

SPECIAL ISSUE ARTICLE

Organic additive engineering toward efficient perovskite light-emitting diodes

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Abstract

Perovskite materials with excellent optical and electrical properties are promising for light-emitting diodes. In the field of perovskite light-emitting diodes (PeLEDs), organic materials additive engineering has been proved to be an effective scheme for enhancing efficiency and stability in PeLEDs. Most impressively, the reported external quantum efficiency of PeLEDs based on perovskite-organic composite has reached over 20%. Herein, we will review the important progress of the organic materials' additive-modified PeLEDs and discuss the remaining problems and challenges and the key research direction in the near future.

KEYWORDS

perovskite, light-emitting diodes, organic, additive, stability

1 | INTRODUCTION

Metal halide perovskites with excellent electrical, optical, and magnetic properties have been widely researched in the semiconductor field, including photovoltaics,¹⁻⁹ light-emitting diodes,¹⁰⁻¹⁵ detectors,¹⁶⁻²⁰ lasers,²¹⁻²⁴ and memristors.²⁵⁻²⁷ The general chemical formula of perovskite materials is ABX_3 , where A is a monovalent cation, such as $CH_3NH_3^+(MA^+)$, $CH(NH_2)_2^+(FA^+)$, Cs^+ ; B is a divalent cation, such as Pb^{2+} , Sn^{2+} ; X is a halide anion, such as Cl^- , Br^- , and I^- .²⁸⁻³⁰ Varying the component of A, B, or X can widely tailor the bandgap of perovskites, which meets the precondition of achieving different colors from red to green to blue. Furthermore, perovskite materials demonstrate high photoluminescence (PL), color purity, and carrier mobility. Hence, metal halide

perovskites are promising materials for next-generation, light-emitting diodes.

The perovskite light-emitting diodes (PeLEDs) in early 1990s reports were achieved at liquid nitrogen temperature.³¹ The room temperature PeLED was reported in 2014.³² After that, intense efforts have been made to improve the efficiency of PeLEDs. Lee and coworkers demonstrated a $MAPbBr_3$ -based PeLEDs with an external quantum efficiency (EQE) of 8.5%.³³ Afterwards, effective PeLEDs were reported based on two-dimensional (2D) perovskite materials^{15,34-38} and perovskite quantum dots (QDs).³⁹⁻⁴² Recently, organic materials' additive engineering has been proven an effective way to achieve efficient PeLEDs. The EQE based on the perovskite-organic composite has reached 21.6% for near-infrared light.⁴³

In this review, we discuss the application of organic additive materials in perovskite emitting layers. In Section 2, we discuss the effect of organic additives in

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PeLEDs and the progress of PeLEDs based on the perovskite-organic composite. In Section 3, we present a summary of perovskite-organic composite films serving as a light conversion layer for displays. Furthermore, in Section 4, we outline the remaining problems and challenges of PeLEDs and present possible research directions in the near future. Finally, we present a conclusion and outlook in Section 5.

2 | PEROVSKITE LIGHT-EMITTING DIODES

Generally, the perovskite materials as emitting layers for PeLEDs can be classified into 3D structure, 2D (or quasi-2D) structure, and QDs (or nanocrystals, NCs). All three structures have led to impressive PeLED device performance. On the other hand, there are several problems in terms of device efficiency and stability, which need to be resolved. Mixing organic materials (including small organic molecules as well as polymers) into perovskite materials has been reported to be a promising method to resolve these problems.

2.1 | 3D PEROVSKITES

The EQE of PeLEDs based on 3D perovskites was lower than 1% at the beginning.³² One reason responsible for lower EQE was the poor film morphology that restricted

the performance severely. Radiative charge recombination in the emitting layer is a prerequisite for efficient PeLEDs. However, poor quality perovskite films tend to have pinholes, which act as shunting paths for the injected charges. A large number of electrical charges bypass the perovskite layers through pinholes, which causes undesirable leakage current.

To reduce leakage current, a uniform perovskite emitting layer is preferred. Greenham and coworkers introduced a polymer material polyimide (PIP) into MAPbBr₃ perovskites forming a perovskite-PIP composite system.⁴⁴ Upon mixing of PIP, perovskite NCs were embedded into the PIP matrix. A uniform compact emitter film was obtained (Figure 1A), which decreased the leakage current substantially. Consequently, EQE was improved by over 2 orders of magnitude in a green PeLED device.⁴⁴

In addition to the insulating PIP, an ionic conducting polymer poly(ethyleneoxide) (PEO) was also mixed with MAPbBr₃ perovskite to control the film morphology.⁴⁶ Similarly, Gao, Ma and coworkers incorporated the PEO polymer into CsPbBr₃ perovskite to optimize the film formation process.⁴⁵ PEO prevented perovskite from growing into large grain, generating a more uniform compact perovskite-PEO composite film. A local electrical current mapping revealed that the perovskite-PEO composite film had better electrical properties, as shown in Figure 1B. Besides, PEO passivated the perovskite surface traps and decreased trap-assisted nonradiative recombination. The photoluminescence quantum yield (PLQY) reached 60%. As a result, an EQE of 4.26% was achieved

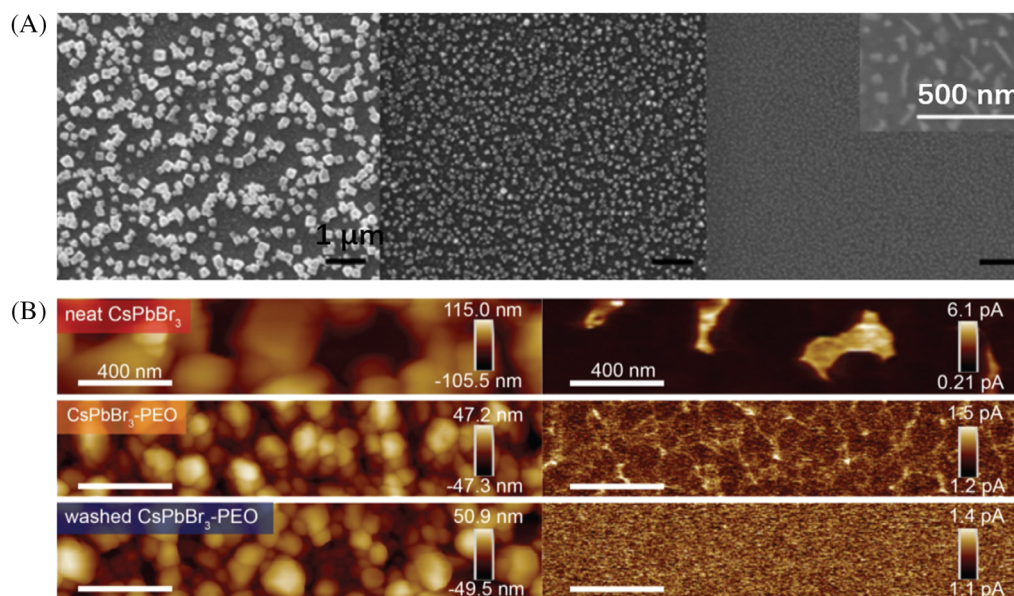


FIGURE 1 A, Scanning electron microscopy images of perovskite:PIP (w/w 1:0), perovskite:PIP (w/w 1:0.1), and perovskite:PIP (w/w 1:0.5) films, respectively. Reproduced with permission.⁴⁴ Copyright 2015, American Chemical Society. B, Local electrical current mapping of perovskite films. Reproduced with permission.⁴⁵ Copyright 2016, John Wiley and Sons. PIP, polyimide

