As depicted in Figure 3, by analyzing the working mechanism of these high performance devices, we conclude that efficient perovskite QLEDs rely on: (i) Robust perovskite crystals with low defect density, (ii) chemically stable constituent layer with efficient and balanced carrier transport capacity, (iii) appropriate device structures and driving modes.

3.1 | Improving the stability of perovskite QDs

To realize highly efficient perovskite QLEDs, the optical properties and colloidal stability of perovskite nanocrystals should be taken into consideration simultaneously. The enhancement of device performance was possible by improving crystal formation energy and choosing proper ligands. On the one hand, increasing the crystal formation energy can strengthen the interaction between crystals, which ameliorates the inherent instability of perovskite. On the other hand, ligands with robust bonding ability and optimum chain length are profitable for high optical properties and stability. Reported methods include component engineering and surface passivation, which will be discussed in detail as follows.

3.1.1 | Inorganic A-site cation substitution

Organic-inorganic perovskites containing organic A-site cations are sensitive to humidity and easily decompose due to the small formation energy.^[84] The introduction of inorganic cations like Cs into $CH_3NH_3PbX_3$ can effectively improve the stability of the devices due to the increased binding energy of perovskite.^[9,129–134] Indeed, it has been recognized that replacement of inorganic Cs^+ offers the $CH_3NH_3PbX_3$ extra thermal stability up to $\sim 500^\circ C$.^[135] Furthermore, adding Cs^+ as co-doping cation in organic-inorganic perovskite QDs also provides a feasible method to solve these problems.^[112,136] And the explanations can

TABLE 1 The recent reported operational lifetime of perovskite QLEDs

Emission layer	Device structure	PLQY	EQE	Operational lifetime ^a	Lifetime measurement conditions	Refs.
Cu:CsPbBrL ₂	ITO/ZnO/PEI/QDs/TCTA/MoO ₃ /Al	94.8%	5.1%	L ₅₀ = ~2.5 minutes	Not mentioned	[111]
CsPb ₂ Br ₅	ITO/PEDOT:PSS/QDs/TPBi/LiF/Al	~35%	1.1%	L ₅₀ = 6 hours	10 mA cm ⁻²	[84]
Cs _x (MA _{0.17} FA _{0.83}) _{1-x} PbBr ₃ (x = 0–0.15)	ITO/PEDOT:PSS/Poly-TPD/QDs/POT2T/LiF/Al	93%	7.36%	L ₅₀ = ~600 seconds	1.58 mA cm ⁻²	[112]
CsPbBr ₃ –CsPb ₂ Br ₅	ITO/PEDOT:PSS/Poly-TPD/PVK/QDs/POT2T/LiF/Al	92%	~0.62%	L ₅₀ = 2 hours	5.5 V	[113]
Q-2D/FA _{0.8} Cs _{0.2} PbBr ₃	ITO/PEDOT:PSS/QDs/TPBi/LiF/Al	/	6.8%	L ₅₀ = 440 seconds	~100 cd m ⁻² ; 298 K; 45% humidity	[114]
CsPbBr ₃ :PMMA	ITO/PEDOT:PSS/PVK/EML/TPBi/LiF/Al	/	0.25%	L ₅₀ = 50 minutes	40.7 mA cm ⁻² ; 25°C; 55% humidity	[83]
CsPbI ₃	ITO/PEDOT:PSS/EML/TPBi/LiF/Al	/	0.21%	L ₇₀ = 16 hours	5 V	[54]
CsPbBr ₃	ITO/PEDOT:PSS/Poly-TPD/QDs/TPBi/Cs ₂ CO ₃ /Al	/	1.7%	L ₅₀ = 630 seconds	10 V	[63]
CsPbI ₃	ITO/PEDOT:PSS/PVK/EML/ZnO/Ag	91%	6.2%	L ₅₀ = 13.5 minutes	150 mA cm ⁻²	[115]
CsPbBr ₃	ITO/PEDOT:PSS/PTAA/QDs/TPBi/LiF/Al	88% (solution) 79% (film)	18.7%	L ₅₀ = 30 minutes	1.5 mA cm ⁻² (1000 cd m ⁻²)	[116]
Zr-modified CsPbI ₃	Si/Ag/ZnO/PEI/QDs/TCTA/MoO ₃ /Au	91%	13.7%	Luminance decreases by 22% after lighting 100 times	8 V	[117]
CsPbBr _x Cl _{3-x}	ITO/TFB/PFI/QDs/3TPYMB/LiQ/Al	~100%	6.3%	L ₅₀ = 99 seconds	4.5 V	[118]
CsPbBr _x I _{3-x}	ITO/PEDOT:PSS/Poly-TPD/QDs/TPBi/LiF/Al	88.7%	6.36%	L ₅₀ = 540 seconds	4.1 V	[28]
CsPbBr ₃	ITO/PEDOT:PSS/Poly-TPD/QDs/TPBi/LiF/Al	69.4%	1.42%	L ₅₀ = 42 seconds	1 mA cm ⁻²	[119]
CsPbBr ₃	ITO/PEDOT:PSS/Poly-TPD/QDs/TPBi/LiF/Al	80%	6.5%	L ₅₀ = 30 minutes	2.5 mA cm ⁻²	[120]
CsPbBr ₃	ITO/ZnO/QDs/CBP/MoO ₃ /Al	23.6% (film) 85% (solution)	0.58%	20 minutes without any decay	3.5 V	[121]

(Continues)

TABLE 1 (Continued)

Emission layer	Device structure	PLQY	EQE	Operational lifetime ^a	Lifetime measurement conditions	Refs.
CsPbBr ₃	ITO/ZnO/QDs/p-Si/Al	70%	0.1%	L ₅₀ = ~5 minutes	AC driving mode; 4 V	[122]
CsPbI ₃	ITO/ZnO/QDs/Poly-TPD/p-Si/Al	/	/	L ₆₄ = 90 minutes	AC driving mode (square pulsed bias, duty cycle: 50%, frequency: 10 Hz, V _{pp} : 8 V)	[123]
CsPbBr ₃	ITO/PEDOT:PSS/poly-TPD/PFI/QDs/TPBi/LiF/Al		0.06%	L ₅₀ = 10 minutes	5 V; N ₂ filled glovebox	[92]
CsPbBr ₃	Au/p-MgNiO/CsPbBr ₃ /n-MgZnO/n ⁺ -GaN	82.8%	2.39%	L ₈₀ = 10 hours	28°C; 30–50% humidity; 10 V	[59]
CsPbBr ₃	Al/n-ZnO/CsPbBr ₃ QDs/p-NiO _x /ITO	83.6%	3.79%	L ₇₀ = 12 hours 15.1% decay	8.0 V; 75% humidity; without encapsulation 50 mA cm ⁻² ; 393 K; three heating/cooling cycles	[124]
MAPbBr ₃	ITO/CPEs/QDs/TPBi/LiF/Al		5.66%	12 hours without decay	20 mA cm ⁻²	[125]
CsPbBr ₃	Au/p-NiO _x /CsPbBr ₃ QDs/MgZnO/Au NPs/n-ZnO/n ⁺ -GaN		4.626%	L ₈₅ = 60 hours	25°C; 20–30% humidity; 8.0 V	[19]
CsPbBr ₃	ITO/NiO _x /QDs/ZnO/Al		0.11%	L ₇₀ = 1.75 hours	65% humidity; unencapsulated	[18]
CsPbBr ₃	ITO/PEDOT:PSS/poly-TPD/QDs/TPBi/LiF/Al	82%	16.8%	L ₅₀ = 24 minutes	8 mA cm ⁻² , L ₀ ^{nT} = constant (n = 1.5, L ₀ = 100 cd m ⁻²)	[126]
CsPbBr ₃	ITO/PEDOT:PSS/PVK/QDs/TPBi/LiF/Ag		0.35%	L ₅₀ = ~100 seconds	50% humidity; dark room; 7.0 V	[127]
CsPbI ₃	ITO/PEDOT:PSS/poly-TPD/QDs/TPBi/LiF/Al	89%	17.8%	L ₅₀ = 317 hours	30 mA cm ⁻²	[128]

Abbreviations: AC, alternating current; EQE, external quantum efficiency; LED, light-emitting diodes; PLQY, photoluminescence quantum efficiency; QLEDs: QDs-based LEDs.

^aL₅₀, L₆₄, L₇₀ and L₈₅ are defined as the time taken for the luminance to decay from 100% to 50%, 64%, 70% and 85% respectively.

