TOPICAL REVIEW

Metal halide perovskites-based white light-emitting diodes

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Abstract

White light-emitting diodes (WLEDs) serve as a replacement for traditional incandescent light due to their excellent characteristics, such as high brightness, efficiency in energy consumption, and long lifetime. The high-efficiency and low-cost white-emitting materials and light-emitting diode devices has always been the goal pursued in the field of lighting technology. Recently, metal halide perovskites are emerging as one of the most promising luminescent materials for next-generation WLEDs due to their facile synthesis and excellent optoelectronic properties, such as high photoluminescence quantum yields, widely tunable bandgap, and high charge-carrier mobility. Although the luminescence efficiency of perovskite emitters and WLED devices has increased rapidly over the past several years, achieving high-efficiency and stable WLEDs remains great challenges. In this review, we focus on the recent progresses of WLEDs based on metal halide perovskites including color-conversion WLEDs, tandem structure of WLEDs, double-emissive-layer of WLEDs, and single-emissive-layer of WLEDs. Importantly, we highlight the WLEDs based on a single emissive layer that show white electroluminescence directly from the perovskite emitters. Finally, we will give an outlook of future research avenues on how to reach the goal of efficient and stable perovskite-based WLEDs.

1. Introduction

Light sources have always played a key role in the history of human development, which experienced changes from fire to electricity [1]. Since the development of incandescent light bulbs in the late 1800s, various lighting technologies have been extensively investigated (figure 1) [2, 3]. With the advent of bright double-heterostructure blue light-emitting diodes (LEDs) for the first time in 1993, it is now possible to generate white light using LEDs and enables the rapid development of WLEDs [4, 5]. Since then, white-light sources-based reliable and efficient LEDs are used to replace incandescent bulbs and compact fluorescent lamps. However, the commercial mainstream white LEDs (WLEDs) based on yellow phosphor-coated blue LEDs have some drawbacks, such as low color rendering index (CRI) and expensive epitaxy growth techniques [6–8].

The efficient light sources that combine high brightness, high CRI, and good stability are in a strong worldwide demand for next-generation lighting technology [9, 10]. Over the past few decades, a variety of luminescent materials (e.g. organic molecules, quantum dots (QDs), metal halide perovskites) have been explored and used in the development of WLEDs [11–18]. In 1995, Kido et al reported the first white OLEDs based on vacuum-deposited organic thin films [19]. Since then, white OLEDs have made great achievements in the field of lighting, bringing the major advantages of high light utilization rate and flexibility [20], but the light-emitting devices are limited by their operational lifetimes [21, 22]. These problems can be solved by integrating semiconductor QDs into inorganic LEDs due to their excellent thermal stability and high resistance to photo-oxidation [23–25]. Compared with conventional QDs (such as CdSe and InP), metal halide perovskites possess high defect tolerance, resulting in bright luminescence without any electronic surface passivation, and low-cost fabrication for lighting applications [26–29]. Therefore, the development of single-color perovskite LEDs has made rapid progress in the past few years, reaching high external quantum...
efficiencies (EQEs) of more than 20% of green, red, and infrared light-emitting devices [30–37]. Moreover, recent studies have shown that metal halide perovskites have broadband emission or multi-color characteristic, which have demonstrated great potential in high-quality lighting applications [38–40].

In this review, we present the recent progresses of perovskite-based WLEDs from the point of view of device construction, such as color converters, multiple-emissive-layer, double-emissive-layer, and single-emissive-layer of devices. We then focus on the WLEDs based on a single emissive layer fabricated by the solution method that enables more cost-efficient lighting technology. Finally, we put forward an outlook on the realization of high-efficiency and stable WLEDs based on metal halide perovskites.

2. Color-conversion WLEDs

In general, the emission of metal halide perovskites exhibits narrow line width (the full width at half maximum (fwhm) of <40 nm), which can be adjusted to cover the visible spectrum by changing the proportions of halides as well as the size [41, 42]. One easy way to obtain white light is to optically mix several emission colors of the perovskite materials (e.g. red, green, and blue) in appropriate proportions. However, the ion migration in mixed-halide perovskites originates from the ionic character and low migration activation energy of halide vacancy [43–46], resulting in the rapid change of emission color during device operation, which severely hinder the practical application of perovskite-based WLEDs. Therefore, the color-conversion is an effective strategy to achieve multi-color characteristic and finally realizing a white light emission [47–53].