in CsPbBr₃-PEO-based PeLEDs. Meanwhile, similar effects were demonstrated in FAPbBr₃-PEO-based PeLED devices.⁴⁷ Yu and coworkers further added poly(vinylpyrrolidone) (PVP) into CsPbBr₃-PEO.⁴⁸ Because PVP facilitated the dispersity of perovskites in PEO, a more uniform CsPbBr₃-PEO-PVP composite film was fabricated. The maximum luminance of PeLEDs reached ~600 000 cd m⁻² and an EQE of 5.7% was achieved in a green PeLED.

In addition to optimized film morphology, the reduced grain size after incorporating organic materials is another reason for EQE improvements.⁴⁹ 3D perovskites are excellent materials for photovoltaics, partly thanks to their small exciton binding energy, which is helpful for electron-hole dissociation. Unfortunately, the small exciton binding energy is unfavorable for PeLEDs, because the recombination of electrons and holes is of importance to achieve efficient PeLEDs. Reducing the grain size is a general strategy to enhance electron-hole radiative recombination. Perovskite films have a smaller grain size in the perovskite-polymer composite, which is favorable for the radiative recombination of injected charges. Yip and coworkers added a polymer poly(2-ethyl-2-oxazoline) (PEOXA) into the perovskite film to tune the grain size.⁵⁰ Because the polymer can control the crystal growth of the perovskite film, different grain sizes can be acquired by changing

the PEOXA ratio. The perovskite films with smaller size grains demonstrated enhanced bimolecular radiative recombination. On the other hand, the smaller grain size will increase the specific surface area, accompanied with a higher trap density. Meanwhile, a large number of charge carriers confined in perovskite films will enhance the possibility of Auger recombination. Hence, it is important to obtain a balance among bimolecular recombination, trap-assisted non-radiative recombination, and Auger recombination, when the grain size of perovskite is decreased. Secondly, trap-assisted non-radiative recombination and Auger recombination should be suppressed via passivation, balanced charge injection, and so on. PEOXA can coordinate with undercoordinated lead ions (Pb²⁺) in perovskite films due to the lone pair of electrons leading to passivation of surface traps. Passivation engineering in perovskites was demonstrated to be effective in reducing trap-assisted nonradiative recombination.⁵¹⁻⁵³ Finally, a MAPbI₃ PeLED with an EQE of 5.4% was achieved.

Wang, Huang and coworkers introduced an organic molecule 5-aminovaleric acid (5AVA) into perovskite and achieved a discrete submicrometer-structured perovskite possessing the light extraction effect, as shown in Figure 2.⁵⁴ Such a structure helps extract the light out of devices. Meanwhile, 5AVA molecules can

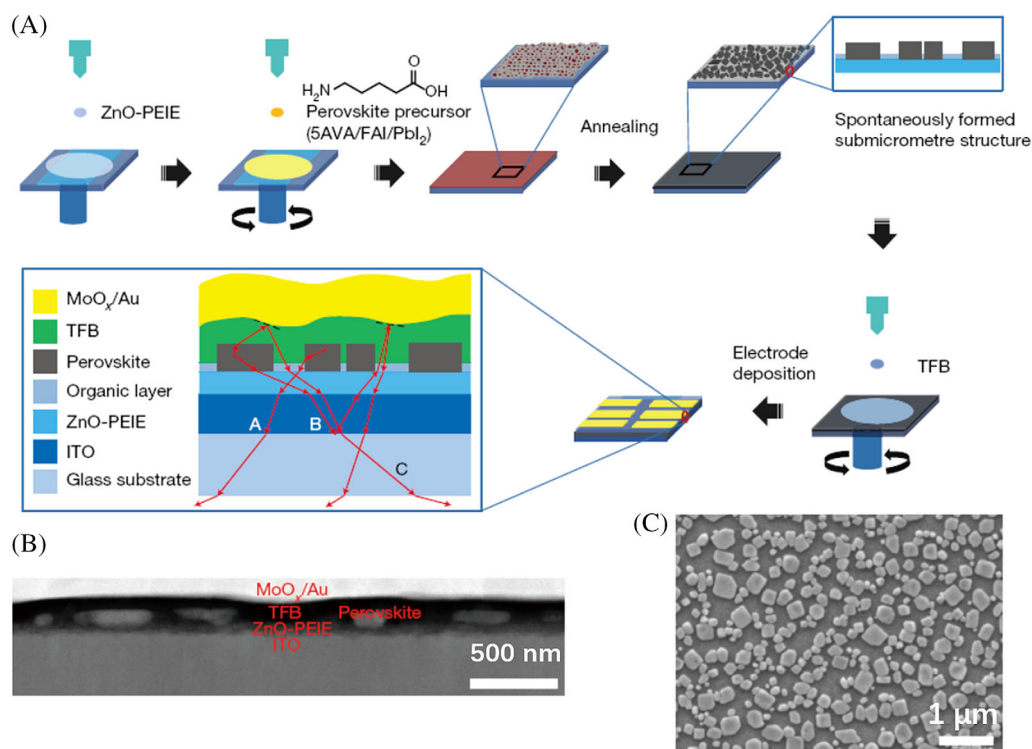


FIGURE 2 A, Schematic drawing showing the fabrication process of discrete submicrometer-structured perovskites. B, Cross-sectional scanning transmission electron microscopy image of a device. C, Top-view scanning electron microscopy images of the perovskite film. Reproduced with permission.⁵⁴ Copyright 2018, Springer Nature

passivate the surface defects of perovskite and improve the radiative recombination of charges. Importantly, 5AVA can have a dehydration reaction with the underlying electron transport layer, which forms an insulating layer between the perovskite grains. This insulating layer can block the direct contact between the hole transport layer and electron transport layer from the perovskite grain spaces and prevent the leakage current. As a result, an EQE over 20% was achieved in a near-infrared PeLED.

