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Inorganic perovskite composites for light emitting diodes

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ABSTRACT

In recent years, there has been significant interest in all-inorganic perovskite materials due to their remarkable qualities, which include extended stability, adjustable light absorption spectra, suitable energy bandgap, extensive carrier diffusion length, and elevated photoluminescence quantum yield (PLQY). These extraordinary optical characteristics and stability make them highly promising contenders for optoelectronic devices. This paper explores inorganic perovskite composites and their applications in the development of light-emitting diodes (LEDs). Firstly, the fundamental concepts of all-inorganic PeLEDs have been explicated. Subsequently, the introduction of inorganic perovskite composites PeLEDs has been presented. Particularly, we have comprehensively provide a review the synthesis methods employed for all-inorganic nanocrystals (NC) , including hot-injection, ultrasonication, room-temperature and vapor deposition techniques. Furthermore, we have outlined various strategies aimed at enhancing the efficiency of all-inorganic composite PeLEDs.

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1. Introduction

Light-emitting diodes (LEDs) have become increasingly vital in our daily lives since their introduction in the 1960s [1-4]. They have the potential to replace traditional light sources in lamps and displays due to their superior energy efficiency, longer lifetime, versatility, and other advantages. Lately, the revival of metal halide perovskites, with a common formula ABX_3 (A representing a monovalent organic cation like formamidinium (FA), methylammonium (MA), or an inorganic cation B, Cs⁺ representing a metallic cation like Sn²⁺ or Pb²⁺, and X representing a halide anion such as Cl⁻, Br⁻, or I⁻), has led to the emergence of a fresh line of LEDs [5].

The appealing optical and electrical characteristics of organic-inorganic perovskites have garnered significant interest [6]. In particular, all-inorganic perovskite materials have captivated researchers due to their exceptional carrier mobility, efficient radiation recombination, excellent color purity, and adjustable bandgap [2]. These materials hold great promise as potential

candidates for the development of next-generation LED technology [7].

All-inorganic perovskite compositions, such as CsPbBr₃, exhibit superior thermal stability when compared to materials featuring a hybrid mix of organic and inorganic perovskite. The inorganic perovskites, such as CsPbX₃ (with X = Cl, Br, and I), as well as hybrid halide systems like Cl/B and Cl/Br, have been found to possess enhanced thermal stability [8-10]. Furthermore, the remarkable properties of all-inorganic perovskites, such as their ability to emit narrowband light with a full-width at half-maximum (FWHM) < 20 nm, impressive external quantum efficiency (EQE), current efficiency (CE), high photoluminescence quantum yield (PLQY), efficient carrier diffusion, low rates of nonradiative recombination, and wide range of tunable wavelengths 400 to 700 nm have generated significant enthusiasm for the advancement of LEDs [11-13].

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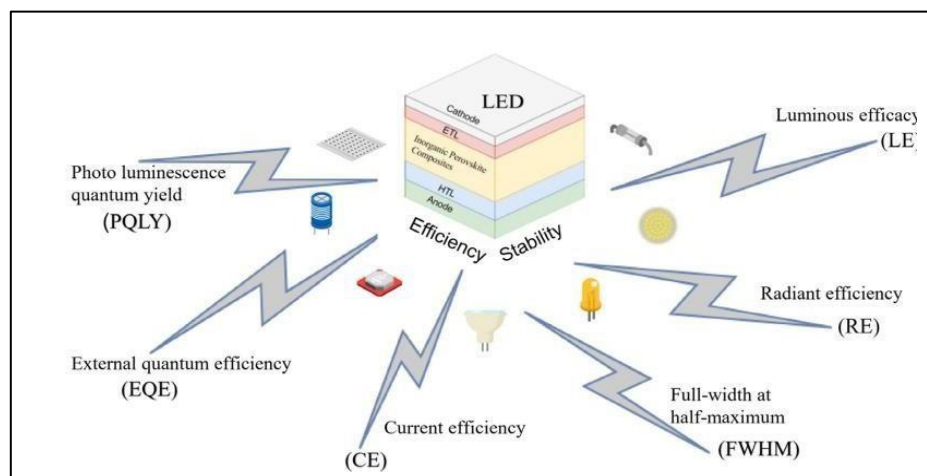


Fig.1. Remarkable properties of perovskites-LED

Nevertheless, the photoluminescence of the inorganic perovskite CsPbBr₃ was relatively feeble, as observed in large single crystals [14, 15]. However, by incorporating nanocrystals of CsPbBr₃ into optically inactive Cs₄PbBr₆ [16], the efficiency of light emission could be significantly improved. This enhancement can be primarily attributed to surface passivation, the spatial confinement of CsPbBr₃ nanocrystals and interfacial emissive states [17].

The improved efficiency of radiative recombination when subjected to optical excitation can potentially result in an enhanced electroluminescence effect when incorporated into LED devices [18]. Apart from the recombination of charge carriers, the behavior of charge transport behaves is also essential in influencing the efficiency of LEDs. Previous studies have demonstrated that optimizing the CsPbBr₃ | Cs₄PbBr₆ composite perovskite layer can effectively address the issue of inefficient charge carrier injection by adjusting the thickness and ratio of the layer [19, 20].

