REVIEWS

A recent advances of blue perovskite light emitting diodes for next generation displays

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Abstract: The halide perovskite blue light emitting diodes (PeLEDs) attracted many researchers because of its fascinating opto-electrical properties. This review introduces the recent progress of blue PeLEDs which focuses on emissive layers and interlayers. The emissive layer covers three types of perovskite structures: perovskite nanocrystals (PeNCs), 2-dimensional (2D) and quasi-2D perovskites, and bulk (3D) perovskites. We will discuss about the remaining challenges of blue PeLEDs, such as limited performances, device instability issues, which should be solved for blue PeLEDs to realize next generation displays.

Key words: halide perovskite; light emitting diodes; blue emission

1. Introduction

The halide perovskite is a promising material with ABX_3 formation and perovskite structure. A monovalent cation is used in the A-site; methyl ammonium (MA), formamidinium (FA), Rb, and Cs. For the B-site, divalent metal, typically Pb, is used. For X site, halides are used; I, Br, and Cl. These halide perovskites attracted many researchers with fascinating properties, such as high charge carrier mobility[1], high photoluminescence quantum yield (PLQY)[2], high absorption coefficient[3], easily tunable optoelectronic bandgap[4], narrow emission spectra[5], wide color gamut[6], defect tolerance[7]. Moreover, with light weight, cheap material price, and solution processability, the halide perovskites are considered as economical materials for various optoelectronic devices such as solar cells[8, 9], photodetectors[10], and light emitting diodes (LEDs)[11].

The perovskite LEDs (PeLEDs) have been expected to be the leading candidate for the next generation displays due to narrower emission spectrum than other materials, and the PeLEDs have been rapidly developed in past few years with intensive research. The external quantum efficiency (EQE) of the PeLEDs have been enhanced from below 1% to over 20% for green and red colors[12–14], approaching the theoretical limits[15]. However, the blue PeLEDs are showing relatively slow development.

This review introduces the recent progress of the blue PeLEDs first, which focuses on the development of emissive layers. The emissive layer is categorized into three groups, perovskite nanocrystals (PeNCs), 2-dimensional (2D) and quasi-2D perovskites, and bulk (3D) perovskites. Subsequently, the developments of interlayers and interfacial engineering methods will be discussed secondly. Next, we discuss the remaining huddles in blue PeLEDs such as performance limitations,

device instability issues, and presents perspective of the blue PeLEDs.

2. Developments in emissive layer

2.1. PeLEDs with perovskite nanocrystals (PeNCs)

2.1.1. Synthesis methods of blue emissive PeNCs

The halide perovskite nanocrystals (PeNCs) are nanometer-sized perovskite crystals, wrapped with organic ligands. By the organic ligands, the PeNCs are dispersed in organic solvents. The presence of organic ligands increases the exciton binding energy of the PeNCs, because electrically insulating ligands confine the charge transport behavior. Increased exciton binding energy enhance radiative recombination rates, and this is the main reason of most of PeNCs shows higher PLQY results than other types of perovskites. Unlike two other types of perovskites, the 2D perovskites and bulk (3D) perovskites, the PeNCs must be synthesized before fabrication of emissive layers. The first synthesis of PeNCs was reported by Schmidt *et al.*[5] with $MAPbBr_3$ perovskite with green color emission, then, various research on synthesis, post-treatments, and device applications of PeNCs were published. We will briefly introduce the synthesis methods of blue color emissive PeNCs.

Hot injection method

The general synthesis method of the PeNCs is based on the hot-injection method, which is well known and widely using method in the quantum dot fields[16]. The hot-injection method has the advantage of yielding nanoparticles with uniform size, and controlling the size and shape of the nanoparticles easily. Furthermore, methods of controlling properties of the PeNCs were quickly developed by applying the knowhow of previous hot-injection methods for quantum dots.

A common synthesis process of blue emissive PeNCs is injecting Cs precursors into the $PbBr_2$, $PbCl_2$, and organic ligand mixed precursors at high injection temperature[6]
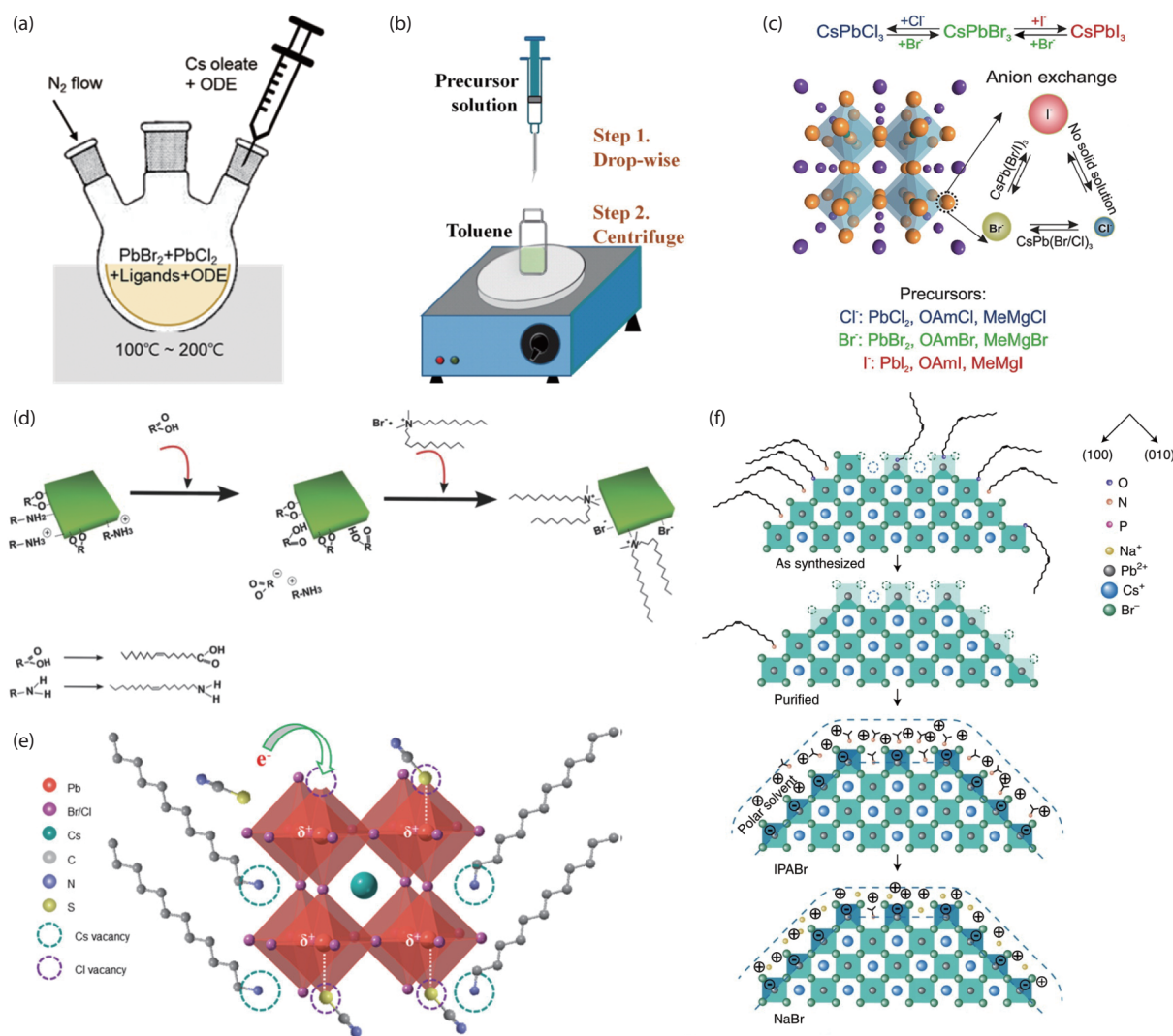
Fig. 1. (Color online) Synthesis and engineering methods of PeNCs to improve the performance of the blue PeLEDs. Schematic diagrams of synthesis method of the blue emissive PeNCs through (a) hot injection method, (b) LARP method[20], and (c) halide exchange method[23]. Copyright © 2015, American Chemical Society. Schematic diagrams of strategy to improve the blue PeLEDs through (d) ligand exchange method[28], (e) halide defect passivation method[35], and (f) bipolar shell strategy on PeNCs[18]. Copyright © 2016, John Wiley and Sons, Copyright © 2020, American Chemical Society. Copyright © 2020, Springer Nature.