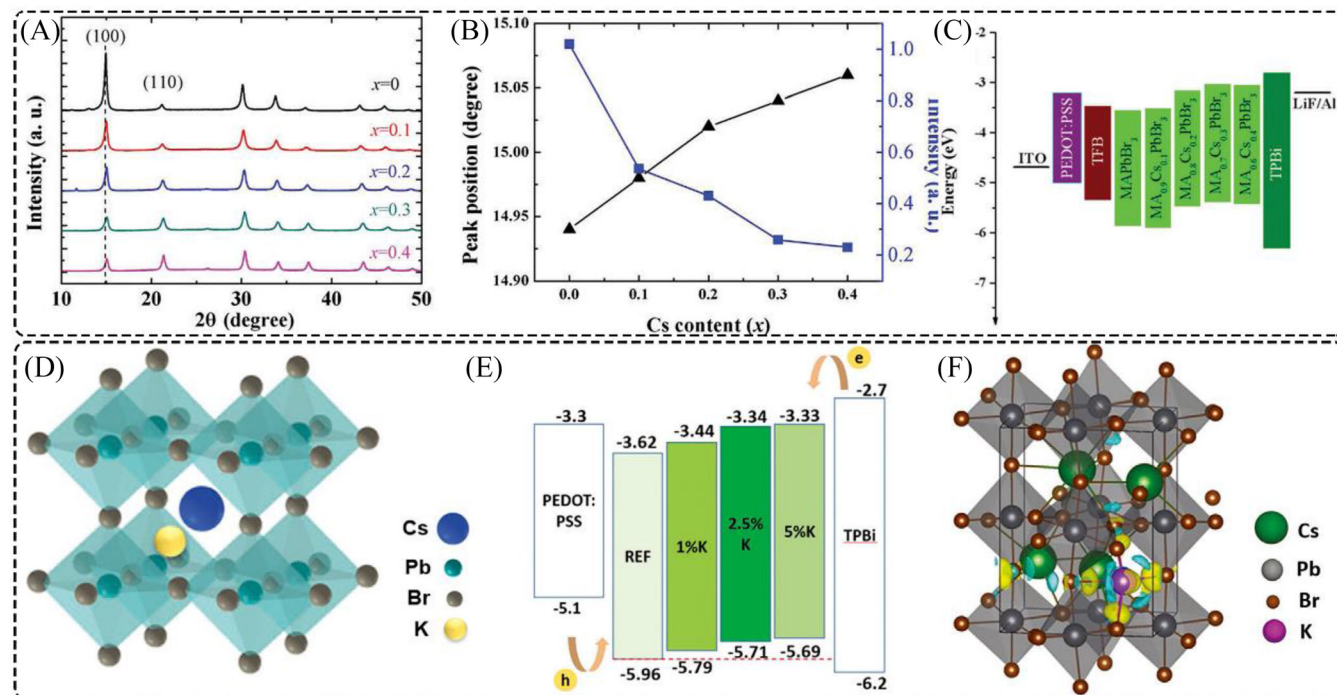


FIGURE 4 A, The XRD patterns obtained for the MACsPbBr₃ perovskites. B, The (100) peak position and (100) peak intensity upon increasing x; (C) The energy band diagram of the perovskite QLEDs structure. Reproduced with permission.^[137] Copyright 2017, Royal Society of Chemistry. D, Graphical illustration of perovskite crystal structure with potassium acting as interstitial site; (E) Band energy alignment of perovskite nanocrystal films adjacent to hole injection and electron injection layer; (F) Optimized structure of K-doped CsPbBr₃ where K⁺ stays in the quarter interstitial site. The blue and yellow areas show the charge density that represent the electron depletion and electron accumulation, respectively. Reproduced with permission.^[138] Copyright 2020, John Wiley and Sons

be ascribed as follows: (i) lattice shrinkage caused by smaller Cs atom can improve the formation energy of perovskite, (ii) the inorganic A-site cation shows better thermal and humidity stability than organic one and (iii) the energy levels match well after introducing Cs^[137] (Figure 4A-C).

Previous studies have confirmed that the incorporation of potassium ions helps to inhibit the formation of photoinactive δ -phase in mixed cationic perovskites or CsPbI₃ solar cells to enhance phase stability.^[137] A recent work shows that K can be introduced into perovskite crystal and stay as interstitial defects to enhance crystallinity, photoemission as well as stability of the nanocrystals^[138] (Figure 4D). The doping of K can modify energy levels of the nanocrystal, leading to satisfying energy alignment with TPBi and PEDOT:PSS used in the devices (Figure 4E). Furthermore, K⁺ are prone to stay at interstitial site in the perovskite compound and draw out bromide from the perovskite lattice selectively, leading to excess bromide on the surface^[139] (Figure 4F). Consequently, more ligands prefer to bond on the nanocrystal surface with bromide, which can passivate the surface trap and thus, stabilize the nanocrystals.^[47]

3.1.2 | Enhanced formation energy by B-site cation doping

Incorporating ions of different elements such as Ce³⁺,^[140] Mn²⁺,^[141] Sr²⁺,^[142] and Cu²⁺^[111] into host lattices has been extensively explored as an effective approach for nanomaterials to stabilize the structure and to adjust the performance. The formation energy of mixed halogen perovskite CsPbBrI₂ can be enhanced through substituting Pb²⁺ by Cu²⁺. This can be interpreted as the Cu²⁺ ions with a smaller radius can increase tolerance and the bond strength of metal-halides, which makes the structure of cubic perovskite more stable^[111] (Figure 5A-D). On the one hand, appropriate introduction of Cu²⁺ cations can stabilize the α -CsPbBrI₂ nanocrystals. On the other hand, the partial replacement of Pb²⁺ may determine the octahedral voids, which match well with the size of Cs⁺. However, as for CsPbI₃, Cu-substitution is invalid for the improvement the stability. This can be interpreted as follows. The [BX₆]⁴⁻ octahedra is the basic unit in the perovskites ABX₃ and if octahedral parameter (μ) is too small, the unit may become unstable.^[143] Since the ionic radius of Cu²⁺ is much smaller than that of Pb²⁺, substituting Cu²⁺

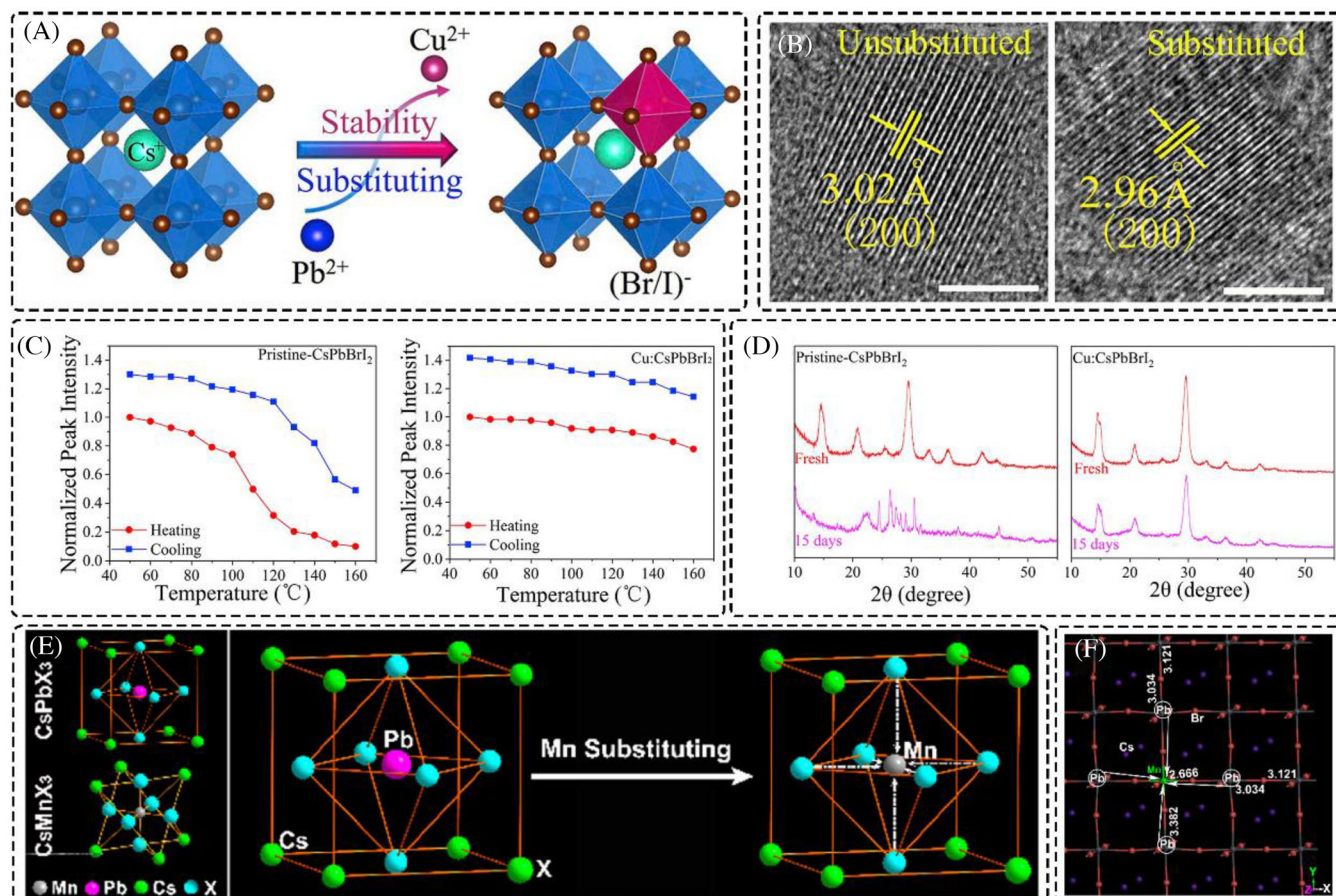


FIGURE 5 A, Schematic illustration showing the structural properties of perovskite CsPbBr_2 crystals before and after the substitution of Pb^{2+} with smaller Cu^{2+} ions; (B) HR-TEM images of unsubstituted CsPbBr_2 and Cu^{2+} -substituted CsPbBr_2 nanocrystals (scale bars: 5 nm); (C) temperature dependent PL behavior of peak emission intensity of unsubstituted and substituted CsPbBr_2 nanocrystals in hexane; (D) PXRD patterns recorded for the unsubstituted and substituted CsPbBr_2 nanocrystals thin films stored for 15 days. Reproduced with permission.^[111] Copyright 2019, Elsevier. E, Schematic illustration showing the crystal structures of perovskite CsPbX_3 and CsNiCl_3 -structured CsMnX_3 with identical octahedral coordination environment of host cation formed by six halide atoms; (F) Calculated three-dimensional stacking diagram of a $\text{CsPbBr}_3:\text{Mn}$ crystal when Pb^{2+} ion was substituted by Mn^{2+} ion with an actual concentration of 2.08 mol % by using first-principle calculations based on DFT, showing the lattice contraction in the (001) lattice plane of an orthorhombic perovskite CsPbBr_3 crystal induced by Mn^{2+} doping. Reproduced with permission.^[141] Copyright 2017, American Chemical Society