2.1. Perovskites combined with blue/UV chips for WLEDs

As mentioned above, WLEDs based on a blue/UV LED component with phosphors are the most common technique for generating a bright white emission [54, 55]. The light generated from blue/UV LED chips passes through a color conversion layer and excites the phosphors. As excitons relax, light with longer wavelength is emitted, achieving color-conversion from original wavelength of light into various colors [12]. The metal halide perovskites offer the possibility of obtaining high CRI and appropriate correlated color temperature (CCT), and even emitting suitable visible parts of sunlight with less UV/blue component, thus achieving healthy lighting [7]. Based on this concept, WLEDs were assembled by combining blue LED chip with perovskite QD/poly(methyl methacrylate) (PMMA) composites (figure 2(a)) [56]. The electroluminescence (EL) spectra of LED devices contain three luminescent peaks (figures 2(b) and (c)), which correspond to blue chips with a central wavelength of about 460 nm, green perovskite, and red perovskite QDs, respectively. Meanwhile, the color temperature of the WLEDs can be tuned ranging from 2500 to 11 500 K through controlling the ratio of green to red perovskite QDs. However, the intrinsic instability of perovskite materials with soft ionic crystal structure is biggest obstacle to the commercialization of perovskite WLED lighting [57–59]. In recent years, many studies attempted to overcome the low device stability [60]. An efficient strategy for improving the stability of perovskites is to cover them with stable and transparent polymer materials [61–64]. For example, Zhong et al prepared MAPbX3 NCs embedded polyvinylidene fluoride composite films via an in-situ fabrication strategy [65]. The composite films exhibit
enhanced PL properties and improved stability to water and ultraviolet radiation. Furthermore, they produced a highly efficient wide-gamut WLED with luminescent efficiency of 109 lm W\(^{-1}\) and color gamut of 121% National Television System Committee (NTSC) by combining green luminescent composite films and red luminescent K\(_2\)SiF\(_6\):Mn\(^{4+}\) phosphor with a blue InGaN chip. In addition, more and more types of materials are developed for the synthesis of perovskite composites \([51, 66–68]\), such as mesoporous silica, mesoporous alumina, and metal–organic frameworks as matrices in which to host perovskite, exhibiting much improved material stability and high photoluminescence quantum yield (PLQY) values. These strategies are also beneficial to further improve the performance of WLEDs.

It is worth noting that doping with the targeted impurities has proved to be one of the effective strategies to regulate the optical properties of perovskites \([69–71]\). Particularly, Mn\(^{2+}\) has been intensively investigated due to its functionality in both strengthening the binding of perovskites matrix and acting as emitting center to produce bright red fluorescence, which can be identified as one of the promising down conversion phosphors for WLEDs \([72, 73]\). Bai et al have doped Mn\(^{2+}\) ions into 2D (PEA)\(_2\)PbBr\(_4\) to achieve dual-color emission center, including 410 nm from the exciton transition of the host and 600 nm from the d–d transition of Mn\(^{2+}\) \([74]\). The Mn\(^{2+}\) ions introduce a new exciton recombination channel within the band gap of perovskites, which leads to the \(4^1T_1\rightarrow6^1A_1\) internal transition emission of Mn\(^{2+}\) ion at 600 nm through the energy transfer of excitons (figure 2(g)). Furthermore, WLEDs was prepared by uniformly coating the mixture of the efficient dual-color emitting Mn\(^{2+}\) doped (PEA)\(_2\)PbBr\(_4\) NCs, CsPb(Cl/Br)\(_3\) NCs (495 nm emission), and PMMA/toluene on UV-LED chips (405 nm) (figure 2(f)). In lead-free perovskite systems, Zhang et al implemented a melting-crystallization strategy without solvent to prepare kilogram-scale Bi\(^{3+}\)-doped Cs\(_2\)Na\(_x\)Ag\(_{1–x}\)InCl\(_6\) microcrystals \([75]\), which show a white light broadband emission (400–850 nm) and a high PL efficiency (figure 2(e)). By coating perovskites combined with commercial blue and green phosphors (figure 2(d)), they fabricated WLED with a high CRI of 97.1 and Commission Internationale de l'Eclairage (CIE) color coordinates of (0.331, 0.339).