(Fig. 1(a)). Synthesized PeNCs are purified by centrifugation and finally dispersed in nonpolar organic solvents before use. Successful synthesis of blue PeNCs was achieved by Protesescu *et al.* with $CsPb(Br/Cl)_3$ perovskites with high PLQY (~90%)[6]. Interestingly, in the hot-injection method, various parts in the synthesis process affect the property of the blue PeNCs[17]. The shape of the PeNCs could be controlled by selection of organic ligands and the concentration of ligands affects the PeNCs size. The injection temperature also determines the average size of the PeNCs. Furthermore, the emission spectra could be adjusted by reducing the size of the nanocrystals, by the quantum confinement effect[6, 18, 19].

Ligand-assisted reprecipitation (LARP) method

The LARP method is simpler than the hot-injection method for obtaining the PeNCs. When perovskite precursors and organic ligand mixed solution is added into nonpolar solvent, nanometer-size perovskite crystals are immediately synthesized[20] (Fig. 1(b)). Like hot-injection method, blue PeNCs were obtained using mixed halide precursors, and with further assistance of quantum confinement effect[21]. Since the

LARP method does not require heating instruments and inert gas supply, the LARP method has great advantages in synthesis process steps and could easily modify the scale of the synthesis.

Halide exchange post-treatment method

After the PeNCs synthesis, the halide ratio in the PeNCs could be tuned more to obtain blue emissive PeNCs[22–24]. The halide exchange post-treatment methods have been considered as an efficient way to obtain blue emissive PeNCs without use of insoluble $PbCl_2$ precursors (Fig. 1(c)). Usual halide exchange process is mixing extra halide precursors with the PeNCs. Then the halides from the supplied precursors are spontaneously exchanged with the halides at the surface of the PeNCs. Because the halides in the perovskite crystals are mobile, the exchanged halides on the PeNCs surface are spontaneously infiltrated into the inside of the PeNCs, forming well blended (Br and Cl) halide PeNCs. Furthermore, the halide exchange method provides enough of halides to the PeNCs, the surface halide defects of nanocrystals are naturally passivated, driven to improved PLQY.

2.1.2. PeLEDs with PeNCs

The first blue PeLEDs based on the PeNCs were produced by Song *et al.* with CsPb(Br/Cl)$_3$ PeNCs[25]. The PeNCs were synthesized through the hot-injection method, and the PeLEDs showed EQE of 0.07% and maximum luminance of 742 cd/m^2. After that, various methods have been proposed to improve the performance of the blue PeLEDs with PeNCs. The common strategy for improving the blue PeLEDs was the post-treatment method to improve the characteristics of the PeNCs[26, 27]. Pan *et al.* introduced ligand exchange technique to achieve better charge injection[28] (Fig. 1(d)). They used didodecyldimethylammonium chloride (DDAC) ligands to control the ligand density of PeNCs as well as to supply the Cl anions to the PeNCs. They achieved performance of sky-blue PeLEDs, EQE of 1.9% and maximum luminance of 35 cd/m^2 at 490 nm emission peak. Likewise, Shin *et al.* used mixture ligands with DDAC and didodecyldimethylammonium bromide (DDAB) to obtain pure blue PeLEDs, showing EQE of 0.44%[29] at 470 nm emission peak. Ochsenbein *et al.* reported that zwitterionic sulfobetaine exchanging ligands could form more stable bond with PeNCs and provide better long-term stability, and enhance the PLQY[30]. They also fabricated blue PeLEDs with performance of over 1% of EQE at 461 nm emission peak.

The basic purpose of these ligand exchanging techniques is removing original ligands such as oleic acids or oleylamines. These common ligands are easy to handle and provide good colloidal stability with long alkyl chain, however, become a barrier to charge carrier injection. Therefore, exchanging ligands should have shorter alkyl chain to improve the charge carrier injection. In a similar vein, the ligand density in PeNCs could also be a considerable factor in the performance of blue PeLEDs[31]. High ligand density certainly confines electronic band structure of PeNCs and results high PLQY, but too high ligand density lowers the performance of blue PeLEDs from inhibited charge carrier injection. On the other hand, too low ligand density lowers colloidal stability of PeNCs and leads severe aggregation that disturbs solution process. Moreover, too low ligand density in PeNCs leads creation of defects on the surface of PeNCs and lowers the radiative recombination. Therefore, to achieve high performance blue PeLEDs, the ligand density should be controlled to appropriate level, which depends on what exchanging ligands are used. Furthermore, the ligand density of PeNCs is unpredictably decreased in purification conditions, so, ligand exchanging techniques require delicate experimental steps to obtain high quality PeNCs with optimized ligand density condition.