In the perovskite-organic structure-based PeLEDs, the properties of organic materials play a significant role. Recently, Gao, Huang, Liu and coworkers presented that the ational design of organic materials is of importance to achieve effective traps passivation.⁴³ Several amino-functionalized organic materials (Figure 3A) were incorporated into perovskites separately. These organic materials can all passivate traps and decrease trap-assisted non-radiative recombination. However, the efficiencies showed obvious differences, as shown in Figure 3B, which was due to the oxygen atoms that changed the interaction between the organic material and the perovskite material. The strong hydrogen bonds between the organic material and the perovskite material restricted the passivation effect. Certain positions of oxygen atoms in organic materials weakened the hydrogen bonding and the optimized configuration led to enhanced interaction (ie, passivation) between organic materials and traps in perovskites enhancing EQE (Figure 3B). Finally, as shown in Figure 3C, 2,2'-[oxybis

(ethylenoxy)]diethylamine (ODEA)-passivated FAPbI₃-based PeLED achieved an EQE of 21.6%.

The organic additive engineering demonstrated the effects in 3D PeLEDs. The poor film morphology and traps on perovskites caused a large leakage current and trap-assisted nonradiative recombination. Upon incorporation of organic additives, a uniform perovskite film and a low concentration of traps can be achieved.

2.2 | 2D PEROVSKITES

2D or quasi-2D perovskites that served as emitting layers have a general formula of A'₂A_{n-1}B_nX_{3n+1}, where A' is a series of long alkylammonium organic cations, such as benzylammonium (BA), phenylethylammonium (PEA), 1-naphthylmethylamine (NMA), and phenylbutylammonium (PBA), and <n> is the number of perovskite layers.⁵⁵⁻⁵⁷ Generally, several layers (<n> = 1, 2, 3, etc.) of BX₆ are separated and sandwiched by the long alkylammonium organic cation layers. These layered perovskites form quantum well structures that have a large binding energy owing to the quantum confinement effect. A larger binding energy is favorable for the formation and recombination of excitons in perovskite materials compared with that with a lower binding energy. The 2D or quasi-2D perovskites have a multiphase structure typically, where different layered quantum wells coexist and are stacked together

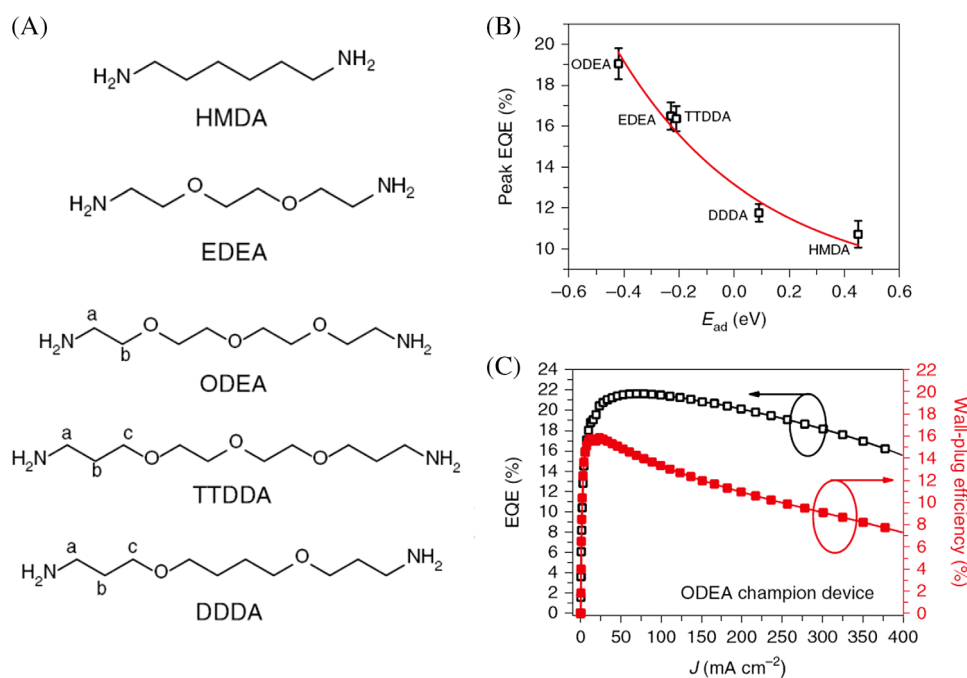


FIGURE 3 A, Chemical structures of passivating molecules: HMDA is hexamethylenediamine, EDEA is 2,2'-(ethylenedioxy)diethylamine, TTDDA is 4,7,10-trioxa-1,13-tridecanediamine, and DDDA is 4,9-dioxa-1,12-dodecanediamine. B, Average EQE values of devices incorporating different molecules. C, EQE of devices without or with ODEA. Reproduced with permission.⁴³ Copyright 2019, Springer Nature. EQE, external quantum efficiency

due to van der Waals forces. With the decrease in the number of perovskite layers $\langle n \rangle$, the corresponding bandgap increases, and a cascade energy level forms among different perovskite layers. The cascade energy provides a funneling channel for energy transfer from the perovskites with a wider bandgap to those with a narrower bandgap.^{35,36} Injected electrons and holes form excitons in the wider bandgap (small $\langle n \rangle$) perovskite layers and then transfer to the narrower bandgap (large $\langle n \rangle$) perovskite layers, followed by emission of photons.

Compared with that of 3D perovskites, 2D perovskites have a large excitonic binding energy and usually a higher PL efficiency, which is attractive for light-emitting diodes. However, there are several challenging issues for 2D perovskite materials, such as poor film morphology, inefficient charge transport, and undesirable trap-assisted nonradiative recombination. Incorporation of organic materials is an effective method to resolve these problems. Ma and coworkers chose BA as the alkylammonium organic material to mix with CsPbI₃ and fabricated a quasi-2D perovskite material as the emitting layer in PeLEDs. An EQE of 2.72% was achieved using the quasi-2D perovskite material. To improve the efficiency of PeLEDs, PEO was incorporated into the quasi-2D perovskite materials and a smooth perovskite-PEO film without large crystals was obtained.⁵⁸ The root mean square roughness of the perovskite-PEO film was reduced to 1.41 nm, which was much lower than that of the pristine perovskite film (2.69 nm). Meanwhile, PEO passivated the surface traps of perovskites, which reduced trap-assisted nonradiative recombination and improved the PL efficiency of perovskite films. Because of the film morphology improvement and the trap passivation effect of PEO, a red PeLED with an EQE of 6.23% was achieved.