2.2. Integrated perovskite-based conversion layer and EL for WLEDs
The integrated perovskite-based conversion layer and EL for white emission is another promising strategy in lighting technology, which can effectively avoid the use of blue/UV-LED chips. In 2018, Congreve et al have
produced WLED devices with a CIE coordinate of (0.311, 0.326) via placing red and green perovskite NCs on the glass substrate of perovskite-based blue LEDs (figures 3(a) and (c)) [76]. The EL centered at 469 nm is consistent with the blue perovskite LEDs. The down-converted emission centered at 511 and 627 nm is matching the PL measured on the red and green perovskites individually (figure 3(b)). The inferior performance of WLED (the device has a maximum brightness of 102 cd m$^{-2}$ and an EQE of 0.25%) is mainly attributed to the waveguide losses in the thin films. Furthermore, Chen et al demonstrated that the captured photons in the waveguide and surface plasmon polariton modes can be efficiently extracted by optimizing the device structure to improve the performance of perovskite WLED [77]. It comprised a layer of red perovskite NCs, used as a down converter, coated on blue perovskite LEDs with ultra-thin transparent top electrodes (figure 3(d)). Based on this, the light extraction efficiency in the device has been increased by more than 50% (figure 3(e)), and achieving high-performance WLED device with a high EQE of 12% and approximately 2000 cd m$^{-2}$, respectively (figure 3(f)), which represent the most efficient perovskite-based WLEDs (table 1).

### 3. Tandem structure of perovskite-based WLEDs

Tandem structures of LEDs with stacked two or more light-emitting layers are an efficient technique to achieve white emission, which have been commonly used to improve device performance of WLEDs [78, 79]. In LEDs with tandem structures, the different emitting layers are usually connected by intercalation layers. Therefore, white emission can be realized by tandem structure of perovskite-based LEDs on the basis of solving the major challenge of ion exchange in mixed-halide perovskites [80]. As a typical example, Choy et al have demonstrated all-perovskite WLEDs based on two dimensional red-light perovskite (CH$_3$CH$_2$CH$_2$NH$_3$)$_2$CsPb$_2$I$_7$ (PA$_2$CsPb$_2$I$_7$), blue-light CsPb(Br/Cl)$_3$ and an interlayer of bis(1-phenyl-1H-benzo[d]-imidazole)-phenylphosphine oxide (BIPO), poly(4-butylphenyldiphenyl-amine) (poly-TPD) (figure 4(a)) [81]. The design of the intercalation layer is to ensure a uniform distribution of carriers between the different layers. As shown in figure 4(b), the PL spectrum of PA$_2$CsPb$_2$I$_7$ was at 695 nm with a fwhm of 37 nm, and the PL spectrum of CsPb(Br/Cl)$_3$ was at 492 nm with a fwhm of 20 nm. As shown in figure 4(c), the white light emission can be achieved by combining these two perovskites. With this tandem structure of LEDs, the typical white light emission with steady CIE coordinates of (0.32, 0.32) and a CCT of $\sim$6000 K was achieved in a wide range of driving current densities (figures 4(d)–(f)). It is worth mentioning that the WLEDs based on red perovskite and sky-blue organic p–i–n heterojunction have realized by Su et al in 2022 [82]. They sequentially stacked organic p–i–n heterojunctions on perovskite layers by vacuum deposition.

![Figure 3](image-url)
Figure 4. WLEDs based on the tandem structure. (a) Energy level of tandem structures WLEDs based on PA$_2$CaPb$_2$I$_7$/interlayer/CaPb(Br,Cl)$_3$. (b) Normalized PL and absorption spectra of perovskite films. (c) CIE chromaticity diagram of PA$_2$CaPb$_2$I$_7$/CaPb(Br,Cl)$_3$. (d) Normalized EL spectra of LED devices. (Inset: photo of a WLED). (e) EL spectra and (f) CIE coordinates of WLEDs based on PA$_2$CaPb$_2$I$_7$/BIPO:poly-TPD/CaPb(Br,Cl)$_3$. Reprinted with permission from [81]. Copyright (2018) American Chemical Society.

The organic p–i–n heterojunction includes p-type hole transport layer, ultrathin undoped organic phosphorescent interlayer and n-type electron transport layer. Furthermore, the red emission of the perovskite layer combined with the blue emission of the organic p–i–n heterojunction simultaneously produces white emission with a high EQE of 7.35%, CIE coordinates of (0.424, 0.363), and a low CCT of 2868 K. It should be emphasized that the WLED showed good spectral stability and higher operating lifetimes (ten-fold longer than perovskite-only LEDs). In summary, there are few studies to realize perovskite WLEDs based on tandem structures. The main challenges are the selection of interlayer and orthogonal solvent and the design of device structure. The interlayer needs to be able to effectively avoid the ion exchange between these two perovskites and to facilitate carrier transportation and distribution in the architecture. In addition, since perovskites are easily destroyed by polar organic solvents, it is important to carefully select appropriate solvents when stacking these perovskites layer-by-layer.