Another main strategy to increase the luminescence efficiency of PeNCs is the halide defect passivation. The halide defects are a major factor that creates trap-states in electronic band of the PeNCs and lowers radiative recombination in the PeNCs. Therefore, removing the halide defects is an efficient way to improve the PeLEDs and lots of methods were proposed[17]. Since the halide defects could be passivated by just supply enough of halides to PeNCs, in most cases, the halides were provided with proper exchanging ligand pairs to improve charge carrier injection as mentioned[28, 32, 33]. However, there are several interesting methods to passivate the halide defects without using halides for blue PeLEDs. By

using tetrabutylammonium p-toluenesulfonate (TBSA) and sodium dodecylbenzenesulfonate (SDSA), Ye *et al.* successfully exchanged halides in the PeNCs and passivated halide vacancies[34]. They achieved blue PeLEDs with performance of 2.6% of EQE for 496 nm emission peak. To passivate the Cl vacancies, Zheng and coworkers used n-dodecylammonium thiocynate (DAT)[35]. The thiocyanate group in DAT acted as pseudo halides, passivated the halide vacancies, and removed electronic trap states (Fig. 1(e)). The blue PeLEDs based on this PeNCs achieved EQE of 6.3% at 471 nm emission peak.

Recently, Dong *et al.* reported surface engineering method on CsPbBr$_3$ quantum dots for blue PeLEDs[18] (Fig. 1(f)). They used 4 nm sized PeNCs with a strong quantum confinement effect to achieve blue emission spectra. The surfaces of PeNCs were treated with isopropylammonium bromide (IP-ABr) to passivate the Br vacancies on the surface and to form anionic inner shell. After then, a NABr solution was added to exchange the IPA$^+$ on the surface with Na$^+$ and to form outer shell. The PeNCs with bipolar shell structure exhibited near-unity PLQY result. The PeLEDs with this PeNCs achieved EQE of 12.3% and maximum luminance of ~ 250 cd/m^2 at 479 nm emission peak, which is the highest performance of the blue PeLEDs with PeNCs. This bipolar shell strategy suggested that stable blue PeNCs inks could be achieved without use of any organic ligands. The ionic bipolar shell successfully passivated halide defects and greatly reduced charge carrier injection barrier, so resulted with the highest performance of blue PeLEDs with PeNCs for now. Even more, as the PeNCs in this method were based on a single halide perovskite, the blue PeLEDs from this method could be expected to have enhanced spectral stability due to the lack of halide segregation.

2.2. PeLEDs with 2-dimensional (2D) and quasi-2D perovskites

2D perovskites and quasi-2D perovskites have formation of $R_2A_{n-1}B_nX_{3n+1}$, where R is an aryl or alkyl group, and n value represents the number of layers of perovskite crystals. Considering that $n = 1$ is 2D perovskite and $n = \infty$ is 3D perovskite, the perovskites with the n value bigger than 1 called as quasi-2D perovskites (Fig. 2(a)). These 2D and quasi-2D perovskites are formed with the assistance of organic spacing molecules, and importantly, the number of perovskite layers are tuned by selection of the organic spacing molecules and by controlling the condition of those molecules. For ideal case ($n = 1$), the 2D perovskite could be expressed as $(R-NH_3)_2BX_4$, also known as the Ruddlesden-Popper perovskite[36]. The big advantage of the 2D and quasi-2D perovskite for blue PeLEDs is that blue emissive perovskite could be obtained without use of any Cl precursors by quantum confinement effect. The first PeLEDs with 2D perovskites was suggested by Liang *et al.*[37] using 2-phenylethylammonium (PEA) as spacing molecule. The fabricated $(PEA)_2PbBr_4$ film showed luminescence peak at 410 nm and the PeLEDs resulted EQE of 0.04%. Cheng *et al.* used 4-phenylbutylamine (4-PBA) for the 2D perovskites[38]. They made sky-blue color perovskites films and PeLEDs showed EQE of 0.015%. Unfortunately, too much organic spacing molecules in the film can disrupt and degrade the charge carrier transport, and can lead to decrease of radiative excitons in the emissive layers. Moreover, the performance of the PeLEDs using 2D perovskites is limited by the
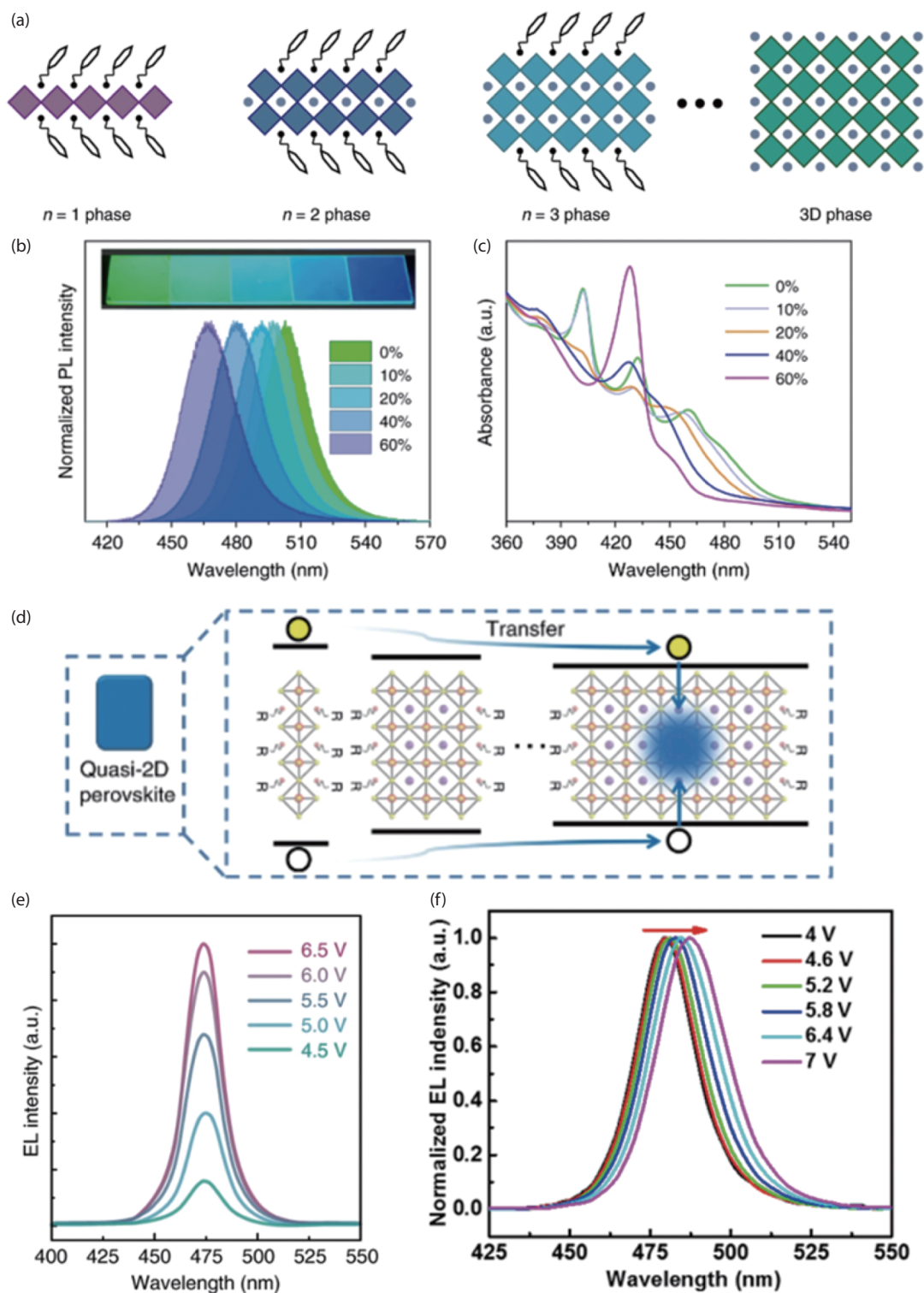
Fig. 2. (Color online) (a) Schematic diagrams of the Ruddlesden-Popper perovskite and quasi-2D perovskites[44]. Copyright © 2019, Springer Nature. (b) Emission spectra and (c) absorption spectra of quasi-2D perovskites with control of n values by adjustments of optical spacing molecule concentration[56]. Copyright © 2018, Springer Nature. (d) Schematic diagram of energy transfer in a quasi-2D perovskite film with mixed *n* values[45]. Copyright © 2019, Springer Nature. Operational stability of blue PeLEDs based on quasi-2D perovskite with (e) single halide composition[44] and (f) mixed halide composition[52]. Copyright © 2019 and 2020, Springer Nature.