Incorporating phenylethylammonium bromide (PEABr) into perovskites can decrease the grain size via tailoring the crystal growth and form an energy funneling quantum well perovskite structure. Consequently, the exciton radiative recombination events can be enhanced. However, this energy funnel is limited by the size distribution and phase separation between the perovskite and the organic phase. Sun, Sirringhaus, Deschler and coworkers added an organic molecule of 1,4,7,10,13,16-hexaoxacyclooctadecane (crown) into the 2D perovskite films to enhance the funnel process.⁵⁹ Because of the steric hindrance of crown, phase aggregation of PEABr was suppressed, which led to an efficient energy transfer funnel among perovskite layers. Trap-assisted nonradiative recombination was reduced, as shown in Figure 4A. The PLQY of perovskite film reached 70% (Figure 4B). An EQE of 15.5% was achieved in a green PeLED.

Tang, Lee, Gao and coworkers incorporated PEABr into CsPbBr₃ to fabricate 2D perovskite emitting layers. However, PeLEDs based on the 2D perovskites generated an EQE less than 3%. The reason of the low EQE was attributed to the poor film morphology and grain boundary traps causing a leakage current and trap-assisted nonradiative recombination. After incorporating a polymer polyethyleneglycol (PEG) into 2D perovskites, the efficiency of devices improved significantly. PEG as a Lewis base coordinated with the lead ions on perovskites, which passivated the traps at grain boundaries and decreased trap-assisted nonradiative recombination in perovskite emitting layers. Additionally, the perovskite-PEG composite led to a smooth and compact film, which reduced the leakage current. The suppressed trap-assisted nonradiative recombination and the reduced leakage current improved the radiative recombination probability of injected charges. The maximum luminance of devices improved from

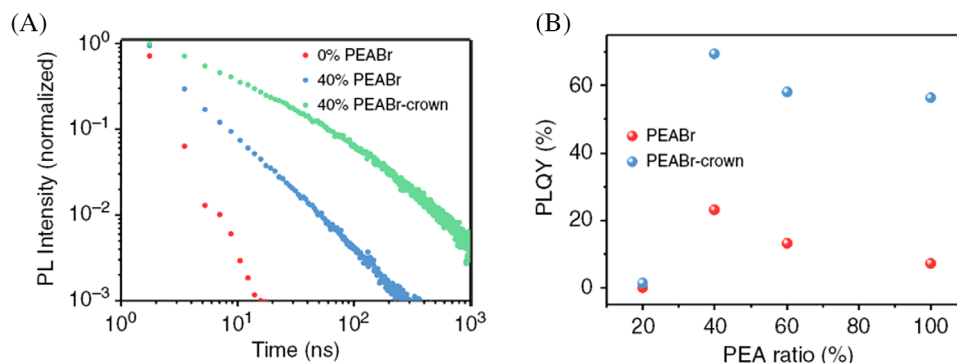


FIGURE 4 A, Time dependence of the PL intensity of perovskite films, where the red curve corresponds to the CsPbBr₃ film; the blue curve corresponds to the PEABr-CsPbBr₃ film; the green curve corresponds to the PEABr-CsPbBr₃ film mixed with crown. B, PLQY of the PEABr-incorporated perovskite films with or without crown. Reproduced with permission.⁵⁹ Copyright 2018, Springer Nature. PEABr, phenylethylammonium bromide; PL, photoluminescence; PLQY, photoluminescence quantum yield

11,430 to 45 990 cd m^{-2} after mixing PEG. An EQE over 13% was achieved in a green PeLED.⁶⁰

Inspiringly, Di and coworkers achieved a near-infrared PeLED with an EQE up to 20.1% by using a quasi-2D perovskite-polymer heterostructure as the emitting layer.⁶¹ The quasi-2D perovskite was prepared by dissolving NMAI, FAI, and PbI_2 in the *N,N*-dimethylformamide solvent. The polymer poly(2-hydroxyethyl methacrylate) (poly-HEMA) was introduced into the perovskite materials to form a perovskite-polymer heterostructure. Injected charges form excitons in lower $\langle n \rangle$ perovskites for the first time, followed by transfer to the larger $\langle n \rangle$ perovskites within ~ 1 ps, as shown in Figure 5A,B. The faster transfer process eliminated almost all trap-assisted non-radiative recombination, and an external PL quantum efficiency of nearly 100% was achieved (Figure 5C).

The long alkylammonium organic material is helpful for enhancing radiative charge recombination. On the other hand, the insulating property of long alkylammonium organic materials can block charge injection and transport within the perovskite layer, causing a larger turn-on voltage and a lower efficiency. A balanced ratio between the long alkylammonium organic material and charge transport is of importance for the fabrication of efficient PeLEDs. On the other hand, the remaining long alkylammonium organic material is still a disadvantage for PeLEDs. Removing the excessive long alkylammonium organic material after the perovskite film deposition was an alternative scheme to improve the transport property. Solvent washing is a feasible method to remove the excessive long alkylammonium organic material. However, the commonly used solvent such as isopropanol also dissolves the 2D perovskite materials. Tan et al demonstrated that the polymer PEO can act as a protection layer for the 2D perovskite (BABr-CspbBr_3) material during the washing process.⁶² As illustrated in Figure 6A, incorporation of PEO protected the 2D

perovskite material from solvent damaging during the washing process. Without incorporating PEO, the 2D perovskite material changed to 3D and the PL intensity decreased by 82% after the solution washing treatment (Figure 6B). On the contrary, after incorporating PEO, only the parts with low $\langle n \rangle$ perovskites were removed and the 2D perovskite-PEO composite film still retained its 2D property after the washing posttreatment (Figure 6C). In addition, charge transport properties were improved. Meanwhile, the ionization energy decreased from 5.86 to 5.65 eV, as shown in Figure 6D, indicating a reduced energy barrier for hole injection. Consequently, the turn-on voltage decreased from 3.4 V to 2.9 V. The maximum luminance of PeLEDs improved from 191 to 33 532 cd m^{-2} . A green PeLED with an EQE over 8% was achieved.