into the CsPbI_3 host may result in the undesirable lattice collapse. Compared to Cu^{2+} , the substitution of Pb^{2+} with Mn^{2+} might induce more appropriate lattice contraction and substantially stabilize perovskite lattice^[141] (Figure 5E and 5F). As a result, $\text{CsPbX}_3:\text{Mn}$ QDs with much better stability can be obtained, which is beneficial to fabricate the high-performance perovskite QLEDs.

Zn^{2+} have been proven to be another effective doping element for improving the performance of perovskites.^[144] A recent work showed Zn^{2+} can improve tolerance factors and stability of nanocrystal by progressive lattice contraction without changing the crystal form. Simultaneously, the perovskite crystal changed to nearly ambipolar with the incorporation of Zn, eliminating the hole injection barrier of QLEDs.^[145,146] Because of both CsPbBr_3 and CsSnBr_3 show the same crystal structure, it is expected

that Sn^{2+} doping in CsPbBr_3 QDs is easier.^[147,148] Unfortunately, it is widely known that Sn^{2+} are easily oxidized to Sn^{4+} in the air, endowing the perovskite QDs undesirable instability.^[149] Especially, the introduction of excessive Sn will also decrease PL strength dramatically due to the mass defects caused by oxidation.^[150,151]

3.1.3 | Multi-phase heterojunction

In addition to the two methods mentioned above, multi-phase regulate is another way to obtain the stable perovskite. An early research indicated that the structural transformation from CsPbBr_3 into CsPb_2Br_5 could be triggered by excess PbBr_2 at low temperature.^[134] On one side, CsPb_2Br_5 prefer to attach on the surface of the CsPbBr_3

nanocrystals to lower the overall energy of compound. On the other side, the coulomb attraction between them also plays a crucial role to form the composite nanocrystals. The generation of CsPb_2Br_5 nanoparticles not only facilitates radiative recombination but also suppresses the nonradiative decay, increasing the performance of QLEDs. Similarly, another work also pointed out the outstanding thermal stability should be attributed to the interfacial stabilization effect between CsPbBr_3 and CsPb_2Br_5 .^[113] The CsPbBr_3 nanocubes with small size can distribute and anchor on the surface of CsPb_2Br_5 (Figure 6A and 6B). In this case, the strong interfacial effect would make CsPbBr_3 nanocubes isolated from each other during the annealing process. Therefore, the composite film showed great thermal stability (Figure 6C-E).

3.1.4 | Robust ligands with stronger binding capacity

Dangling bonds and uncoordinated atoms produced by ligands shedding are the predominant defect species in perovskite QDs.^[152,153] Due to the dynamic combination ability, these ligands are easily detached from perovskite surface upon purification and spin coating, which induce the numerous non-radiation recombination and poor colloidal stability. Therefore, a large number of methods have been devoted to optimize the colloidal QDs such as surface passivation, ligand exchange and so on, which are proven to be an effective way to enhance the radiative recombination and stability of perovskite QDs.^[154]

Recent research indicated that novel surface ligands such as quaternary ammonium salts, phosphonic acids,^[120] and amphoteric molecules show the stronger adhesion to the surface of nanocrystals. In addition, in order to achieve proper carrier injection capability, the length of the ligand should also be taken into consideration. Octylphosphonic acid (OPA) are proven to have stronger binding interaction than oleic acid, which allows the perovskite to endure multiple purifications.^[120] At the same time, a uniform and compact nanocrystals film with an impressive stability can also be obtained. Benefiting from the reduction of leakage current and defect density, the device exhibits satisfactory performance (Figure 7A and 7B). L-phenylalanine is an aromatic bi-functional ligand, which prefers to coordinate with both the cations and anions on the QDs surface.^[154] The introducing of L-phenylalanine during colloidal synthesis can fill the surface vacancy and stabilize the QDs eventually.^[155] Indeed, L-phenylalanine possess larger adsorption energy than OLA and OA, which is beneficial for stabilizing the QDs. Furthermore, the formation energy of vacancy is also enhanced, indicating the advantages of this ligand

(Figure 7C and 7D). Based on the hard and soft acids and bases theory, quaternary ammonium bromide can bind to under-coordinated lead sites robustly, decreasing the surface trap state of perovskite nanocrystals.^[119]

While the passivation based on organic molecules can eliminate some of defect states, such dielectric coatings will introduce an undesirable barrier for charge injection and dramatically reduce the conductivity of perovskite QDs films. Introducing inorganic ligands should be an effective way to overcome the challenges mentioned above.^[156] Potassium halide (KBr or KCl) was proved to be an effective ligand for this purpose. Different from the A-site substitution mentioned in 3.1.1, K ions were preferred to combine with halogen anions on the surface of perovskite through ionic bonds, reducing the surface defects and organic ligands (Figure 7E). Therefore, the fluorescence performance and conductivity of film were enhanced simultaneously.^[157] However, it is worth pointing out that excessive K^+ in the system will degrade the morphology of film because of K^+ can affect the growth of crystals, resulting in a bumpy surface. Another work indicated that zirconium acetylacetonate could be introduced into CsPbI_3 QDs for passivating the surface and adjusting the recombination rates.^[117] High-resolution XPS spectra show that this inorganic ligand is more likely to combine with Pb^{2+} and I^- on the surface of nanocrystals, suppressing the diffusion of these ions.^[158] At the same time, it is obvious that the energy levels of CsPbI_3 QDs were raised with the introduction of zirconium acetylacetonate, and the electron and hole mobilities are getting closer in modified QDs films. Therefore, the Auger recombination caused by unbalanced charge transport was suppressed effectively. In addition, organic-inorganic co-passivation is also an effective way to stabilize perovskite structure. For instance, trioctylphosphine-oxide (TOPO) can easily bind with Zn ion to form TOPO-Zn complex.^[159,160] This organic-inorganic complex with Lewis acid properties can effectively combine with halide and attach to the surface of perovskite QDs. Since O^{2-} tends to react preferentially with outer Zn rather than with other parts, the stability is improved.

The proper ligand exchange using shorter chains with stronger adhesion could be considered to improve the stability of nanocrystals and shrink the QD-to-QD distance in films, resulting in the fascinating device performance.^[160] In this process, various ammonium salts are generally used and the chain length of ligands is also optimized to achieve proper carrier injection^[119,121,161] (Figure 7F and 7G). In the mixed perovskite, I ions with a larger ionic radius induce the decreased decomposition energy.^[28] Ligand exchange method in this system is proven to the passivation of the perovskite nanocrystals while preserving the cubic shape of QDs.^[162] Through