strong exciton-phonon coupling[39]. Therefore, pure 2D perovskites have limits for emissive applications.

The emissive layers with quasi-2D perovskites have less organic spacing molecules in the film, resulted with improved optoelectronic properties by reduced exciton-phonon coupling. Additionally, the quasi-2D perovskites form an efficient en-

ergy level structure. When a quasi-2D perovskites are formed, the film usually has a mixed phase state with several *n* values, not a uniform phase with a single *n* value. This mixed phase state naturally constructs energy funnel structure that provides better charge transport, higher radiative recombination chances, and improved device performance[40] (Fig. 2(d)).

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Several successful results for PeLEDs using quasi-2D perovskites have been reported, supported by nice stability and pinhole-free morphology[41−45]. The most important part determining the performance of the blue PeLEDs with quasi-2D perovskite is organic spacing molecules, which determine the number of perovskite crystal layers and charge carrier transport behavior, so various organic molecules have been tested for the quasi-2D perovskites and PeLEDs.

Kumar et al. proposed a blue PeLEDs with quasi-2D perovskite that have formation of $OLA_2MA_{n-1}Pb_nBr_{3n+1}$, where $n = 3–5$ and OLA = oleylammonium[46]. They got device performance of EQE of 0.024% and maximum luminance of ~1 cd/m² at 456 nm emission peak. By using n-butylammonium (BA), the Congreve et al. achieved a blue PeLEDs EQE of 0.0054%[47]. With introduction of 2-phenoxyethylamine (PEOA) group as spacing molecules, device performance was improved to EQE of 1.1% and maximum luminance of 19.25 cd/m² at 480 nm emission peak[48]. Wang et al. reported quasi-2D perovskites with ethylammonium bromide (EABr)[49]. They achieved deep blue PeLEDs with EQE of 2.6% and maximum luminance of 100 cd/m² at 473 nm emission peak. Vashishtha et al. achieved successful blue PeLEDs with quasi-2D perovskites with BA, with performance of EQE of 6.2% and maximum luminance 3340 cd/m² at 487 emission peak[50]. By using mixed organic molecules with guanidine (GA) and PEA, Zhang et al. reported blue PeLEDs with EQE of 8.2% at 492 nm emission peak[51]. Chu et al. boosted performance of blue PeLEDs with quasi-2D perovskites over 10%[52]. They used PEA molecules to form quasi-2D phases and used EA molecule to control the crystal structure and achieved EQE of 12.1%, which is the highest performance result with quasi-2D perovskites for blue PeLEDs for now.

The blue PeLEDs with quasi-2D perovskite with single halide composition, Br, have intensively been studied due to easy of fabrication, free from the halide segregation ensuring stable color spectra (Fig. 2(e)), and excellent performances. However, most of quasi-2D perovskites exhibited sky-blue color, at around 490 nm emission peak. One of the main reasons for the sky-blue color result is that it is hard to obtain pure single phase quasi-2D perovskites with low n value through solution process. The emission spectra of quasi-2D perovskites at mixed phase state is determined by the largest n value in the film. Therefore, if the film could not have pure phases with low n value, which is hard to achieve, the emissive layer could not exhibit the pure blue color emission. Additionally, as the emission peak of the quasi-2D perovskites is not continuous with n values, it is also hard to obtain desired specific emission peak at around 470 nm. Therefore, obtaining pure blue color PeLEDs around 470 nm with quasi-2D perovskites with a single halide composition is difficult. Moreover, the emission spectra from single halide quasi-2D perovskite films have shown broader or multiple emission peaks that reduces great advantages of the PeLEDs, by uncontrolled radiative recombination from the mixed phases in the films. Several attempts were conducted to obtain pure blue emission spectra with quasi-2D perovskites, with participation of Cl elements[53−56], and device performances were improved up to EQE of 3.1% by Zhang et al. at emission peak at 474 nm[57]. However, as participation of Cl elements means there could be the halide segregation problem, improving the spectral stability of the quasi-2D based pure blue PeLEDs

with mixed halide composition is a major remaining problem (Fig. 2(f)).

2.3. PeLEDs with bulk (3D) perovskites

Low-dimensional perovskites have alternative way to obtain blue emission with assistance of the quantum confinement effect, and the properties can be enhanced by organic molecules. However, it is inevitable to use Cl element to achieve the blue emission with bulk perovskites, which is the biggest problem to obtain highly efficient blue PeLEDs. The inorganic Cl precursors are rarely solved in most of solvents, which leads poor film morphology resulting limited performances (Fig. 3(a)). Fortunately, Cl precursors with organic counter cations have better solubility and demonstrated the potential of bulk perovskites for blue PeLEDs.

The first blue PeLEDs with bulk perovskite was reported by Kumawat et al. with $MAPb(Br_{1−x}Cl_x)_3$ ($x = 0–1$), with clear blue color emission[58]. They showed the emission spectra could be easily tuned with control of the halide precursor ratio. They also successfully fabricated blue PeLEDs with EQE of ~1 × 10⁻⁴ % and maximum luminance of ~2 cd/m² at 482 nm emission peak. This limited performance could be attribute to poor morphology with many pinholes in the film[59]. Several attempts of modifying precursor materials were conducted to improve the solution processability and quality of the perovskite films. For example, lead acetate was introduced in precursor solution to achieve better solution processability and pinhole-free film[60].