Wang and coworkers chose 2D perovskites of PEABr-CspbBr_3 mixed with PEO as the emitting layer and achieved an EQE of 12% in green PeLEDs. Furthermore, NaBr was incorporated into the perovskite layers to improve the performance of PeLEDs.⁶³ Because of the smaller size, Na ions possessed a smaller steric hindrance in the formation of perovskite materials. Hence, Na ions can be incorporated into perovskite materials partially substituting PEA. The substitution of PEA changed the transport resistance of charges, leading to more balanced charge transport between electrons and holes. Meanwhile, Na ions filled the vacancies in perovskites, which passivated the traps and decreased trap-assisted nonradiative recombination. The introduction of NaBr reduced the perovskite grain size, leading to stronger quantum confinement and improved radiative recombination of excitons. As a result, the EQE was improved to 17.4% with a corresponding current efficiency of 54.6 cd A^{-1} .

The long alkylammonium organic material in perovskites can assist in the formation of the 2D or quasi-2D quantum well structure, improving radiative recombination of charges. In addition, the long alkylammonium organic material can lead to better perovskite films and passivate surface traps. On the other hand, the resistance of long

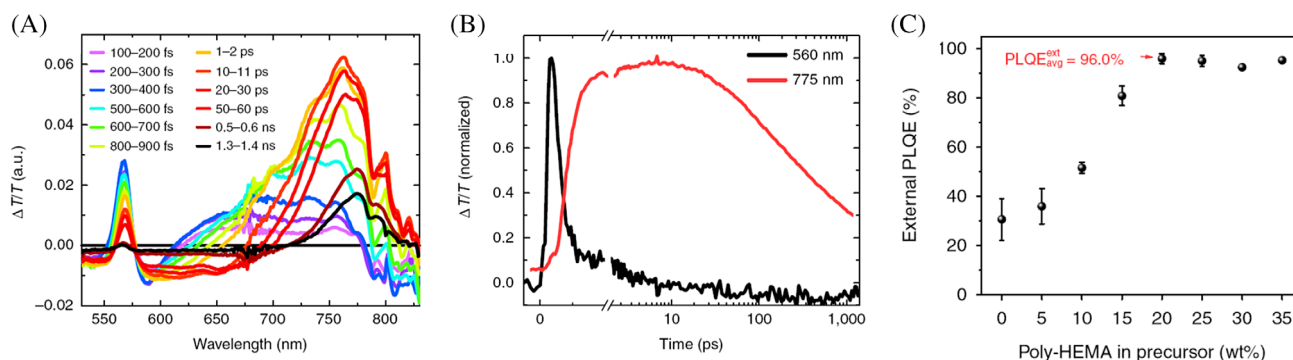


FIGURE 5 A, Transient absorption spectra. B, Transient absorption kinetics. C, External PL quantum efficiency of perovskite films with different ratios of a polymer. Reproduced with permission.⁶¹ Copyright 2018, Springer Nature. PL, photoluminescence

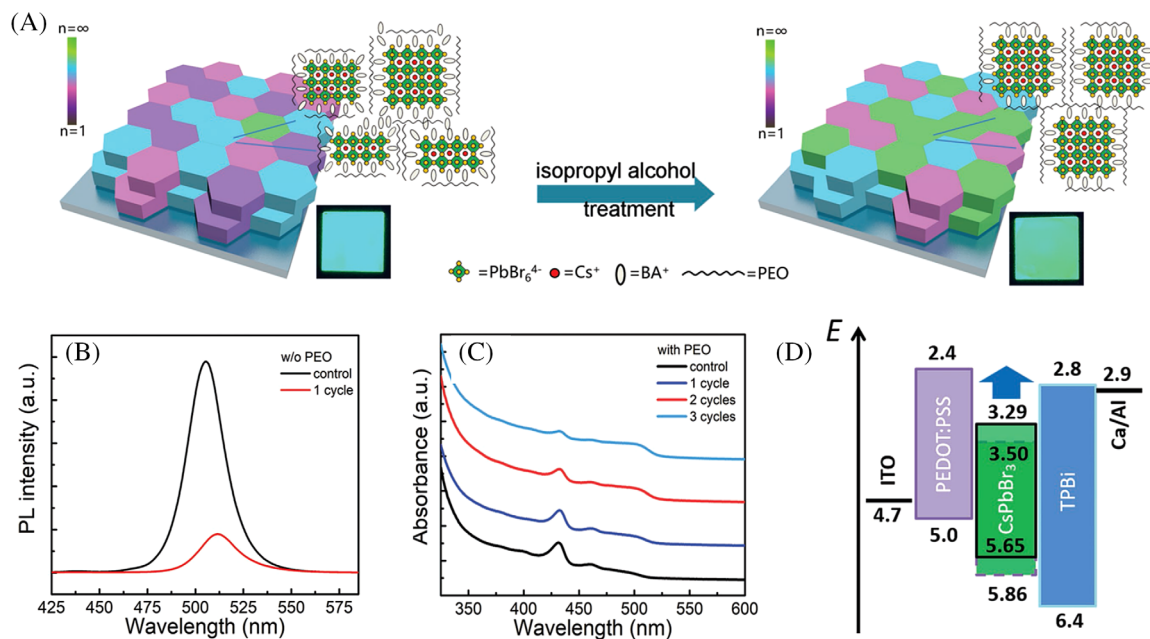


FIGURE 6 A, Schematic drawing showing the perovskite-PEO composite films post-treated by washing with the solvent of isopropyl alcohol. B, PL spectra of perovskite films before and after the solvent post-treatment. C, Absorption spectra of the perovskite-PEO composite films before and after the solvent post-treatment. D, Energy-level alignment of PeLEDs. Reproduced with permission.⁶² Copyright 2018, John Wiley and Sons. PeLEDs, perovskite light-emitting diodes; PEO, poly(ethyleneoxide); PL, photoluminescence

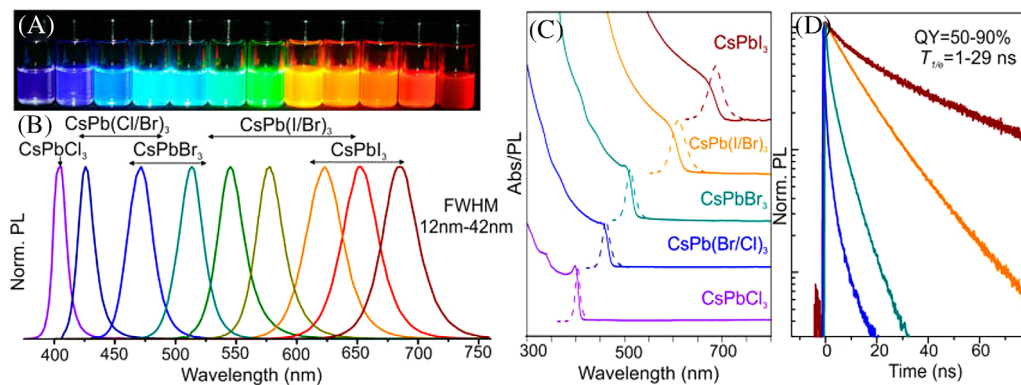


FIGURE 7 A, Photograph of colloidal CsPbX_3 NCs ($X = \text{Cl}, \text{Br}, \text{I}$) perovskite NCs in toluene under UV illumination. B, PL spectra exhibiting composition-tunable bandgap energies covering the entire visible spectral region with narrow and bright emission. C, Absorption and PL spectra corresponding to optical absorption and emission spectra of colloidal CsPbX_3 NCs. D, Time-resolved PL decays of colloidal CsPbX_3 perovskite NCs. Reproduced with permission.⁶⁴ Copyright 2015, American Chemical Society. NCs, nanocrystals; PL, photoluminescence

alkylammonium organic material decreases charge or energy transfer between perovskite layers, which is unfavorable for device performances. Organic additives can be incorporated into 2D or quasi-2D perovskites to control the resistance caused by long alkylammonium organic materials.