4. Double-emissive-layer of WLEDs

In order to simplify the architecture of device, researchers have developed a series of dual emitter layer configuration of WLEDs. In this system, the choice of solvent is particularly important, and orthogonal solvents can effectively avoid perovskite dissolution. In addition, it is also critical to improve the energy transfer efficiency and achieve a reasonable distribution of carriers between the light-emitting layers. For example, it has proven to be an effective strategy to form double emission via spin-coating traditional QDs or organic light-emitting layers on top of perovskite films [83]. As a typical example, Ercan et al mixed perovskites with thermoplastic polyurethane to form multicolor luminescent nanofibers [84]. Meanwhile, white-light emission was achieved by introducing orange-light-emitting poly(2-methoxy-5-(2′-ethylhexyloxy)-1,4-phenylenevinylene) (MEH-PPV) into TPU shell and coupled the blue-light-emitting MAPb(Br$_{1-x}$Cl$_x$)$_3$ nanoparticles. Furthermore, the WLED device was integrated by spin-coating a blue polyfluorene (PFO) emissive layer on top of orange-light-emitting MAPb(I$_{1-x}$Br$_x$)$_3$ nanofibers (figures 5(a)–(c)), showing the CIE coordinates of (0.27, 0.35). In addition, Nripan et al reported WLED devices through combining the blue perovskite materials CsPbA (PBA$_{2m}$mBr$_{3m+1}$, PBA = CsC$_6$H$_5$(CH$_2$)$_2$NH$_3$) films with a commercial organic emitter, Livilux SPR-001, leading to an EQE of 1.3%, as shown in figures 5(d)–(f) [85]. Here, the structural design of the double light-emitting layers can significantly strengthen the spatial separation and suppress the energy transfer, so that the calculated energy transfers efficiency reaches about 30%. However, it is still a challenge to achieve efficient WLED device via combining perovskites with organic fluorescent emitter. Improvements have also been accomplished through the incorporation of phosphorescent emitter in WLEDs. For example, Lee et al report a WLED device via
using a phosphorescent emitter (Ir(BT)$_2$(acac)) with blue quasi-two-dimensional perovskites [86], as shown in figures 5(g)–(i). To improve the energy transfer efficiency from blue perovskites to orange phosphors, they introduced a charge-confinement device structure of quantum wells to enhance carrier trapping, resulting in maximum EQE of 10.81%. Meanwhile, the WLEDs can get a stable white light emission while enabling tunable colors from warm white to daylight by adjusting the concentration of Ir(BT)$_2$(acac) in the organic light-emitting layer.

5. Single-emissive-layer of WLEDs

WLED based on single emissive layer are gaining more and more attention due to their features of multi-color or broad-spectrum, which means that the single layer can emit white light. It can, on the one hand, avoid the problems of self-absorption and color instability that other approaches may bring about. On the other hand, it can simplify the structure of the WLED device [87]. In this section, we focus on the possible strategies to achieve high-efficiency WLED devices based on single emissive layer, which is widely seen as an ideal way in high-quality lighting application.

5.1. WLEDs based on blending perovskites with other emitters

Recent studies have shown that blending metal halide perovskites with organic molecules like oligomer, polymer, or others to create a single emitting layer is a direct way to generate effective white emission (figure 6(a)) [18]. Yang et al obtained white-light emission by blending orange polymer materials, MEH-PPV, with the blue perovskite NCs (CsPbBr$_3$Cl$_{3−x}$ NCs) as the active layer, where excitons transfer from perovskites to the organic polymers via Dexter or Förster energy transfer [88]. As shown in figure 6(b),
the EL spectra showed two emission peaks at blue (470 nm) and orange (560 nm) corresponding to CsPbBr\textsubscript{x}Cl\textsubscript{3-x} and MEH-PPV, respectively. Meanwhile, the relative intensity of the emission peaks varied with the different ratio of CsPbBr\textsubscript{x}Cl\textsubscript{3-x} to MEH-PPV. As a result, white light was obtained with the CIE coordinate at (0.33, 0.34) When the weight ratio of CsPbBr\textsubscript{x}Cl\textsubscript{3-x} to MEH-PPV was 9:1. It is worth noting that the white light can be modulated by varying the ratio of halides in perovskites. Similarly, Liu et al have demonstrated a hybrid WLED with the 9,9-dihexylfluorene co-oligomer and CsPbBr\textsubscript{1.5}I\textsubscript{1.5} QDs for white light emission [89], which exhibits a maximum brightness \(\sim 1200\ \text{cd m}^{-2}\) (figure 6(c)) and CIE coordinates of (0.28, 0.33).