Producing emissive layers through mixed A-site cations could be an effective way to obtain high-quality films. By mixing the A-site cations, the formation energy of the perovskite could be increased, the defect density in the perovskite crystals could be reduced, and the device performance could be enhanced. Moreover, with reduced defect density, the halide segregation could be reduced to have better spectral stability (Figs. 3(c) and 3(d)). Kim et al. used three types of cations (Cs, MA, FA) for bulk perovskites and successfully fabricated blue PeLEDs with EQE of 1.7% at 475 nm emission peak[61]. Yuan et al. developed this mixed cation concept. They used five types of cations to fabricate the bulk perovskite and achieved EQE of 2.01% at 484 nm emission peak[62]. The mixed cation perovskites were further developed with vapor-assisted crystallization technique. Karlsson et al. used three cations (Rb, Cs, FA) for perovskite films and the PeLEDs resulted EQE of 11.0% and maximum luminance of 2180 cd/m², which is the highest performance of blue PeLEDs with bulk perovskites for now[63].

Though mixing cation is effective strategy to improve the performance of the blue PeLEDs, the bulk perovskites with organic cations have some problems that moisture absorbing properties of organic cation combined Cl precursors, and resulted with high defect density in the pure blue color PeLEDs. However, the inorganic bulk perovskites for blue PeLEDs are much more difficult to fabricate due to even lower solubility of inorganic Cl sources. To solve the problem of insoluble Cl precursors, alternative ways to get wide optical bandgap were tried. Wang et al. reported that introducing the Rb cations into the Cs based perovskite causes lattice distortion and widens the optical bandgap[64]. They successfully fabricated inorganic blue PeLEDs with RbCl precursors with EQE of 0.062% for blue PeLEDs. Yoon et al. introduced anion
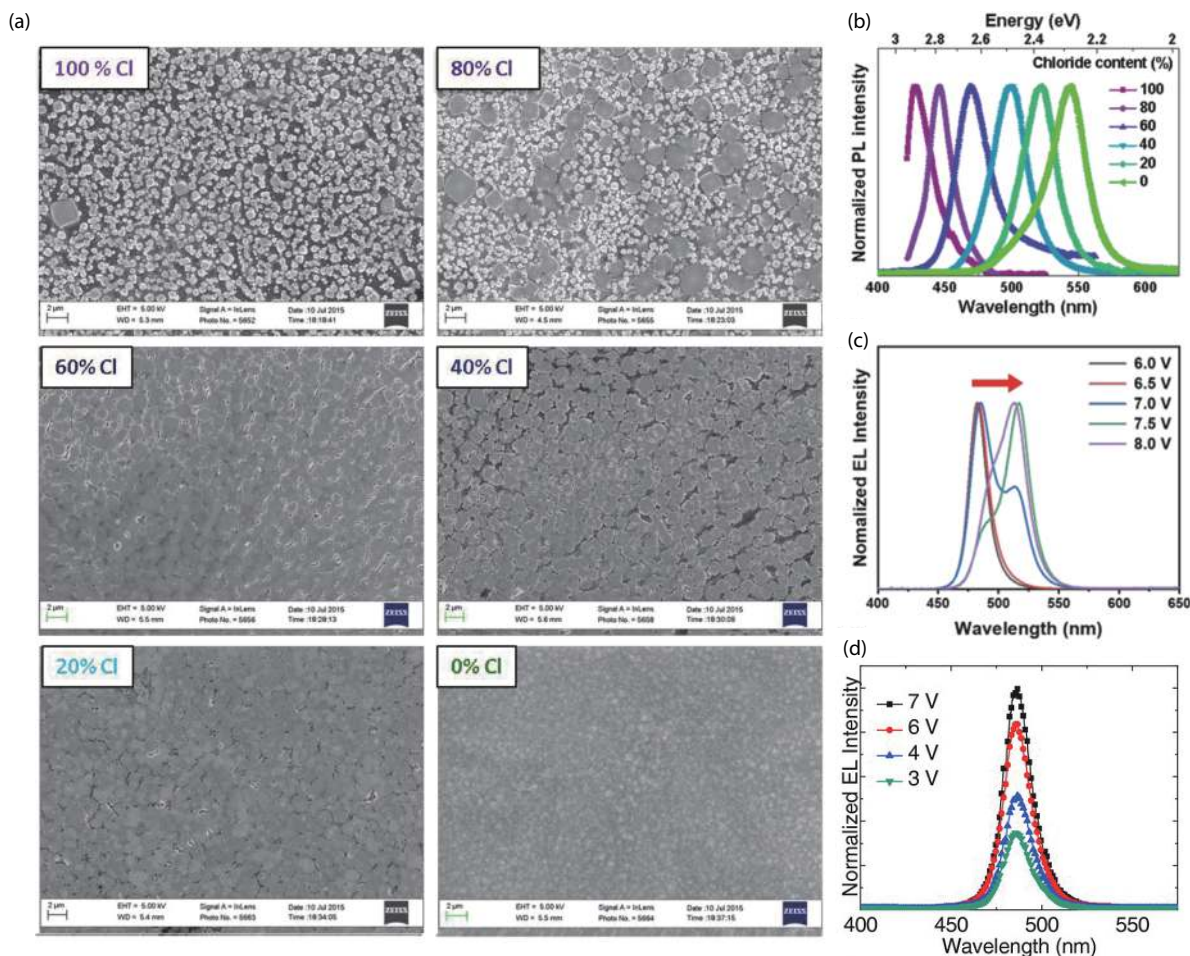
Fig. 3. (Color online) (a) Surface images of bulk (3D) perovskite with varying Cl contents in the film and (b) corresponding PL spectra of bulk perovskites[60]. Copyright © 2015, American Chemical Society. EL spectra operated under different applied voltages of the blue PeLEDs based on the (c) single A site perovskite (CsPbX$_3$)[65] and (d) triple A site perovskite ((Cs/MA/FA)PbX$_3$)[61]. Copyright © 2021, American Chemical Society. Copyright © 2017, John Wiley and Sons.

exchange technique for inorganic bulk blue PeLEDs[65]. They fabricated bulk CsPbBr$_3$ film and then exchanged anions with simple post-treatments and blue PeLEDs showed EQE of 0.32% with excellent stability. With assistance of additives, Cheng *et al.* reported simple fabrication method to obtain high-quality inorganic perovskite films[66]. By adding poly-oxyethylene sorbitan monolaurate (Tween) and tetraphenylphosphonium bromide (TPPB) into the precursor solution, the solution processability was improved with controlled solution wettability, the film morphology was improved, and the halide segregation was effectively prevented. The device showed EQE of 4.13% and maximum luminance of 9352 cd/m^2 at 482 nm peak.

The bulk perovskites have disadvantages such as difficulties in film morphology, exciton quenching between charge transport layers and emissive layer due to lack of electronic band confining organic molecules, and halide segregations. However, the bulk perovskites have advantages on cheap precursors, simpler fabrication methods, stable to most of non-polar solvents which enables fabricating organic layers on top of emissive layers by solution processing methods. In addition, the bulk perovskites are free from the charge blocking organic molecules, the bulk perovskite could have better maximum luminance with higher current injection when the material quality and stability are guaranteed.