2.3 | PEROVSKITE QDs

Perovskite QDs or NCs have demonstrated a potential for efficient PeLEDs, because of their high PLQY, narrow emission width, tunable spectral region, and wide color gamut

covering, as shown in Figure 7.⁶⁴⁻⁶⁶ Since the QD PeLED with an EQE of ~1% was reported by Zeng and coworkers,³⁹ several research works have been reported to improve the performance, and an EQE of 21.3% was achieved in a CsPb(Br/I)_3 QD-based device.⁶⁷

Surface ligands are critical to the properties of perovskite QDs. Ligands can improve the dispersity of QDs in solvent. However, the insulating property of ligands limits charge transport and causes poor conductivity in QD films, which is one of the reasons why perovskite QDs with an excellent PLQY cannot achieve a high device efficiency.⁶⁸⁻⁷⁰ Removal of ligands decreases the dispersity of QDs and consequently

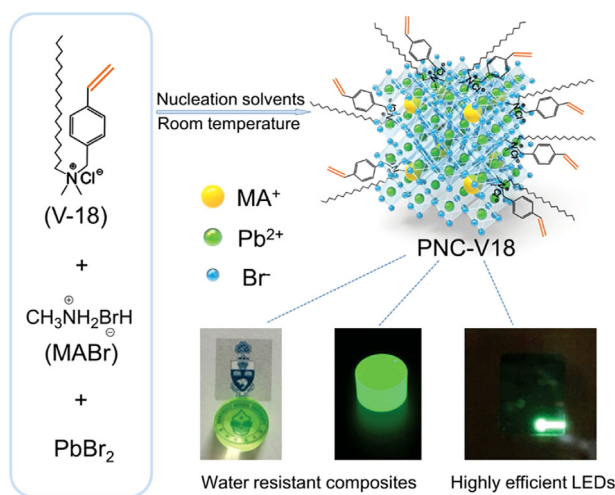


FIGURE 8 Schematic of perovskite NCs treated by ligand V18 and photographs of water-resistant perovskite composites and PeLEDs. Reproduced with permission.⁷² Copyright 2017, John Wiley and Sons. NCs, nanocrystals; PeLEDs, perovskite light-emitting diodes; V18, 4-vinylbenzyl-dimethyloctadecylammonium chloride

leads to a poor film morphology. Rogach, Choy, and coworkers introduced an organic material polyhedral oligomeric silsesquioxane (POSS) into the CsPbBr₃ NC solution to resolve the dispersity problem.⁷¹ POSS is favorable to increase perovskite dispersity. Compared with the pristine perovskite film, a much denser film was obtained after adding POSS. Meanwhile, POSS blocks the holes transporting to the electron transport layer, which improved charge radiative recombination in perovskite emitting layers. CsPbBr₃-NC-based PeLEDs showed an EQE of 0.35%.

Stability of perovskite QD films is still an obstacle for the application of devices. Seferos, Sargent, and coworkers applied a crosslinking material 4-vinylbenzyl-dimethyloctadecylammonium chloride (V18) as a ligand to improve the stability of perovskite NCs, as shown in Figure 8.⁷² V18 molecules passivated surface traps and improved the efficiency of devices. In addition, the styryl group of V18 ensured a crosslinking process that connected the perovskite NCs. The crosslinked perovskite composite exhibited excellent water resistance properties. Huang, Chen, and coworkers chose PEO as a capping agent and alleviated the instability problem of MAPbBr₃ perovskite films.⁷³ After the surface of QDs was coated by PEO, the stability of QDs was improved, and the QDs solution retained its original color for several days. In addition to the environmental stability, PEO improved the electrical properties of QD films through passivating the quenching sites on QDs. When the perovskite-PEO composite film was used as an emitting layer, an EQE of 2.38% was achieved. The relevant parameters of PeLEDs are summarized in Table 1.

The properties of additives play an important role in improving efficiency. These properties include (a)

morphology improvements, (b) grain size control, (c) passivation of defects, (d) solvent resistance, (e) barrier reduction, (f) suppression of phase segregation, and (g) capping agent. A summary of these functional molecules related to the above properties can be found in Table 1. A strategy about how to rationally choose organic additives warrants more research efforts. According to Table 1, film morphology control and passivation of electronic traps are the main factors that can lead to efficiency improvements. Perovskite grain growth is influenced by organic additives owing to the interaction between perovskite precursors and organic additives.⁶⁰ In PeLEDs, perovskite thin films with smaller grains are preferred for attaining high efficiencies. Organic additives can induce the formation of smaller perovskite grains with uniform and pinhole free perovskite-organic composite films.^{45,46} Additionally, the passivation agent, such as oxygen and/or nitrogen-containing groups, could form chemical bonds or other interactions (eg, van der Waals force) with traps in perovskites, such as undercoordinated lead ions (Pb²⁺).⁵⁰ Upon formation of bonds between organic additives and traps (ie, passivation), trap-assisted nonradiative recombination is suppressed and consequently the efficiency of PeLEDs is improved.

The efficiency of QD PeLEDs with organic additives still lags behind compared with that of 3D- and 2D-based PeLEDs. However, organic materials are beneficial for the stability of perovskite QDs or NCs. Mixing organic materials and perovskite QDs or NCs is a feasible method to achieve display applications, which is discussed in the following section.