In addition, Mora-Seró et al prepared colloidal QD-based WLEDs by mixing the solution of carbon QDs (CQDs) and CsPbI\textsubscript{3} QDs [90]. It is possible to obtain a complex colloidal solution that emits ‘warm’, ‘neutral’ and ‘cool’ white light, which is sought after for indoor lighting applications via varying the concentration of perovskite QDs (figure 6(d)). Moreover, the WLEDs show a high CRI of 92 as well as a CCT range between 3000 and 7500 K (from warm white to cool white). In addition, the chromaticity coordinates of white emission can be further adjusted by the applied voltage (figure 6(e)). To expand the solvent compatibility of perovskites with organic molecules, some studies have introduced organic materials into polar solvents (such as: dimethylformamide, dimethyl sulfoxide) to form perovskite-organic composite films for white emission [91, 92]. In order to generate white EL, Chao et al blended a sky-blue perovskite with an orange-near-infrared compound (TPA-T-DCV-Ph), obtaining WLED devices with the CIE coordinates of (0.3, 0.49) (figure 6(f)) [92]. Subsequently, they mixed sky-blue MAPb\textsubscript{(0.6,Cl\textsubscript{0.4})} with orange–red rhodamine 6G for achieving white emission [91]. As a result, the CIE chromaticity coordinates of WLEDs were (0.33, 0.4) and (0.36, 0.40) when the rhodamine 6G concentration was 2 wt% and 3 wt%, respectively.

5.2. WLEDs based on metal ion-doped perovskites

Metal ion doping is an effective strategy to achieve multi-color emission in metal halide perovskites, which could potentially be used as a new class of direct white light-emitting materials [93, 94]. For example, doping Mn\textsuperscript{2+} into blue perovskites gives dual emission, including the intrinsic emission of perovskites and the red color from the Mn\textsuperscript{2+}, showing potential in single-emissive-layer WLEDs (SEL-WLEDs) [7]. Meanwhile, a perovskite family including lead-free double perovskites, lead-halide perovskites, and cesium copper halides offer unique self-trapped exciton (STE) characteristics, which shows broadband emission with a large Stokes shift, make them extremely promising materials for WLEDs [47, 95–97]. In 2018, Tang et al reported the lead-free double perovskite Cs\textsubscript{3}AgInCl\textsubscript{6} with alloying Na\textsuperscript{+} and Bi\textsuperscript{3+} that exhibits efficient and stable white-light emission with a high PLQY of 86% via STEs [98]. However, the double perovskites-based WLEDs...
showed poor performance with a luminance of ~50 cd m⁻² due to their poor charge transport characteristics, as shown in figures 7(a)–(c). Furthermore, Qu et al prepared Cs₂AgInₙ₀₉Bi₀₁Cl₆ QDs via a hot-injection method [99], which were used as light-emitting layer in electrically excited WLED (figure 7(d)). It is revealed by microzone optical and electronic characterizations that the broadband emission of Cs₂AgInₙ₀₉Bi₀₁Cl₆ comes from STEs (figure 7(e)). The Cs₂AgInₙ₀₉Bi₀₁Cl₆ QD based-WLEDs have a stable EL emission (figure 7(f)), superior operational stability (T₅₀ of 48.5 min), a maximum luminance of 158 cd m⁻², high CRI of 94.5, and CIE coordinates of (0.32, 0.32).

In another work, Song et al reported the Sm³⁺ doped CsPbCl₃ NCs with white-light emission [100], which can be modulated by varying the concentration of dopant (figure 7(h)). In figure 7(g), the energy-transfer process from the CsPbCl₃ host to Sm³⁺ was shown, which was mediated by strong exchange coupling between carriers. Meanwhile, the overall PLQY of Sm³⁺-doped CsPbCl₃ NCs is close to 85% via optimization of reaction conditions, such as reaction temperature, dissolvability of SmCl₃, and adjustment of surfactant. The WLED based on doped NCs had an EQE of 1.20%, a luminance of 938 cd m⁻², EQE of 1.20%, and a high CRI of 93 (figure 7(i)).