3. Developments in charge transport/ injection layers

3.1. Hole transport/ injection layers (HTL/HIL)

With excellent wetting property to form all three types of perovskite emissive layers, poly(3,4-ethylenedioxythiophene): polystyrenesulfonate) (PEDOT:PSS) is widely used for blue PeLEDs, and the p–i–n structure that using PEDOT:PSS as a substrate is common structure for the blue PeLEDs. However, due to the deep valence band maximum of blue emissive perovskites and shallow HOMO (highest occupied molecular orbital) of the PEDOT:PSS, there is a large energy barrier to inject holes from PEDOT:PSS layer to the emissive perovskite layers. Moreover, the acidic property of the PEDOT:PSS layer could be potential origin of degradation of devices. Therefore, to match the energy levels with emissive layer, to improve the charge injection, to match the charge injection balance, and to enhance the device stability, various genuine HTL/HIL materials and related engineering techniques to improve the properties of HTL/HIL were suggested for all three types of blue emissive perovskite materials (Fig. 4(a)).

Jang *et al.* improved wettability, compatibility of PEDOT:PSS, which results better perovskite crystal growth process, with assistance of conjugate polyelectrolytes (CPEs)[67]. In-
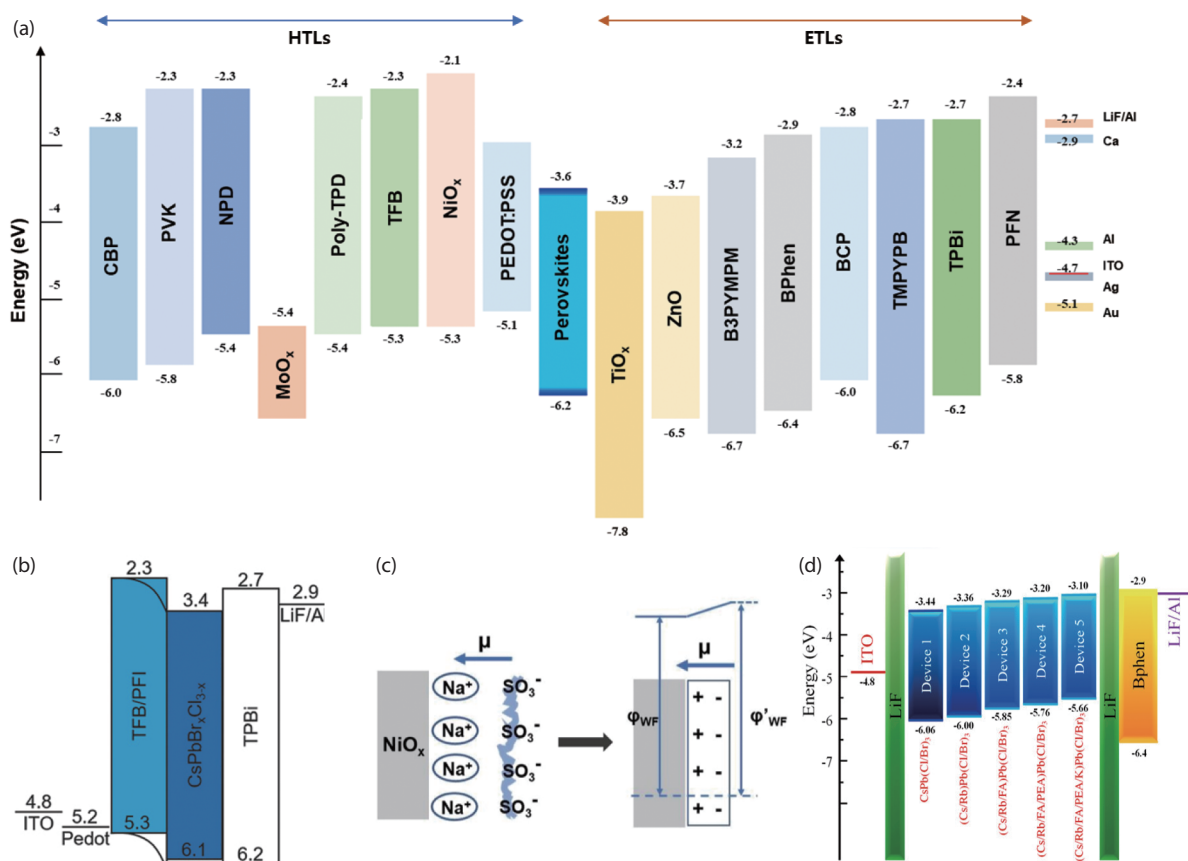
Fig. 4. (Color online) (a) Energy levels of various charge transport/ injection layer materials with blue emissive perovskite. (b) TFB/PFI bilayer structure strategy to reduce hole injection barrier[72]. Copyright © 2018, John Wiley and Sons. (c) Dipole assisted energy level tuning strategy to reduce hole injection barrier[75]. Copyright © 2019, John Wiley and Sons. (d) Interfacial engineering with thin insulating layer to prevent leakage current[62]. Copyright © 2020, American Chemical Society.

troducing the neutral HTL/HIL material with deep HOMO level is also effective method to improve the charge injection and chemical stability. The polymers that have deeper HOMO level, such as poly[N,N'-bis(4-butylphenyl)-N,N'-bisphenyl-benzidine] (poly-TPD)[29, 68], Poly(9-vinylcarbazole) (PVK)[69, 70], CBP[30], have widely been used for better charge injection with PeNCs. Additionally, the energy level of these polymeric HTL/HIL could be modified with further engineering methods[71]. Gangishetty et al. reported the successful charge injection barrier reduction using poly[(9,9-dioctylfluorenyl-2,7-diyl)-co-(4,4'-(N-(4-sec-butylphenyl) diphenylamine)) (TFB) and Nafion perfluorinated ionomer (PFI) bilayer structure[72] (Fig. 4(b)). Shin et al. also reported hole injection barrier could be reduced with CPEs with formation of dipole moment, improving EQE of blue PeLEDs with PeNCs to 1.34% at 470 nm emission peak[73].

However, the polymeric HTL/HIL could disturb perovskite crystal growth due to low wettability of precursor solutions in polar solvents for the bulk perovskites. Considering the disadvantages of organic HTL/HIL, the inorganic oxide HTL/HIL could be promising HTL/HIL for the bulk blue LEDs[74]. Ren et al. introduced PSS Na to form a dipole moment on the NiO$_x$ layers to modify the energy level of the NiO$_x$ layers (Fig. 4(c)). With modified energy level, the hole injection was improved, and non-radiative recombination was reduced. The performance of the blue PeLEDs was improved to EQE of 1.45%[75].