3 | PEROVSKITE LIGHT EMISSION FILMS

In addition to working as emitting layers in PeLEDs, the perovskite materials can serve as a light conversion layer for displays. And the perovskite-organic composite presents an alternative scheme for this purpose. For example, Snaith and coworkers embedded perovskite crystals into a transparent polymer of polystyrene (PS) matrix and achieved a wider color emission, as shown in Figure 9A.⁷⁸ PS matrix stabilized the perovskite structure and held its emission spectra within 400 minutes under continuous illuminance. This proved that perovskite-PS composite is promising for application in display. Alivisatos and coworkers chose poly(styrene-ethylene-butylene-styrene) (SEBS) as an encapsulation material to protect the perovskite QDs from water and oxygen,⁸⁰ because the alkyl chain on SEBS was similar to the alkyl chain ligands on the QD, which optimized the interface between QDs and SEBS and generated a better blend effect. With the protection of SEBS, perovskite films that immersed in water demonstrated stable emission spectra after 60 days.

TABLE 1 Summary of the reported PeLED devices using a composite of perovskite materials and organic additives as emitting layers

Year	Emitting layers		Device structure ^a	Additive functions	Champion EQE (%)	EL ^b (nm)	Ref.
	Perovskite materials	Organic additives					
2015	MAPbBr ₃	PIP	ITO/PEDOT:PSS/emitter/F8/Ca/Ag	Morphology	1.2	534	44
2016	CsPbBr ₃	PEO	ITO/PEDOT:PSS/emitter/TPBi/LiF/Al	Morphology/passivation	4.26	521	45
2016	CsPbBr ₃	PEO/PVP	ITO/emitter/In-Ga	Morphology	5.7	522	48
2016	CsPbBr ₃ QD	POSS	ITO/PEDOT:PSS/PVK/emitter/TPBi/LiF/Ag	Morphology	0.35	517	71
2017	MAPbBr ₃	PEO	ITO/PEDOT:PSS/emitter/TmPyPB/CsF/Al	Morphology	4.0	520	74
2017	CsPbBr ₃	PEO	ITO/PEDOT:PSS/emitter/TPBi/LiF/Al	Grain sizes	4.76	525	75
2018	MAPbI ₃	PEOXA	ITO/poly-TPD/emitter/TPBi/LiF/Al	Passivation/grain sizes	5.4	760	50
2018	BAI:CsPbI ₃	PEO	ITO/PEDOT:PSS/poly-TPD/emitter/TPBi/LiF/Al	Morphology/passivation	6.23	680	58
2018	BABr:CsPbBr ₃	PEO	ITO/PEDOT:PSS/emitter/TPBi/LiF/Al	Solvent resistance/ Barrier reduction	8.42	514	62
2018	PEABr:CsPbBr ₃	PEG	ITO/PEDOT:PSS/emitter/TPBi/LiF/Al	Morphology/passivation	13.14	514	60
2018	PEABr:CsPbBr ₃	Crown	ITO/poly-TPD/emitter/TPBi/LiF/Al	Phase aggregation suppression	15.5	514	59
2018	PEABr:CsPbBr ₃	Crown	ITO/PVK/emitter/TPBi/LiF/Al	Phase aggregation suppression	13.9	512	76
2018	PEABr:CsPbBr ₃	Crown	ITO/poly-TPD/PFN/emitter/TPBi/LiF/Al	Phase aggregation suppression	14.4	512	77
2018	NMAI:FAPbI ₃	Poly-HEMA	ITO/MZO:PEIE/emitter/TFB:PFO/MoO ₃ /Au	Heterostructure	20.1	800	61
2018	FAPbI ₃	5AVA	ITO/ZnO:PEIE/emitter/TFB/MoO ₃ /Au	Morphology/passivation	20.7	803	54
2019	MAPbBr ₃ QD	PEO	ITO/PEDOT:PSS/PVK/emitter/TPBi/LiF/Al	Capping agent/passivation	2.38	511	73
2019	PEABr:CsPbBr ₃	PEO	ITO/NiO/PVK/emitter/TPBi/LiF/Al	Morphology/passivation	17.4	512	63
2019	FAPbI ₃	ODEA	ITO/ZnO:PEIE/emitter/TFB/MoO ₃ /Au	Passivation	21.6	800	43

Abbreviations: 5AVA, 5-aminovaleric acid; EQE, external quantum efficiency; ODEA, 2,2'-[oxybis(ethylenoxy)]diethylamine; PeLEDs, perovskite light-emitting diodes; PEG, polyethyleneglycol; PEO, poly(ethyleneoxide); PEOXA, poly(2-ethyl-2-oxazoline); PIP, polyimide; PVP, poly(vinylpyrrolidone); poly-HEMA, poly(2-hydroxyethyl methacrylate); POSS, polyhedral oligomeric silsesquioxane; PS, polystyrene.

^aEmitter is a composite of perovskite materials and organic additives.

^bEL is the electroluminescence peak position of devices.

Zhang and coworkers blended CsPbBr₃ crystals with poly(methyl methacrylate) (PMMA), and achieved a stable composite film that retained the quantum yield over a month.⁷⁹ Even immersed in water for 1 month, the composite film still demonstrated stable PL spectra. In particular, after coating a red emission phosphor layer and a green emission perovskite composite layer onto a blue LED (Figure 9B), a white color light emission was achieved (Figure 9C). Xie,

Wang, and coworkers also mixed CsPbBr₃ crystals with PMMA by a different method and demonstrated similar results.⁸¹ Yang and coworkers incorporated poly(maleic anhydride-alt-1-octadecene) into CsPbBr₃ perovskite films as a protection agent. The perovskite composite film nearly maintained its PL intensity after 40 days in the air.⁸² These research works presented a promising application of perovskite-organic film in display field.