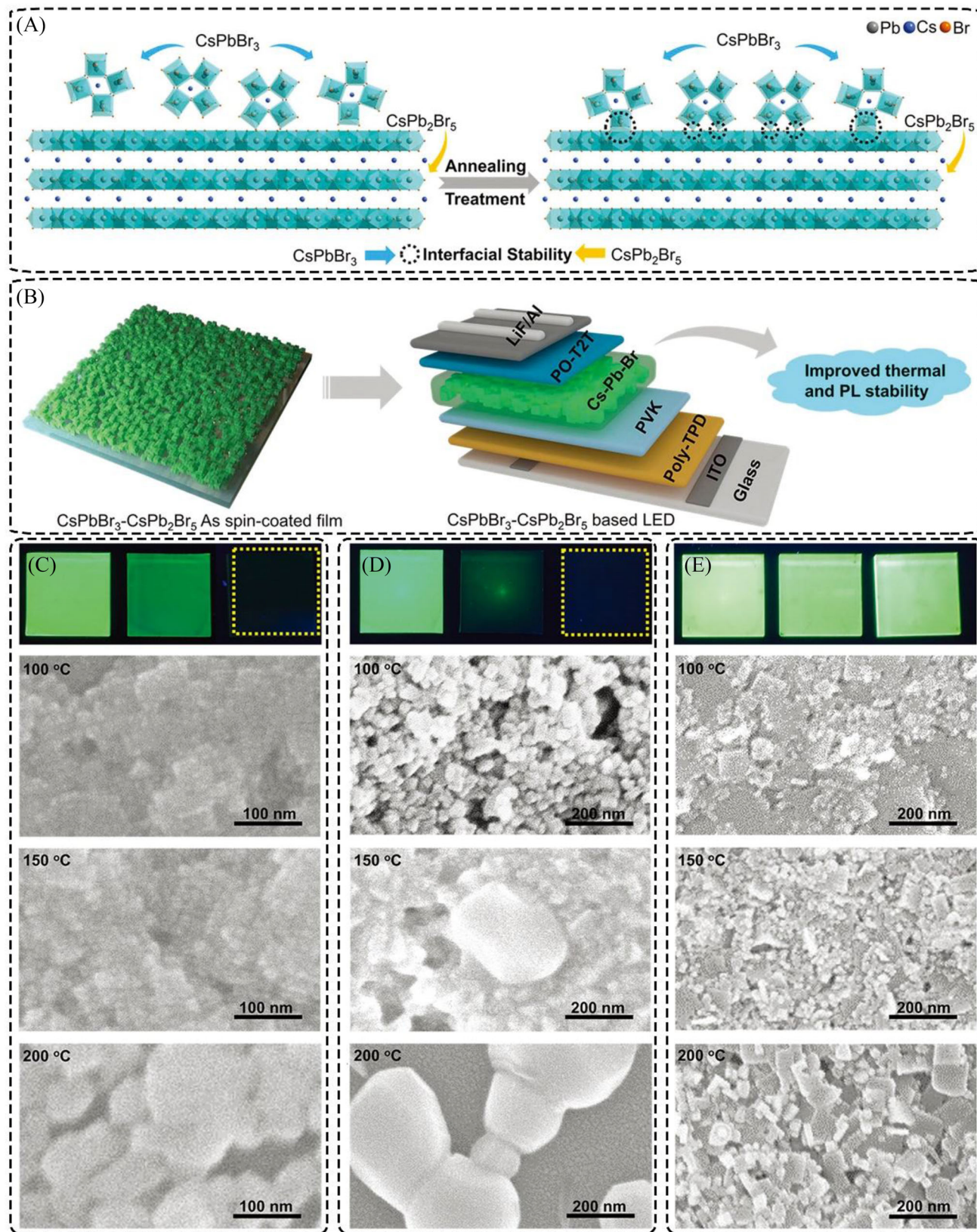


FIGURE 6 A, Schematic illustration of the interfacial stability of dual phase CsPbBr₃-CsPb₂Br₅. B, Schematic illustration of a typical multi-layer structured perovskite QLEDs device by using CsPbBr₃-CsPb₂Br₅ nanocrystals as stable green light emitters. C, Green light emission photographs and typical SEM images of all inorganic Cs-Pb-Br perovskite nanocrystal films on glass slides (C) for HI, (D) for LARP-SP, (E) for LARP-DP treated with different annealing temperatures between 100 and 200 °C under ambient conditions. Reproduced with permission.^[113] Copyright 2018, Royal Society of Chemistry

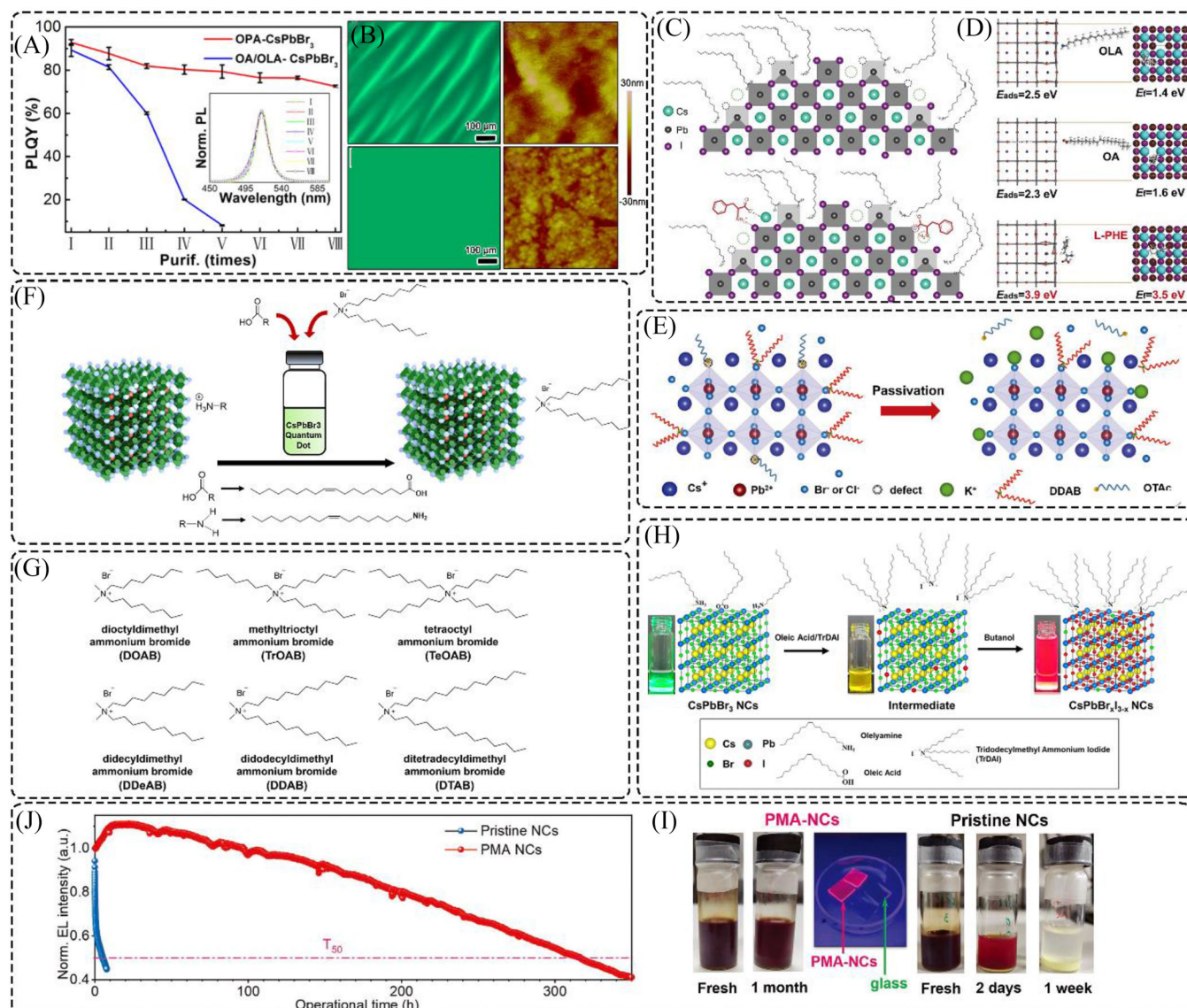


FIGURE 7 A, PL QY of OPA-CsPbBr₃ and OA/OLA-CsPbBr₃ nanocrystal solution in hexane with one to eight purification cycles. Inset shows the PL spectra of OPA-CsPbBr₃ NC solution with different purification cycles. CsPbBr₃ NC films' morphologies. B, Fluorescence photographs of OA/OLA-CsPbBr₃ and OPA-CsPbBr₃ NC films purified with two cycles. AFM height images of OA/OLA-CsPbBr₃ and OPA-CsPbBr₃ NC films with two purified cycles, respectively. Reproduced with permission.^[120] Copyright 2018, American Chemical Society. C, The surface of as-synthesized CsPbI₃ perovskite QDs with and without L-PHE passivation. D, DFT calculated adsorption energy (E_{ads}) of OA, OLA and L-PHE ligands on the optimized CsPbI₃ surfaces, and the corresponding formation energy of vacancy (E_f) on CsPbI₃ surfaces after the adsorption of different ligands. Reproduced with permission.^[155] Copyright 2020, John Wiley and Sons. E, Schematic illustration of potassium passivation. Reproduced with permission.^[157] Copyright 2020, John Wiley and Sons. Schematic of (F) ligand exchange process and (G) chemical structures of various QAB ligand materials with different bulkiness and length. Reproduced with permission.^[161] Copyright 2019, American Chemical Society. H, Schematic of the ligand exchange process and chemical structure of ligand materials and optical properties along with treating the ligand, TrDAI for 0.46 wt % concentration CsPbBr₃ nanocrystals. Reproduced with permission.^[28] Copyright 2020, American Chemical Society. I, Comparison of the aged colloidal β -CsPbI₃ nanocrystals with and without PMA incorporation. J, The operational lifetime of the CsPbI₃ perovskite QLEDs at a constant current density of 30 mA cm⁻². Reproduced with permission.^[128] Copyright 2021, John Wiley and Sons

the exchange strategy using tridodecylmethyl ammonium iodide (TrDAI), halogen vacancies on the surface are effectively passivated and band energy is increased, leading to efficient charge carrier collection, which can be used to obtain the perovskite LEDs with satisfactory optical properties and structural stability (Figure 7H).