3.2. Electron transport/ injection layers (ETL/EIL)

As the p–i–n structure is common structure in the blue PeLEDs field, usually the ETL/EIL are fabricated upon the blue emissive perovskite layers. The main problem is that the perovskite layers have weak chemical stability and are easily decomposed with polar solvents, making difficult to fabricate ETL/EIL with solution processing methods. Therefore, most of the ETL/EIL for blue PeLEDs are formed through a thermal evaporation method, which is a difficult method to control the chemical properties of the materials. In addition, most of the ETL/EIL materials used for blue PeLEDs already have shallower energy level than perovskite materials (Fig. 4(a)), so the bottle neck of charge injection is HTL/HIL usually. As a result, not many approaches have been reported to make significant changes to control the properties of the ETL/EIL materials itself for blue PeLEDs.

For the n–i–p structure, perovskite emissive layers are fabricated onto the ETL/EIL. Because most known ETL/EIL materials are organic materials that disturb the use of polar solvents due to poor wetting properties, bulk perovskites are almost impossible to fabricate onto the organic ETL/EIL materials. However, the PeNCs or the quasi-2D perovskite emissive layers already have PEDOT:PSS material that has superior wetting properties for the p-i-n structure. Therefore, only a few n–i–p structure based PeLEDs with bulk perovskites were tested with oxide ETL/EIL materials[60, 61]. But, since the typical oxide ETL/EIL materials have a deeper conduction band en-

ergy level than that of the blue emissive bulk perovskites, the basic development strategy is modifying conduction band energy level to have shallower level to reduce the electron injection barriers. For instance, Sadhanala *et al.* successfully demonstrated Mg-doped ZnO could be used as a successful ETL/EIL for blue PeLEDs with better device electronic band structure[60].

3.3. Recombination zone control

It is obvious that the electron and hole transport/injection behavior should be carefully controlled to recombine with each other in the proposed recombination zone, the emissive perovskite layers, as well as to balance the charge carrier injection. However, as the perovskite emissive layers are very thin, less than few tens of nanometers, the charge carriers could be recombined not at the emissive layer but at the adjacent interlayers. Generally, the position of the recombination zone could be controlled by adjusting the thickness of interlayers[76]. Li *et al.* successfully tuned the recombination zone by tuning the thickness of the PEDOT:PSS layer[45] and improved device performances with blue PeLEDs.

Additionally, introducing a very thin insulating materials between the interlayers and the emissive layers could support the charge carriers to recombine at the emissive perovskite layers. The thin insulating layers allow charge injection by tunnelling effect and provide energy barrier to prevent the leakage of holes to ETL/EIL or electrons to HTL/HIL[42]. Yuan *et al.* introduced thin LiF layer for both HTL/HIL and ETL/EIL interfaces (Fig. 4(d)), prevented charge carrier leakage, and successfully confined recombination zone with improved performance of blue PeLEDs with 3D perovskites[62]. Furthermore, because the emissive perovskite materials can react with ZnO ETL[77], introducing thin insulating layers could be protection[78] to improve chemical stability of blue PeLEDs and could be effective method to control the recombination zone.

4. Remaining challenges

4.1. Quantum efficiency of emissive layers

The EQE of the PeLEDs is determined by internal quantum efficiency (IQE) and the outcoupling efficiency of the device. The IQE is the product of charge carrier balance, the fraction of excitons capable of radiative decay, and the effective radiative quantum yield[15]. To enhance the device performance, the IQE must be improved. As the fraction of excitons capable of radiative decay is related to material characteristic and hard to control, and the charge carrier balance could be simply adjusted through optimizing the interlayers and device structure, therefore, the main challenges to improve the device efficiency is matter of improving the effective radiative quantum yield, which is also related to the PLQY of emissive layers. As wider bandgap materials tend to have more sub-bandgap trap states[79], the blue emissive perovskite materials tend to have more non-radiative trap-assisted recombination than the green, red, and near-IR emissive perovskite materials, resulting lower PLQY. Therefore, the underlying strategy for improving the blue PeLEDs is improving the effective radiative quantum yield of the device through the passivation and prevention of defects in emissive layers that creates the sub-bandgap trap states.

For PeNCs, surface defects are the main cause of trap states that lowers the effective radiative recombination. The common ligands, oleic acids and/or oleylamines, could be easily detached with purification process or environmental reasons, so exchanging these ligands to the ligands that make stronger bond is effective way to prevent and passivate the surface defects of PeNCs. For now, various ligands, such as ligands that have secondary amines[70], diamine groups[28, 29, 80], bifunctional groups[81], aromatic groups[82], zwitterionic ligands[30], shorter alkyl chains[68], and much more results have been showed improved PLQY for blue emissive PeNCs. Another strategy to improve the PLQY of the PeNCs is doping metal ions, passivating non-radiative surface cationic vacancies[83]. Successful improvements of PLQY for blue emissive PeNCs were conducted with Mn^{2+}[84], Zn^{2+}[85], Sn^{2+}, Cd^{2+}[86], Nd^{3+}[87], metal cation doping.

For 2D and quasi-2D perovskites, various successful defect engineering techniques were reported[32, 88], however, reducing exciton-phonon interaction is another key issue to improve the effective radiative quantum yield. With control of ligand configuration, Gong *et al.* successfully suppressed electron-phonon coupling and improved PLQY of the blue quasi-2D perovskites to 79%[89]. Peng *et al.* also reported suppressed phonon coupling and enhanced PLQY with control of phases of quasi-2D perovskites[90]. As energy level of the bulk perovskites are not confined and the carriers in the bulk perovskite are tends to flow out to interlayers, the bulk perovskites for blue emission usually showed lower PLQY values. However, several successful improvements of PLQY with defect engineering in grain boundaries in bulk perovskites were reported in other fields, and we could easily expect that those strategies can also work for blue PeLEDs[91, 92].

Hopefully, attempts to reduce the defect and trap states in the emissive layers have shown successful results with improved PLQY of emissive layers reaching near unity value, and enhanced EQE of the blue PeLEDs. However, despite the extremely high PLQY values, the device performance of the blue PeLEDs remained at around over 10% of EQE. Considering the other color PeLEDs resulted around 25% of EQE which is reaching the theoretical limits, it seems there is still some room for improving the performance of the blue PeLEDs. Assuming the emissive layers are perfect, to improve the device performance, the interlayers should be seriously discussed. The common trend of improving the device performance through interlayer engineering was dealing with the HTL/HIL because the HTL/HIL have inferior carrier transport/injection behavior than the ETL/EIL due to charge injection barriers. Fortunately, many successful attempts on the HTL/HIL improved charge carrier transport/ injection properties, reaching that of commonly used ETL/EIL to balance the charge carrier injection. To achieve the theoretical limit of EQE with blue PeLEDs, it is time to develop ETL/EIL materials to have superior charge transport/ injection behavior than conventional materials, and for HTL/HIL too. From simple strategies to modify the interfaces to have better electronic band structure to synthesizing innovative materials should be conducted to reach extremely efficient blue PeLEDs.