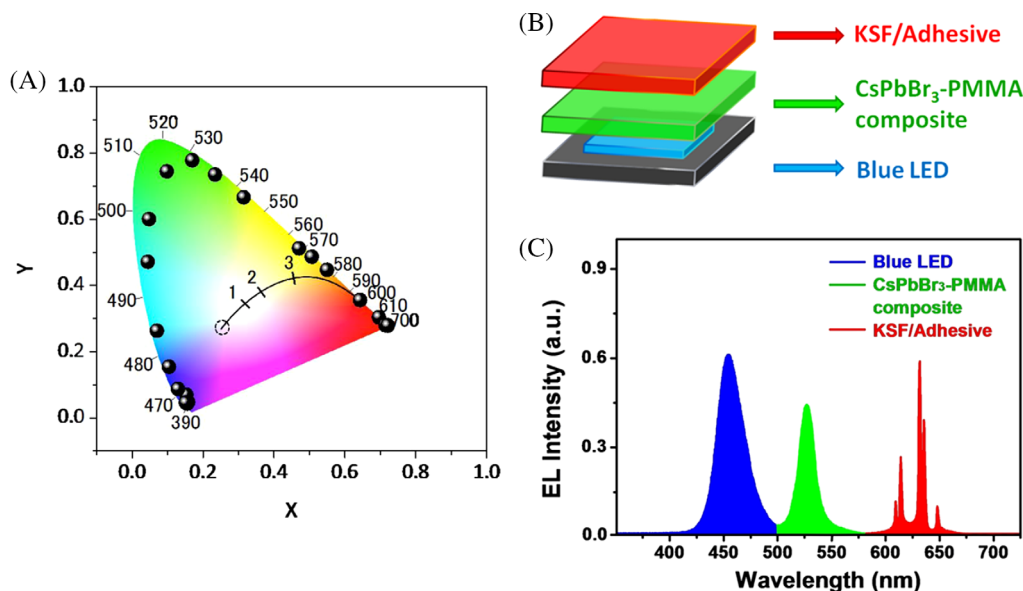


FIGURE 9 A, Commission Internationale de l'Eclairage color coordinates of perovskite composite films. Reproduced with permission.⁷⁸ Copyright 2015, American Chemical Society. B, Prototype of white LED. C, Emission spectra of white LED. Reproduced with permission.⁷⁹ Copyright 2018, American Chemical Society. LED, light-emitting diode

4 | CHALLENGES AND FUTURE DIRECTIONS

4.1 | WORKING STABILITY

The ambient air exposure (such as water and oxygen) related degradation can be resolved by encapsulation technologies.^{83,84} However, the working stability of PeLEDs is still an issue. The possible reasons for perovskite degradation are (a) phase segregation, (b) reaction with electrodes, and (c) even decomposition under high voltages. Injected charges can undergo recombination via several pathways, such as radiative recombination, trap-assisted nonradiative recombination, and Auger recombination.⁸⁵⁻⁸⁷ Both trap-assisted nonradiative recombination and Auger recombination generate Joule heat that can affect the stability of perovskite layers. Hence, to improve operational stability of PeLEDs, trap-assisted nonradiative recombination and Auger recombination have to be suppressed. Trap passivation is an effective method to reduce trap-assisted nonradiative recombination. Achieving balanced electron and hole injection and transport is a feasible method to decrease Auger recombination.

Dion-Jacobson (DJ) structural perovskite materials demonstrate promising properties in perovskite devices.^{88,89} The DJ-type perovskite materials possess a larger decomposition energy compared with that of Ruddlesden-Popper ones. PeLEDs have achieved a half-lifetime over 100 hours with DJ-type perovskite materials as emitting layers.⁹⁰ Hence, DJ-type perovskite materials is a promising alternative solution to achieving stable PeLEDs.

The EQE roll-off is another issue for PeLEDs. Wang, Huang, and coworkers reported that the roll-off is mainly caused by Auger recombination.⁹¹ Due to the quantum well structure of perovskite, a large number of charges are confined in the quantum potential well, which causes severe Auger recombination under high charge injection. After increasing the width of quantum wells, the distributed concentration of charges can be decreased, and consequently the Auger recombination can be reduced. On the other hand, Giebink and coworkers presented that the reason of EQE roll-off was the Joule heating that increased exciton dissociation.⁹² Heremans, Gulbinas, Fakharrudin, and coworkers showed that balanced electron and hole injection reduced the roll-off issue.⁹³ The exact mechanism responsible for the EQE roll-off is still under debate and needs more research works.

4.2 | BLUE PEROVSKITE LIGHT-EMITTING DIODES

The EQE of green, red, and near-infrared PeLEDs have reached over 20%.^{43,54,61,67,94} However, the champion EQE of blue PeLEDs was 9.5%.⁹⁵ One method for achieving blue-color emission perovskites is based on Cl-contained perovskites, such as CsPb(Br/Cl)₃ perovskite. However, according to the tolerance factor of achieving a stable crystal, the size of Cl ions is too small to keep a stable crystal. Hence, the Cl-based perovskites are unstable for blue PeLEDs. As the applied voltage increases, the electroluminescence peak shifts from blue to green color, because the perovskite

emission layer suffers from the halide segregation phenomena leading to nonuniform Cl-rich and Br-rich domains. Red, green, and blue are the basic colors for displays. Hence, the halide segregation problem causing undesirable efficiency in blue PeLEDs needs be resolved. 2D perovskite materials possessing a quantum confinement effect can also achieve blue emission and serve as emitting layers in PeLEDs. However, the adverse factors associated with 2D perovskite, such as traps, various layered distributions, and excessive long alkylammonium organic material, still restrict blue PeLED efficiency. Organic additives have resolved these factors in green, red, and near-infrared PeLEDs effectively, through trap passivation, film morphology control, 2D phase protection, and phase aggregation suppression. 2D perovskites are likely an alternative solution to resolve the problems in blue PeLEDs.

Manganese doping is proven to be effective in improving the spectral stability and lifetime in CsPb(Br/Cl)₃-based blue PeLEDs.⁹⁶ Manganese doping can reduce trap density in perovskite materials, which is favorable for suppressing ion migration and then stabilizing the pristine phase.⁹⁷ Combined advantages of organic additives and metal ions doping simultaneously will be a promising scheme for attaining efficient and stable blue PeLEDs. On the other hand, in general, the number of the studies on PeLEDs is still relatively small and this topic has not been fully investigated in detail, and a clear understanding about the influence of organic additives on ion migration is still lacking at this stage.

5 | CONCLUSION AND OUTLOOK

We review the recent progress of PeLEDs with organic materials as additives and discuss the function of organic additives to the performances of PeLEDs. According to the current challenges facing PeLED applications, different organic additives can be applied to resolve the problem. Furthermore, we discuss the remaining issues, including working lifetime and blue PeLEDs. Organic additives can passivate surface trap, tune the film morphology, enhance charge or energy transfer, and improve stability. Importantly, various organic materials present an abundant choice for resolving the problem and achieving efficient PeLEDs and beyond PeLEDs, including photovoltaics, detectors, lasers, and memristors.

On the other hand, the fundamental principle of choosing the right kind of organic molecules to serve a particular purpose has not been fully understood in PeLEDs. Previous works have preliminarily demonstrated that the molecular structure is of importance for attaining high device performances. However, because of the limited number of studies especially theoretical ones, it is challenging to readily choose

the right effective organic additives that can improve the device performances from the enormous variety of organic materials. Hence, more research including theoretical simulations is required to better understand the intrinsic mechanism of the interaction between perovskite materials and organic additives.

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CONFLICT OF INTEREST

The authors declare no conflict of interest.

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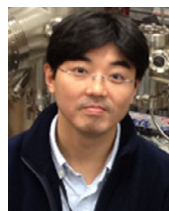
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