In addition to the methods mentioned above, the use of cross-linkable ligands to protect the perovskite is another way to enhance the stability. Recently, Choy and his co-workers demonstrated that poly(maleicanhydride-alt-1-octadecene) (PMA) can be used as a robust ligand to improve the optical performance and stability of perovskite.^[128] Benefiting from the strong bonding energy of Pb-O bond, PMA can regulate the crystallization kinetics of nanocrystal and adsorb on the surface of nanocrystals, stabilizing the perovskite without any crystalline deformation (Figure 7I). Furthermore, these chemically crosslinked PMA increase the formation energy of Pb_{Cs} and passivate the Pb_{Cs} deep defects, ending perovskite outstanding optical performance. It is also worth mentioning that these ligand molecules with the amphiphilic nature and large steric hindrance tend to the formation of a hydrophobicity layer on the surface of perovskite, which will prevent the attacking of polar solvents during the purification. Ultimately, PMA-based perovskite QLEDs show superior operational stability, increasing approximately 60-fold compared to the control one (Figure 7J).

3.1.5 | Moderate purification method

Presence of excess precursors and ligands during the synthesis and post-treatment of perovskite can cause the instability of the required perovskite phase, thereby hindering the injection of charge carriers. In addition, the presence of impurities or residual polar solvents are proven to adversely affect the structure and photophysical properties of perovskite QDs.^[153,163–166] Thus, the selection of purification and dispersion solvents are very important for perovskite QDs. Given the ionicity of perovskites and the weak binding of surface ligands, the solvents with high polarity could completely destroy the structure of perovskite. In contrast, the solvents with low polarity are preferred to be used to purify perovskite. Li et al. have reported the effect of different purification solvents on the properties of perovskite QDs.^[167] The results show that octane and hexane should be used to disperse perovskite QDs since their similar polarity with surface ligands. Compared with 1-butanol, acetone, ethyl acetate and other commonly used solvents, methyl acetate can successfully extract the stable CsPbI₃ QDs with cubic phase and obtain the phase-stable devices.^[26] Furthermore, an innovative method was developed recently for the halide

exchange in CsPbBr₃ QDs using an aqueous-organic solvent system.^[168] The obtained nanocrystals in cubic phase offer high stability and PLQY in both solution and films.

3.2 | Improving the stability of constituent layer

Considering the sandwich structure of LEDs, a uniform perovskite QDs film and robust transport materials as well as sufficient surface contact can effectively improve the stability of the device. From the perspective of emitting layer, the suitable additives or self-assembly of QDs will be beneficial to form a uniform and dense film, which can facilitate charge injection and prevent leakage current. As for transport layers, the stable materials with suitable energy level can not only guarantee the carriers transport efficiently, but also protect the vulnerable perovskite active layer. The unbalanced carrier injection in LEDs is one of the main factors that reduce the stability and efficiency of device. In addition, control over the interface properties aimed on the efficient transport and minimized trapping of charge carriers should also be taken into consideration.

3.2.1 | Construction of uniform emitting layer

Efforts for optimizing QDs mainly focus on improving the properties of nanocrystals in solution. However, the structure and performance of these nanocrystals degrade sharply after transferring to the substrate. Such as, a rough surface will induce the leakage current due to the formation of electrical shunt paths, limiting the electroluminescence efficiency.^[122] Therefore, a uniform QDs film is necessary for achieving efficient devices. To obtain homogeneous perovskite thin films, a general method is the healing treatment, which may result in uncontrollable cracks because of the volume contraction and unexpected redissolution of perovskite.^[169,170] The alternative strategy is to use appropriate additives to control film formation. The drop-casting deposition method with the help of PMMA is proven to form a uniform film without the aggregation of QDs.^[83] Due to the unique properties such as high heat resistant and low moisture absorption of PMMA, the compound film represents excellent air stability. And less EL blinking can be observed under a continuous bias condition (Figure 8A and 8B). However, excessive addition of PMMA will decrease the performance of perovskite QLEDs because of the insulating property of PMMA.

The controllable assembly of nanocrystals is beneficial to maintain high PLQY and structure integrity simultaneously, as well as forming high-quality QDs films.^[63] At high concentrations, the interparticle distance of

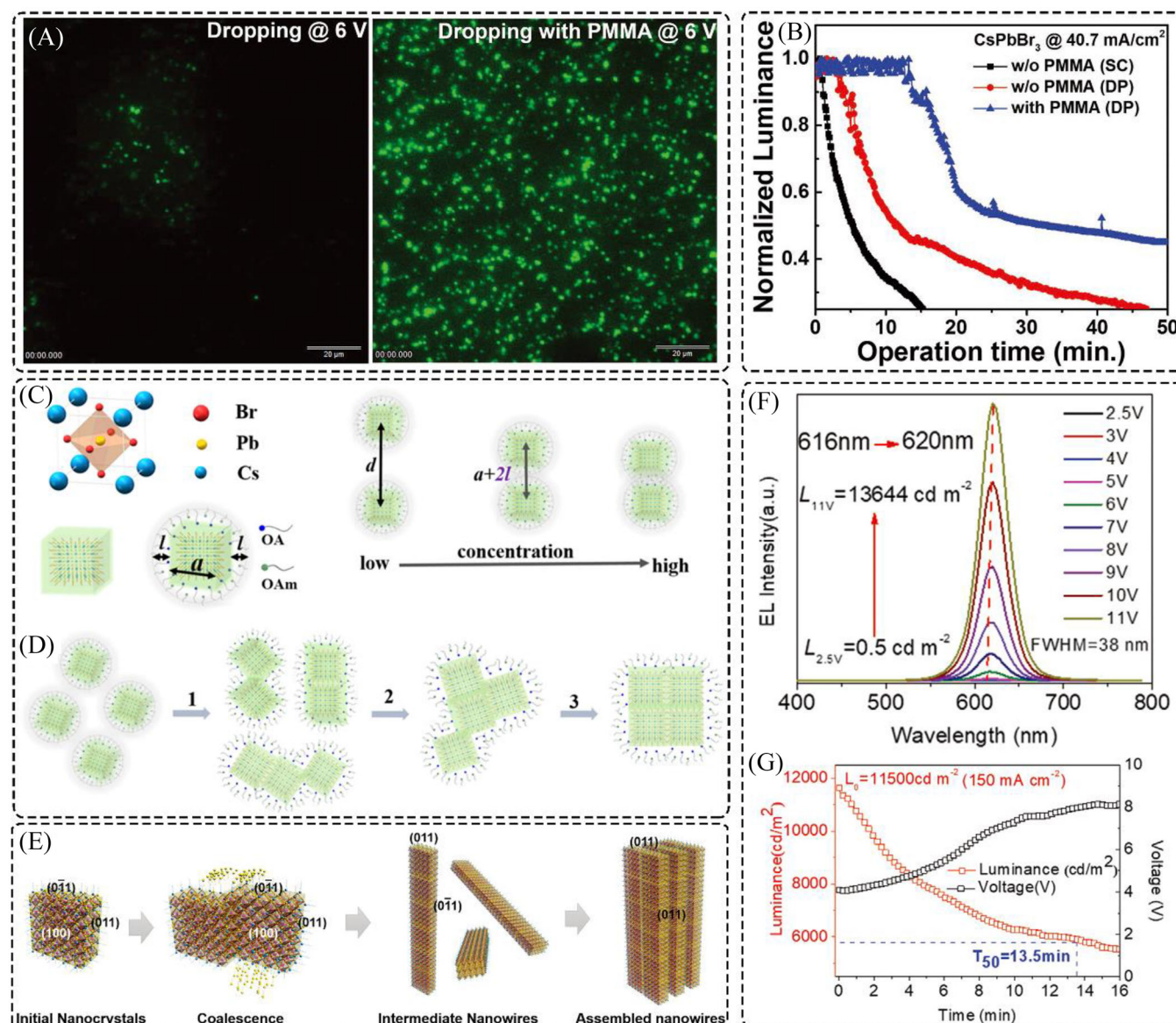


FIGURE 8 A, Optical microscopy image of EL from a perovskite LED with CsPbBr₃ nanocrystal films deposited without and with PMMA at a constant voltage. B, The long-term stability levels of unencapsulated perovskite QLEDs with CsPbBr₃ nanocrystal films prepared by different deposition methods without and with PMMA were evaluated in terms of normalized luminance under ambient air atmospheric conditions (25°C, 55% humidity, with continuous operation). Reproduced with permission.^[83] Copyright 2017, John Wiley and Sons. C, Schematic illustration of crystal structure, the ligands capped nanocrystal and interaction between nanoparticles change with the concentration. D, Illustration of the model for solvent-assisted interface engineering. Reproduced with permission.^[63] Copyright 2018, American Chemical Society. E, Illustration of the shape evolution self-assembly process from initial nanocrystals to intermediate NWs and then to the final assembled NWs; (F) EL spectra with different operated voltage; (G) Constant driving current of 6 mA (150 mA cm²) led to the luminance increasing from 0 to 11 500 cd m² (L₀) and then diminishing. The estimated operational half-life (T₅₀) at 100 cd m² is 694 hours. Reproduced with permission.^[115] Copyright 2020, John Wiley and Sons

nanocrystals will get smaller, and these QDs have the potential to assemble for maximizing the total entropy of the system.^[171] And the dominated interaction could be controlled by the chain length of ligands, solvent polarity and so on (Figure 8C). Benefiting from the sensitivity of perovskite to solvent polarity and dynamic combination with ligands, a previous work reported a method to achieve

the perovskite assembly by adjusting the solvent.^[167,172] As shown in Figure 8D, the hexane was utilized as the trigger for obtaining one-dimensional assemblies in step 1. Ethyl acetate was employed in step 2 to terminate the one-dimensional assembling process and drive the organization in two-dimensional orientation. Finally, the assembly process was ended by evaporating ethyl acetate and

the final products possess a controllable size. This solvent-assisted strategy can activate the self-assembly of QDs and ensure the fabrication of smooth and dense CsPbBr₃ films, which is highly desirable to be used in device.^[63] Perovskite nanowires (NWs) are proven to heal the defects during growth.^[173] A recent research shows that halide vacancies on the surface of nanocrystals can be utilized as ionic anchoring sites to mediate assembly. In this process, excess amine can dominate the anisotropic growth of the QDs, resulting in the formation of NWs, which could be further assembled into clusters driven by vacancy.^[115] It is gratifying that surface defects are self-healed during this process, resulting in a low density of trap states and excellent stability, which could be further made into high-performance devices (Figure 8E-G).