4.2. Operational stability and color stability

The low formation energy is a great advantage of halide perovskites, enabling formation of highly crystalline films

Table 1.   Recent advances of blue PeLEDs.

Type	Perovskite formation	Emission peak (nm)	L_{max} (cd/m^2)	EQE (%)	Ref
Nanocrystal	$CsPb(Br_{0.75}/Cl_{0.25})_3$	452	742	0.07	[25]
	$CsPb(Br/Cl)_3$	490	35	1.9	[28]
	$CsPb(Br_{0.7}/Cl_{0.3})_3$	461	318	1.2	[30]
	$CsPb(Br/Cl)_3$	496	603	2.6	[34]
	$CsPb(Br_{0.59}/Cl_{0.41})_3$	471	465	6.3	[35]
	$CsPbBr_3$	479	250	12.3	[18]
Quasi-2D	$OLA_2MA_2Pb_3Br_{10}$	456	1	0.024	[46]
	$PEOA_2MA_{n-1}Pb_nBr_{3n+1}$ ($n = 1, 2, 3$)	480, 494, 508	19	1.1	[48]
	$EA_2MA_{n-1}Pb_nBr_{3n+1}$	473, 485	100	2.6	[49]
	$(PEA/GA)_2Cs_{n-1}Pb_nBr_{3n+1}$	492	1003	8.2	[51]
	$PEA_2(Cs_{1-x}EA_xPbBr_3)_2PbBr_4$	488	2191	12.1	[52]
	$(PEA/DPPA)_2Cs_{n-1}Pb_n(Br_{0.67}/Cl_{0.33})_{3n+1}$ ($n > 3$)	473	482	8.8	[53]
3D	$MAPb(Br_{0.36}/Cl_{0.64})_3$	482	2	0.0001	[58]
	$(Cs/MA/FA)Pb(Br_{0.5}/Cl_{0.5})_3$	475	3567	1.7	[61]
	$(Cs/Rb/FA/PEA/K)Pb(Br_{0.48}/Cl_{0.48})_3$	484	4015	2.01	[62]
	$CsPb(Br_{0.65}/Cl_{0.35})_3$	482	9352	4.13	[66]
	$(Cs/FA)Pb(Br_{0.6}/Cl_{0.4})_3$	477	2180	11.0	[63]

with solution processing at room temperature condition. However, the low formation energy has another aspect. The low formation energy causes easy and frequent creation of ionic defects which eventually deform and damage the perovskite crystals. These ionic defects sites not only serve as the non-radiative recombination centers, but also impede material stability and operational stability of PeLEDs[93, 94]. Therefore, increasing formation energy of the perovskite can be an efficient way to improve the operational stability of blue PeLEDs. To improve the formation energy of the perovskite, strategy of mixing the A-site cations seems an efficient method. Successfully improved film and device stability results were reported by several groups, and even enhanced stability was reported with mixed A-site cations in photovoltaic fields[95]. Additionally, by using mixed A-site materials for the perovskite materials, the lattice strain of the perovskite could be controlled to improve the phase stability[96].

However, state-of-art mixing A-site technique requires many A-site cations[62], which requires delicate control of fabrication conditions, and it seems most of possible candidates have been tested and hard to find other promising candidates. Even with the state-of-art mixed A-site cation strategy, the stability of blue PeLEDs is far from the commercialization for now. Therefore, another additional strategy to improve the stability of blue PeLEDs that can be combined with mixed A-site strategy should be proposed. As mentioned above, exchanging organic ligands or spacers for PeNCs and quasi-2D perovskite could be one of promising methods to enhance the device stability. Recently, combining 2D perovskite with 3D perovskite strategy was proposed in perovskite photovoltaic devices field[97]. The combined perovskite structure exhibited enhanced carrier lifetime and improved device performance and stability result. Likewise, mixing 2D/3D perovskite strategy could be a promising method for the 3D blue PeLEDs. In addition, introducing the pseudo halide anions to perovskite materials could also be a promising candidate method to improve the device stability[8, 35].

Another big remaining problem that must be solved is color instability. The main cause of this color instability comes from the mobile characteristic of halides in the halide perovskite crystals[98]. The mobile halides in the mixed halide perovskites induce halide segregation phenomena due to thermodynamic reason, which gathers the same type of halides in the crystal, creates unnecessary additional emissive phase, changes overall emission spectra of the perovskite. Interestingly, the origin of halide segregation was also attributed to the ionic defects, which is main reason for degrading operational stability and performance of blue PeLEDs. To suppress the mobile property, eliminating the created ionic defects could be efficient method, and successful stability enhancements were reported with various ionic defect passivation methods[32, 35, 44, 65]. There is also another efficient method to suppress the mobile property, that increasing the formation energy and prevents ionic defects. The formation energy could be increased by using mixed A-site cation strategy[99], and this strategy resulted successful enhancements in both operational stability and color stability for blue PeLEDs[44, 62, 100], especially for blue PeLEDs with the 3D perovskites[61, 63, 64, 101].

However, no matter how well the perovskite composition is controlled, it is impossible to permanently prevent the halide segregation with mixed halide compositions, because the halide segregation strongly affected by uncontrollable entropic factors. Fortunately, we already know the perfect method to prevent the halide segregation, that forming the perovskite with a single halide, Br. Though the Br only perovskites need additional delicate size control to get quantum confinement effect to exhibit blue color emission, but at least there are lots of methods and possibilities to broaden the bandgap for Br only perovskites, while it is almost impossible to prevent the halide segregation with mixed halide perovskites.

5. Conclusions and outlook

We introduced recent developments in the blue PeLEDs (Table 1) and remaining challenges, low quantum efficiency of emissive materials and device instability. With the deep understand of material characteristics, the blue PeLEDs showed dramatic developments in past few years. To improve device performance, the strategy of increasing the formation energy through mixing A-site cations have been effective way

for all three types of perovskites. Reducing Cl contents in the perovskite material and reducing the size of the perovskite crystal could be effective method to obtain efficient and color stable emissive perovskite layer for sky-blue PeLEDs. For the pure blue PeLEDs with narrow emission spectra, the participation of the Cl element seems inevitable for now, so to realize the highly efficient pure blue PeLEDs, the halide defect engineering technique is essential to handle the trap states in the mixed halide perovskites. However, the Cl element should eventually be eliminated to avoid halide segregation and to get enhanced color stability for pure blue PeLEDs in future. Additionally, advanced interfacial engineering techniques and innovative materials for interlayers should be developed to improve charge carrier injection and to reach the theoretical performance limit. With improved performance and long-term device stability, the blue perovskite LEDs will be the best choice for next generation displays.

Acknowledgements

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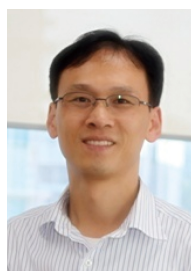
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