5.3. WLEDs based on phase regulation of perovskites

In addition to the effects on doping mentioned above, the phase of perovskites is also considered to be a key issue affecting their optoelectronic properties in recombination kinetics and bandgap. In recent years, researchers have obtained effective white light emission via the phase regulation of perovskites, which is mainly to modulate the emission spectra of metal halide perovskites via controlling their phase transformation [38, 101–103]. First, we pay attention to the phase regulation of lead halide perovskites. For example, Gao et al developed the SEL all-perovskite WLED based on perovskite films with segregated
CsPb(Br$_{1-x}$Cl$_x$)$_3$ and CsPb(Br$_{1-y}$I$_y$)$_3$ grains [103]. In figure 8(a), the image of secondary electron of perovskite films is shown. It consists of clearly separated perovskite grains with different shapes, rod-shaped grains, and plate-shaped grains, which deliver different cathodoluminescence (CL) (figure 8(b)). The rod-shaped grains exhibit a single emission with a peak wavelength of 498 nm. The plate-shaped grains show double emission peaks at 498 nm and 684 nm, indicating that the CsPb(Br$_{1-x}$Cl$_x$)$_3$ and CsPb(Br$_{1-y}$I$_y$)$_3$ structural domains coexist. Further studies have shown that the benzamidine hydrochloride and PbBr$_2$ are the key factors that lead to phase segregation with blue and red emission (figure 8(c)). The perovskite film exhibits inhibition of ion exchange and prevention charge transfer, resulting in dual-emission white EL with an EQE of 0.008% and CIE coordinates of (0.33, 0.33). Similarly, Sun et al. have shown that composition-tunable properties of perovskites have already been utilized in bright white emission [102]. On a nano- or micron-scale of perovskite film, there are possibilities of different halide ions separated in the two phases without uniform mixing. Meanwhile, the electrons will gather at the energy level with the larger band gap and combine with holes to form excited exciton when there is inefficient energy transfer, leading to dual-color emission (figures 8(d) and (e)). Furthermore, by adjusting the concentration of CH$_3$NH$_3$I in the precursor, white emission with CIE coordinates of (0.33, 0.34) could be obtained (figure 8(f)).

A breakthrough in WLEDs was achieved by designing mixed-phase $\alpha/\delta$-CsPbI$_3$, which used as single emissive layer (figures 9(a) and (b)) [38]. The $\alpha$-CsPbI$_3$ and $\delta$-CsPbI$_3$ were uniformly distributed in the emitting layer via controlling the phase transition of CsPbI$_3$ QDs. By combining the superior carrier transport ability of $\alpha$-CsPbI$_3$ with the broad-band emission of $\delta$-CsPbI$_3$, it can demonstrate highly efficient white-light emission (figures 9(c) and (d)). This heterogeneous structure provided a large interface area between the $\alpha$-CsPbI$_3$ and $\delta$-CsPbI$_3$, which enabled more carriers to be injected into $\delta$-CsPbI$_3$ phase. In addition, it also reduced the length scale of $\delta$-CsPbI$_3$ microstructure, which could promote the possibility of carriers of opposite signs meeting each other and recombining radiatively by forming STEs. With EQE and luminance reaching 6.5% and 12 200 cd m$^{-2}$ (figure 9(e)) respectively, these WLEDs are easily fabricated by solution processing and feature a single broadband emitting layer. Meanwhile, the ratio of mixed-phase $\alpha/\delta$-CsPbI$_3$ could be modulated to change the color temperature of white emission through the adjustment of the annealing processes of $\alpha$-CsPbI$_3$. It is worth mentioning that the WLED showed good spectral stability over 300 min during the operation and the half-lifetime ($T_{50}$) was $\sim$230 min under a current density of 1 mA cm$^{-2}$. In summary, the efficient EL was derived from the combination of the excellent charge injection properties of the $\alpha$ phase, the charge transfer between $\alpha$-CsPbI$_3$ to $\delta$-CsPbI$_3$ and the balance radiative recombination of $\alpha$-CsPbI$_3$ to $\delta$-CsPbI$_3$ (figure 9(f)). The research can significantly reduce the production cost of WLEDs, and further advance the development of next-generation displays and lighting application.
Figure 9. WLED based on α/δ-CsPbI₃ heterophase. (a) Architecture of WLED device. (b) EL spectra and picture of the device. (c) Local fluorescence hyperspectral imaging of heterophase interface. (d) PL spectra of different regions recorded in situ. (e) EQE and CE of WLED. (f) The charge injection and recombination mechanism in WLED device. Reproduced from [38], with permission from Springer Nature.