3.2.2 | Efficient charge injection through interface modification

Poor contact and large energy barrier between emission layer and charge transport layers can lead to the charge accumulation, which is harmful to the stability of device. Therefore, it is necessary to control the interface properties in order to achieve effective injection of charge carriers.^[174–176] There are various interfacial materials including small molecules, polymers and inorganic modifiers employed toward this goal. Generally, an ideal interface modifier should possess the following advantages: (i) be able to form compact films easily; (ii) be able to improve the properties of interfaces, such as minimizing the charge injection energy barrier, passivating surface traps, improving the interface contact, suppressing metal ion diffusion and so on; (iii) possess negligible absorption; and (iv) its energy levels should be easily to adjust. Perfluorinated ionomer (PFI) has been widely used as interfacial layer or mixed with metal oxides to optimize the charge injection of LEDs.^[177,178] The self-assembly of this material not only makes the emitting layer show the satisfactory morphology, which increase the contact of constituent layers, but also modifies the energy barrier and makes the carrier transport more convenient. Benefiting from these advantages, the device shows better performance and stability.^[92] Carbon dots (CDs) as a cheap and easy to process material can serve as the surface modifiers to minimize the charge injection/extraction energy barrier. Because of the adjustable surface groups, CDs can make the well-matched energy band alignments, thereby improving device performance^[179] (Figure 9A and 9B). As a cage-like molecular structure with an inorganic siloxane core and eight surrounding organic corner groups, polyhedral oligomeric silsesquioxane (POSS) containing thiol groups can efficiently attach to perovskite

surface and block the undesirable anion exchange.^[180] A previous report indicated that the use of POSS can not only help to keep both electrons and holes located within the emission layer, but also improve the surface contact of the film, leading to a dramatic enhancement of operation lifetime.^[127] It is well known that the perovskite layer is at the center of the sandwich structure between the carrier transport layers in practical devices. Thus, the interface treatment on both sides of perovskite QD film may provide a good way to improve device efficiency and stability. Recently, a bilateral modification strategy was proposed to passivate both top and bottom interfaces of QD film with organic molecules.^[116] Profiting from the strong interaction between the P = O and surface Pb atom, bilateral-passivated molecules could prevent the loss of ligands and endow the films with enhanced stability (Figure 9C and 9D). Interface would provide a flat channel for carrier transportation when defects are passivated, which means the device can reach the same brightness while requiring less current compared to the control one. Furthermore, passivation layer could also prevent the ion migration, leading to the better device stability (Figure 9E-H).

3.2.3 | Robust transport layer with balanced carrier transport capability

The unsatisfactory operating stability of the LEDs can also be attributed to the absence of robust transport layers which lack of sufficient transport capacity of carriers and the effective protection of perovskite active layer.^[59,181] The unbalanced carrier injection in LEDs is one of the main factors that reduce the stability and efficiency of device. Generally, the mobility of electron is faster than hole, which can be effectively reduced by optimizing the electron and HTL materials.^[182] A cross-linkable VB-FNPD (9,9-Bis[4-[(4-ethenylphenyl)methoxy]phenyl]-N2,N7-di-1-naphthalenyl-N2,N7-diphenyl-9H-fluorene-2,7-diamine) was developed as HTL to improve the stability of perovskite QLEDs.^[159] Benefiting from the proper HOMO and LUMO levels, this new HTL with relatively high hole-carrier mobility not only decrease the turn-on voltage and balance the holes and electrons in the device, but also provide the better morphology of the perovskite QDs film, leading to the increased efficiency and lifetime of the device. Functional modification of polymers can be achieved by combining different molecules conveniently, which enable them to be designed as effective transport layer materials. Friend and his co-workers prepared a series of conjugated polyelectrolytes (CPEs) as the hole injection layer used in LEDs.^[125] The backbone, counterions, and side chains of polymer can be used to adjust the energy and electrical transport properties to achieve

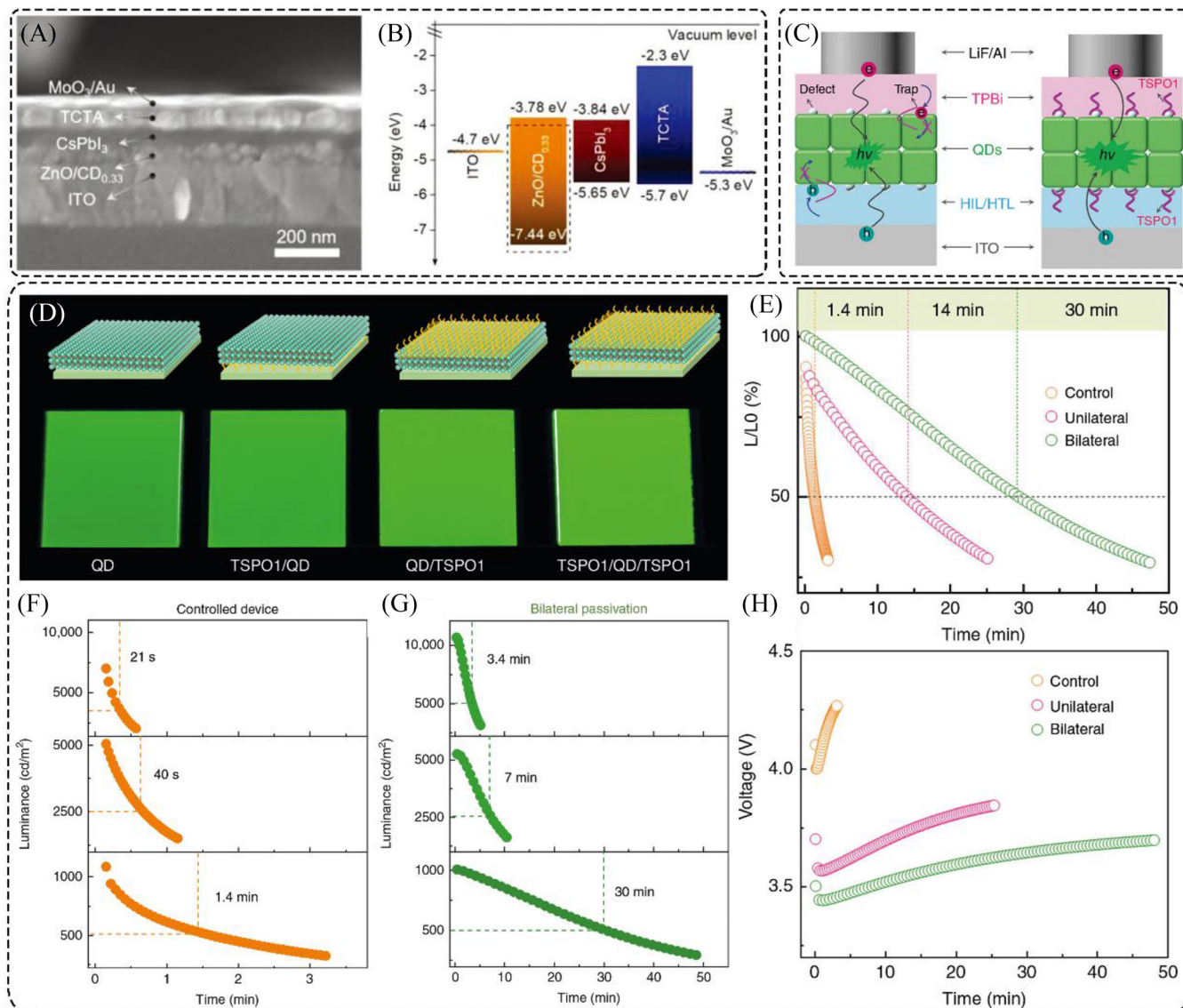


FIGURE 9 A, Cross-sectional SEM image of the CsPbI₃ QD LED, (B) its energy band diagram. Reproduced with permission.^[179] Copyright 2020, John Wiley and Sons. C, The structure of QLED based on QD films passivated without and with passivation, schematic illustration that TSPO1 could passivate defects on the surface of QD films, the defects may trap carriers (e.g., holes, electrons), decrease exciton recombination, and hence degrade the device performances. D, Schematic diagram and photograph of QD films without passivation and with TSPO1 on the bottom side, on the top side, on both sides of QD film under UV light. E, Operational lifetimes of pristine and bilateral-passivated QD films at an initial luminance of about 1000 cd m⁻². F, Operational lifetimes of pristine device tested at different initial values of luminance, 1000 cd m⁻², 5000 cd m⁻², and 7000 cd m⁻². G, Operational lifetimes of bilateral-passivated device tested at different initial luminance, 1000 cd m⁻², 5000 cd m⁻², and 10000 cd m⁻². H, Voltage shifts of perovskite QD LEDs at a constant current density. Reproduced with permission.^[116] Copyright 2020, Springer Nature

effective hole injection and electron blocking behavior (Figure 10A and 10B). Benefiting from these virtues, the long-term stability of perovskite QLEDs are enhanced.