Additionally, cesium copper halides (e.g. Cs₃Cu₂I₅) have a large Stokes shift, broadband emission, and negligible self-absorption, making them ideal candidates for WLEDs [104–107]. For example, Zhu et al discovered a controllable transformation between different phases of Cu-based perovskites [108], which was used to fabricate WLED. The white emission has a high CRI of 94 with CIE coordinates of (0.327, 0.348). In this approach, the reaction time and solvent polarity can control the transformation rate of Cs₃Cu₂I₅ to CsCu₂I₃. The Cs₃Cu₂I₅ and CsCu₂I₃ are coexistent, emitting blue and yellow light, respectively. Moreover, their respective luminescence does not affect each other in the mixed powder or thin film state. Furthermore, Shan et al synthesized copper-based halide composites coexisting with Cs₃Cu₂I₅ and CsCu₂I₃ that emit both blue and yellow light by a simple solution method (figure 10(a)) [109]. It shows a stable white-light emission with warm/cold white-light modulation via simply varying the ratio of CuI/CsI in precursors and the excitation light wavelength (figure 10(b)). As a result, the WLED devices with a luminance of 145 cd m⁻², an EQE of 0.15%, and a high CRI of 91.6 were successfully prepared via using the copper-based halide composites as the emitter. Meanwhile, the devices demonstrated strong operating stability in air environment, achieving a long T₅₀ of about 238.5 min (figure 10(c)). A further enhancement in WLEDs based on cesium copper halides performance was achieved through chemisorbing ether groups of additives onto the surface of metal halides using as electron donors [110]. This approach can improve the PLQY of the metal halide films and increase the surface potential so that the hole injection and transport in WLEDs can be facilitated (figure 10(d)). Furthermore, the chemical interaction between Cs and Tween could delay the nucleation of cesium copper halides (figures 10(e) and (f)), leading to films with enhanced crystallinity. Therefore, warm WLEDs reached an EQE of 3.1% and a luminance of 1570 cd m⁻², which show great potential of lead-free metal halides for the preparation of high-performance WLEDs by the solution method.
Figure 10. WLEDs based on phase regulation of lead-free halides. (a) Crystal structures of CsCu$_2$I$_3$ and Cs$_3$Cu$_2$I$_5$. (b) EL spectra of the WLEDs prepared with various CuI/ CsI ratios (Insets: the photographs of WLEDs). (c) Evolution of WLED brightness with running time. [109] John Wiley & Sons. © 2020 Wiley-VCH GmbH. (d) EL spectra of the WLED under different voltages. (e) Time-resolved GIXWAXS profiles of cesium copper iodides without Tween (e) and with Tween (f). Reproduced from [110]. CC BY 3.0.

6. Summary and outlook

In this review, we have concluded and analyzed the recent progress of metal halide perovskites-based WLEDs in terms of the different ways to generating white-light emission, including color conversion, tandem configurations, double-emissive-layer, and single-emissive-layer of LED devices. As highlighted in this review, the progress of SEL-WLED devices have been accomplished in recent years, which indicates the bright future in meeting the high-quality lighting with low-cost of production. Primary challenges still remain in terms of limited efficiency, operating lifetime, and inherent toxicity of lead.

Table 1 shows the typical WLED devices performance in recent years. Among them, the research of SEL-WLEDs based on heterophase α/δ-CsPbI$_3$ have already basically met the threshold values of the brightness ($\sim 10^3$–$10^4$ cd m$^{-2}$) and EQE ($\sim 6\%$) for lighting requirements, presenting great development potential. It is worth noting that the reported best operation lifetimes of perovskites-based WLEDs are only in the range of several hours, which is still far from meeting the commercial standard. Therefore, it will further prolong the operation stability of WLED device considerably through the design of the light emitting layer, device structure optimization, and suitable packaging engineering. For instance, the trap state can be passivated by introducing defect-passivating additives such as sulfonate derivatives, phosphine oxide derivatives, and acetate derivatives that have a strong binding effect with perovskites to reduce non-radiative recombination center, which can successfully enhance the operational stability of WLEDs. In addition, incorporating a small number of cations such as manganese, strontium, and potassium can also enhance crystal structural stability and suppress ion migration of perovskites for efficient and stable light emissions.

Inspired by the history of OLEDs and QLED, the development of perovskites-based WLED application is relatively new and there is a great opportunity to improve operational stability. In addition, lead is known to be a toxic element whose potential release is quite harmful to both the environment and humans. Therefore, the toxicity of lead halide perovskites will hinder their application in the growing WLED market and other electronic devices. Alternatively, Cu-based perovskites with broadband emission can effectively avoid the toxicity of lead, which has great potential to realize lead-free WLEDs. However, the Cu-based perovskites are usually difficult to prepare high-performance WLEDs due to the large bandgaps and large effective mass of carriers, leading to the poor charge injection and transport in devices. Therefore, when using these lead-free halide materials in SEL-WLEDs, some strategies are needed to improve carrier injection and exciton recombination. The incorporation of organic or inorganic additive such as conjugated polymers, fluorinated molecules, and metal halides, which can modify the grain boundaries of perovskites, may effectively improve the carrier transport properties and recombination efficiency, achieving high-performance lead-free perovskite WLEDs. With in-depth research on light-emitting materials and device structures, we can expect more advanced performance of WLEDs based on metal halide perovskites to meet the commercialization of high-quality lighting applications.
Table 1. Summary of the device performances of the WLEDs.

| Year | Emitter materials | $L_{\text{max}}$ (cd m$^{-2}$) | EQE (%) | Lifetime | CC | CRI (%) | CCT (K) | Reference |
|------|------------------|-----------------|--------|--------|-----|--------|--------|----------|
| 2021 | Sky-blue perovskite/CsPbBr$_{12}$ | ~2000 | 12.2 | 16.1 min | (0.33, 0.33) | NA | 3750–7500 | [77] |
| 2018 | PA$_{2}$CsPbBr$_{1}$/CsPb(BrCl)$_{3}$ | NA | 0.22 | 150 s | (0.32, 0.32) | NA | NA | [81] |
| 2019 | MAPb(Br$_{1-x}$I$_{x}$)$_{3}$/NF | ~98 | NA | NA | (0.27, 0.35) | NA | NA | [84] |
| 2020 | CsPbA-2/SPR | ~80 | 1.3 | NA | (0.33, 0.35) | NA | 5580–8217 | [85] |
| 2021 | PEACl:CsPbBr$_{3}$/Ir(BT)$_2$(acac) | 707 | 10.81 | 155 s | NA | NA | NA | [86] |
| 2017 | CsPbBr$_{1.5}$I$_{1.5}$/HFSO | ∼1200 | ~0.5 | NA | (0.28, 0.33) | NA | NA | [89] |
| 2019 | MAPb(Br$_{0.6}$Cl$_{0.4}$)$_{3}$/Compound 2 | NA | 0.002 | NA | (0.30, 0.49) | 27.5 | NA | [92] |
| 2020 | MAPb(Br$_{0.6}$Cl$_{0.4}$)$_{3}$/6G | <0.2 | <0.01 | NA | (0.30, 0.30) | 92 | 3000–7500 | [90] |
| 2018 | Cs$_3$(Ag$_{0.6}$Na$_{0.4}$)InCl$_6$ | ~50 | NA | 10 min | NA | NA | NA | [98] |
| 2020 | CsPbCl$_{3}$/Sm | 938 | 1.2 | NA | (0.32, 0.31) | 93 | NA | [100] |
| 2018 | Cs$_2$AgIn$_{0.8}$Bi$_{1.2}$Cl$_6$ | 158 | 0.08 | 48.53 min | (0.32, 0.32) | 94.5 | 6432 | [99] |
| 2020 | (Cs$_6$H$_5$C$_2$H$_4$NH)$_2$PbCl$_2$Br$_2$ | ~70 | NA | NA | (0.22, 0.32) | 14 054 | NA | [101] |
| 2021 | CsPbBr$_{1-x}$Cl$_x$/CsPb(Br$_{1-x'}$I$_{x'}$)$_{3}$/CsPb(Br$_{1-x''}$I$_{x''}$)$_{3}$ | 30 | 0.008 | NA | (0.33, 0.33) | NA | NA | [103] |
| 2021 | CsPbBr$_{1-x}$Cl$_x$/CsPb(I$_{1-x'}$Br$_{x'}$)$_{3}$/CsPbBr$_{1-x''}$I$_{x''}$ | ~50 | ~0.02 | NA | (0.33, 0.34) | NA | NA | [102] |
| 2021 | CsPbCl$_{3}$/CQDs | 12 200 | 6.5 | NA | (0.31, 0.34) | >85 | 3000–6000 | [38] |
| 2020 | CsCu$_2$I$_6$/Cs$_3$Cu$_3$I$_5$ | 352.3 | 0.053 | NA | (0.33, 0.35) | 94 | NA | [108] |
| 2021 | CsCu$_2$I$_6$/Cs$_3$Cu$_3$I$_5$ | 145 | 0.15 | 238.5 min | (0.32, 0.33) | 91.6 | NA | [109] |
| 2021 | CsCu$_2$I$_6$/Cs$_3$Cu$_3$I$_5$ | 1570 | 3.1 | NA | (0.44, 0.53) | NA | NA | [110] |

Data availability statement

No new data were created or analysed in this study.

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