However, considering the inherent chemical instability of organic transport materials, the long-term stability of device can still not be guaranteed.^[183] Recent studies have shown that inorganic carrier injectors could be a better choice.^[18,59,184] On the one hand, such inorganic materials show more stability against oxygen, moisture and temper-

ature, endowing the device the long-term survival ability in harsh environment.^[185] On the other hand, the tunable band gaps could form a desirable band alignment with perovskite QDs film, facilitating the injection and confinement of carrier. More importantly, these materials can be used as moisture/oxygen diffusion barriers to protect the emitters due to their desirable scavenging effect^[59,186] (Figure 10C and 10D). The n-type ZnO can act as the electron-injection layer thanks to the appropriate valence-band

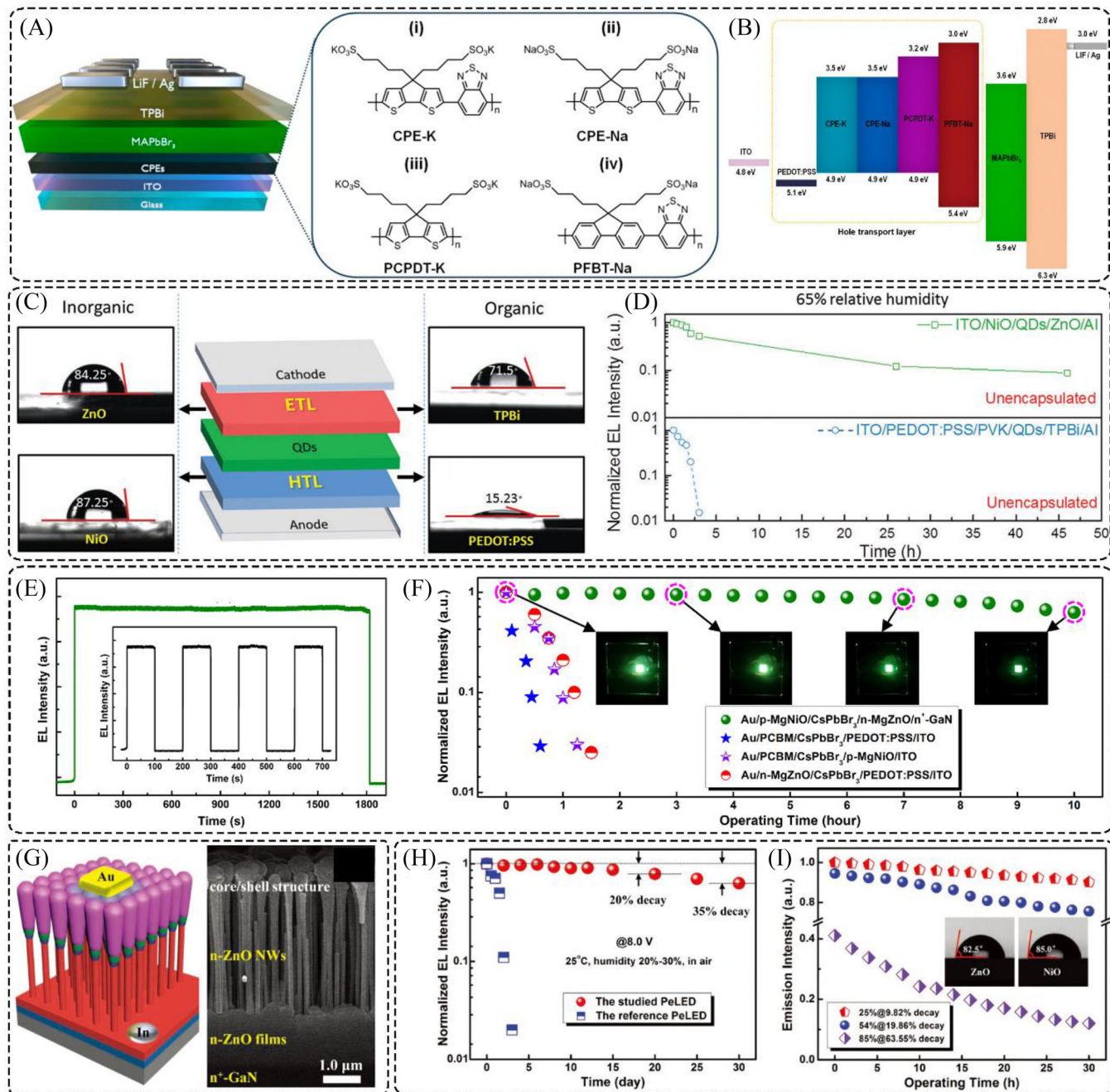


FIGURE 10 A, Device architecture of perovskite QLEDs and chemical structure of CPEs. B, Energy diagrams of perovskite QLEDs for flat band conditions. Reproduced with permission.^[125] Copyright 2018, American Chemical Society. C, Hydrophilic comparison of different charge transport layers. Water contact angles of inorganic charge transport layers (ICTLs) and the commonly used organic charge transport layers (OCTLs) on ITO substrates. D, Normalized luminance versus time of an all-inorganic QLEDs and QLEDs with organic CTLs (ITO/PEDOT:PSS/PVK/QDs/TPBi/Al) under 65% relative humidity. Reproduced with permission.^[18] Copyright 2017, Royal Society of Chemistry. E, Simplified energy band alignment of the multilayered heterostructure showing the conduction and valence band energy levels with respect to the vacuum level. F, Emission intensity of the studied perovskite QLEDs and three reference perovskite QLEDs as a function of running time under a continuous bias of 10.0 V. The insets show the corresponding photographs of the perovskite QLEDs after different running periods. Reproduced with permission.^[59] Copyright 2017, American Chemical Society. G, Schematic diagram of the Au/p-NiO_x/CsPbBr₃ QDs/MgZnO/Au NPs/n-ZnO/n⁺-GaN heterostructure LED and Cross-sectional SEM image of the NiO_x/CsPbBr₃ QDs/MgZnO/Au NPs/ZnO coaxial NWs. H, Device performance of the plasmonic perovskite QLEDs and the reference perovskite QLEDs as a function of storage time in an ambient environment. I, Emission decays of the plasmonic perovskite QLEDs after different running periods under different humidity conditions. The insets show the water contact angles of ZnO and NiO_x layers deposited on n⁺-GaN/c-Al₂O₃ substrates. Reproduced with permission.^[19] Copyright 2018, John Wiley and Sons

energy level. And the band gap could be further modified by decreasing particle size according to quantum confinement effect.^[124,187] In other words, the valence-band offset at the interface between ZnO and CsPbBr₃ becomes larger for smaller ZnO nanoparticles, implying an excellent hole-blocking effect and effective leakage suppression. Moreover, incorporating Mg into ZnO will further induce a decreased electron affinity and a descending valence band, which can not only act as a robust diffusion barrier against H₂O, but also protect the perovskite from the metal contact.^[126] Analogously, nickel oxides (NiO_x) can be used as the hole-injection layer to effectively ameliorate the unbalanced carrier injection in device.^[188] But, NiO_x shows mass nonradiative decay channels at the interface of HTL, limiting the device performance evidently.^[189] Choy's group adopted a bilayer HTL structure composed by a polymer layer of poly(sodium-4-styrene sulfonate) (PSSNa) and NiO_x to solve this problem. The PSSNa will reduce the defect of NiO_x films, suppressing the nonradiative recombination. Furthermore, the lower valence band of the bilayered structure can benefit the efficient hole injection, leading to the better spectral stability and operation lifetime.^[190] In addition to bilayer HTL structure, the MgNiO_x appears to be a more attractive candidate as the HTL material due to its wide and tunable bandgap and inherent material stability (Figure 10E and 10F). Previously, a perovskite QLEDs was designed based on vertically aligned one-dimensional ZnO nanowires arrays.^[19] (Figure 10G) ZnO and NiO_x were employed as the electron and hole-injection layer respectively in such perovskite QLEDs. On the one hand, the high humidity tolerance of the studied perovskite QLEDs benefits from the well-designed all-inorganic coaxial core/shell heterostructures. On the other hand, it has been generally accepted that these inorganic metal-oxide could prevent water from penetrating into the emitters, maintaining the lifetime of device effectively (Figure 10H and 10I).

3.3 | Optimized device structures and driving modes

Joule heat inevitably generated under working conditions will significantly affect the stability of QLEDs.^[117,98] Unfortunately, poor thermal conductivity of device can cause the efficiency roll-off even degradation.^[100] Si substrate with high thermal conductivity was used in a recent report to construct a top-emitting device structure for improving the stability of device.^[117] Because the Si has a high thermal conductivity, the device can dissipate Joule heat generated at high current density efficiently, leading to the stable perovskite QLEDs (Figure 11A). In addition to thermal conduction, suppressing heat generation is another

way to reduce Joule heating. In previous works, the perovskite QLEDs usually working under a direct current (DC) driving condition.^[191,192] The performance drops off dramatically at high driving voltage due to Joule heat and charge accumulation. The alternating current (AC) condition with the reversal voltage and a short operating time can solve the problems mentioned above^[122,123] (Figure 11B-F). Two factors can be used to explain the improvement of device performance under AC driving mode. The first one is the suppressed heating generation since the less operation time compared with the DC one. Another one is the reduced charge accumulation due to frequent reversal of the applied bias.^[193] The carriers will be trapped in the defect states located in emission layer and act as the space charges to impede the following injection under DC mode. In contrast, with applying AC bias, these trapped carriers are more likely to be extracted. Consequently, the device performance and endurance are improved under AC mode.

4 | CONCLUSIONS AND OUTLOOK

In this review, we generalize the origin for the instability of perovskite QLEDs into two aspects. (i) As the emitting material, the ionicity of the nanocrystals and the weak interaction between the crystal and surface ligands endue perovskite QDs with fragile characteristics, which will not only affect the luminescent properties of perovskite, but also damage the device stability greatly. And the degradation can also be triggered by external environments such as humidity, voltage, temperature and illumination. (ii) The inherent instability of the constituent layer materials and the poor contact among them will induce inefficient and unbalanced carrier injection as well as generate defects, leading to degeneration of charge-transport layers and considerable Joule heating. Furthermore, diffusion of metallic species from electrodes can also accelerate the degradation of perovskite QLEDs.

Various reported strategies to overcome the instability of perovskite materials and the limited operational stability of perovskite QLEDs are summarized. By analyzing the working mechanism of these high-performance devices, we believe that the stability of perovskite QLEDs can be improved from the following aspects. (i) Improving crystal formation energy and choosing proper ligands to obtain the robust perovskite QDs with low defect density. For the former, the smaller inorganic A-site and B-site atoms can induce lattice shrinkage, forming highly crystalline perovskite materials which show better thermal and humidity stability. At the same time, the energy levels match better between the perovskite and charge-transport materials, facilitating carrier injection. For the latter, novel

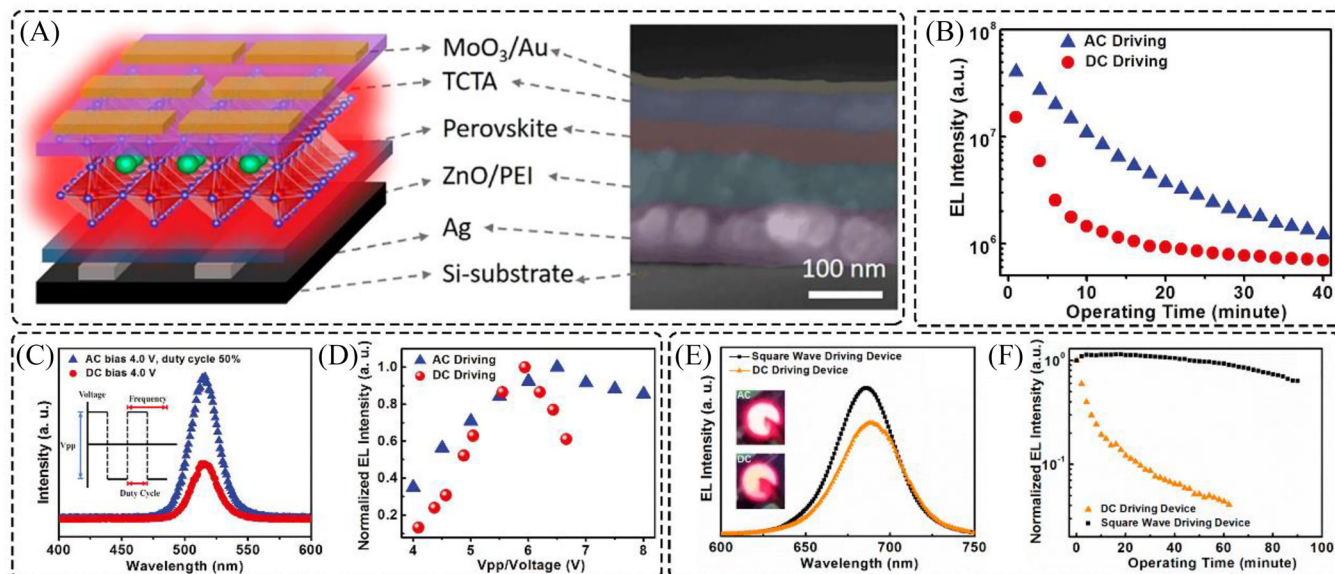


FIGURE 11 A, Schematic diagram and cross-section SEM image of the top-emitting perovskite QLEDs. Reproduced with permission.^[117] Copyright 2020, American Chemical Society. B, Emission intensity of the studied perovskite QLEDs as a function of running time under AC (V_{pp} = 4 V; frequency: 10 Hz; duty cycle: 50%) and DC (voltage 4 V) driving conditions. C, The EL spectra of CsPbBr₃ QDs LED under AC (V_{pp} = 4 V; frequency: 10 Hz; duty cycle: 50%) and DC (voltage: 4 V) driving conditions. The inset is a schematic diagram of the square-pulsed bias. D, Normalized EL intensity of CsPbBr₃ QDs LED as a function of voltage under DC and AC driving conditions (frequency: 10 Hz; duty cycle: 50%). Reproduced with permission.^[122] Copyright 2018, John Wiley and Sons. E, The EL intensity of devices under DC (8 V) and AC (square pulsed bias, duty cycle: 50%, frequency: 10 Hz, V_{pp}: 8 V) driving conditions. The insets are the digital pictures showing the red-light emission from devices under AC and DC driving conditions. F, Normalized emission decays of the perovskite QLEDs after different running periods under DC (8 V) and AC (square pulsed bias, duty cycle: 50%, frequency: 10 Hz, V_{pp}: 8 V) driving conditions. Reproduced with permission.^[123] Copyright 2018, MDPI

surface ligands such as quaternary ammonium salts, phosphonic acids, and amphoteric molecules with appropriate chain length will not only show the stronger adhesion to the surface of nanocrystals, protecting QDs from being destroyed during purification and film formation, but also adjust proper carrier injection capability for the balanced charge-carrier transport. (ii) Robust constituent layers with efficient and balanced carrier transport capacity. Optimizing the morphology of the emitting layer by suitable additives, self-assembly of QDs and interface modification will be benefit to form a uniform and dense film, which limits substantial leakage current and inefficient carrier injection due to the reduction of electrical shunt paths. Chemically stable charge-transport materials with suitable energy level can not only guarantee the balanced transport of carriers, but also protect the vulnerable perovskite active layer from exposure to air, improving device stability significantly. (iii) Adopting appropriate device structures and driving modes to reduce Joule heat. High thermal conductivity substrate and intermittent drive current can diffuse and reduce heat generation effectively, ensuring long-term operation of the device.

We believe that efficient and stable perovskite QDs are the basis of robust device. Although inorganic ion

doping and the use of novel ligands can help stabilize the perovskite lattice, these strategies are difficult to obtain commercially stable QDs considering the ionicity of perovskite. Polymers are used as additives frequently to inhibit the aggregation of nanomaterials, thereby improving film morphology. Generally, they are easily grafted with various chemical groups to achieve the functionalization. In addition, the network structure of these polymers can also limit the movement of molecules and ions as well as protect the materials from being corroded by water and oxygen. A recent report has successfully used polymer in the synthesis of perovskite QDs, and has achieved a stable QLEDs with considerable EQE.^[128] Therefore, it is possible to achieve the ultra-high stability of perovskite QDs by using polymer as a macromolecular ligand. Further research should focus on the grafting of polymers and organic molecules (such as quaternary ammonium salts, phosphoric acid and amphoteric molecules) in order to achieve the more stable combination with perovskite. At the same time, adjusting the energy level of the polymer through functionalization also needs to be taken into account in order to achieve more balanced carrier transport. Furthermore, other elements, such as stable transport materials and better device structure,

affecting stability cannot be ignored. It should be pointed out that even though the inorganic transport layer has a positive effect on stability compared to the organic one, there are still some shortcomings affecting the stability of QLEDs. For instance, considering the perovskite QLEDs will be wet-processed and operated in air, the hydrolysis of ZnO nanoparticles will accelerate the degradation of perovskite films. Therefore, the novel and robust inorganic charge-transport materials which insensitive to moisture should be investigated. We expect that this review can help researchers to comprehend the instability of perovskite QLEDs and make favorable suggestions for future research on improving stability. Considering the immense improvements over a rather short span of time, the next-generation display technology based on perovskite QLEDs will soon be created once its stability problem is resolved.

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DATA AVAILABILITY STATEMENT

Data sharing is not applicable to this article as no new data were created or analyzed in this study.

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