The evaluation of devices utilizing pure and composite perovskites indicated that the incorporation of composites could serve as a viable strategy to enhance the luminosity, efficacy, and durability of perovskite-based LEDs [18].

Inorganic perovskite composites are being recognized as a hopeful substance for LEDs because of their distinctive characteristics, including high color purity, tunable bandgap, excellent charge transport properties, and high PQ efficiency [21]. This paper presents the use of inorganic perovskite composites for LED applications, highlighting their potential for achieving high efficiency and stability. This study showcases techniques of

synthesis inorganic perovskite composites, providing a comprehensive understanding of their optoelectronic properties and device performance. Furthermore, the paper addresses the stability challenges associated with inorganic perovskite composites, proposing innovative strategies to enhance their long-term performance and reliability.

2. Perovskite-Based LED

The EQE serves as a measure to estimate the efficiency of LEDs. The determination of the LED's efficiency is achieved through the calculation of the ratio between the number of photons emitted by the LED and the number of injected electrons. Eq (1) [22, 23] can be used to express the EQE.

$$\text{EQE} = (\text{Number of emitted photons}) / (\text{Number of the supplied electrons}) = \gamma \times \chi \times \eta_{\text{PL}} \times \eta_{\text{oc}} \quad (\text{Eq. 1})$$

Achieving high-performance PeLEDs requires the creation of perovskite-emitting layers that exhibit a high PLQY. The PLQY is impacted by multiple variables, such as the proportion of excitons with spin-allowed optical transitions (χ) and the percentage of injected electrons that generate excitons (γ) and the out-coupling efficiency (η_{PL}). It's important to mention that the effectiveness of LEDs, as measured by EQE, is typically directly proportional to PLQY of layered perovskite. Therefore, enhancing the PLQY of perovskite-emitting layers is crucial for achieving efficient and high-performance PeLEDs [23].

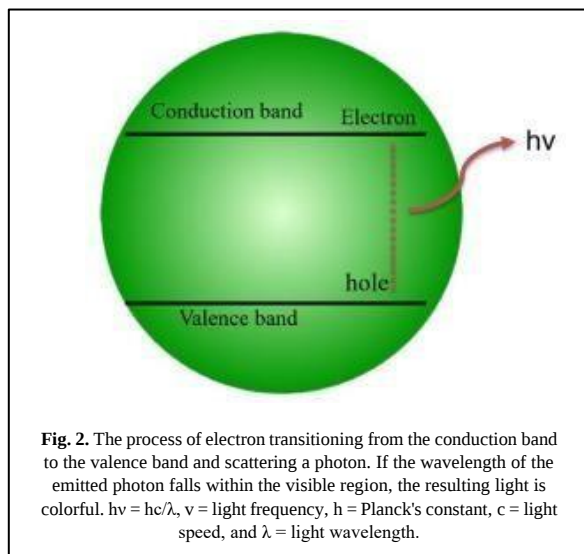
PLQY is the proportion of emitted photons to absorbed ones [2]. For a LED, wall plug efficiency (WPE) or radiant efficiency (RE) refers to the efficiency of converting electrical energy to optical energy and can be defined as:

$$\text{RE or WPE} = \text{EQE} \times \text{LE} = \eta_{\text{injection}} \times \text{IQE} \times \eta_{\text{extraction}} \times L / (I \times V) \quad (\text{Eq. 2})$$

In this context, the EQE is determined by comparing the number of emitted photons to the quantity of electrons injected into the device. The ratio of electrons injected into the active emitter layer to the total number of electrons flowing through the device is denoted as $\eta_{\text{injection}}$. The internal quantum efficiency (IQE) represents the percentage of electron-hole recombination events in the emitter layer that result in the emission of light, as indicated by references [24]. On the other hand, $\eta_{\text{extraction}}$ refers to the percentage of photons produced in the emitter layer that effectively exits the device [25]. Finally, the luminous efficacy (LE) is the proportion of the emitted light to the injected energy provided by the source [26]. The summary of the notable properties of perovskites can be seen in Figure 1. LEDs have been developed with different wavelengths, including infrared, visible, and ultraviolet (UV).

Table 1.
Relationship between light energy, light wavelength, and LED color.

Light color		λ (nm)	Energy (eV)
Violet		400-450	3.1-4.4
Blue		450-500	2.5-3.7
Green		500-750	1.9-4
Yellow		570-590	2.1-2.2
Orange		610-760	2-2.1
Red		610-760	1.6-2.0



LEDs have been developed with different wavelengths, including infrared, visible, and ultraviolet (UV). These LEDs can emit light at various levels, categorized as low, intermediate, or high light emission. When an electric current flows through an LED, it causes the semiconductor device to emit light. The release of photons in the form of energy is a result of the recombination of holes and electrons within the semiconductor material, leading to the emission of light (as shown in Figure 2). The emitted light color from LEDs is directly related to the energy of the photons released, which is determined by the semiconductor's band gap. This information is summarized in Table 1.

The high values of luminescence quantum efficiency serve as evidence for the successful conversion of electrical energy into light, leading to reduced energy consumption and improved performance of LEDs. Perovskites play a crucial role in photoelectric applications due to their exceptional efficiency values. By modifying the structure and chemical composition of perovskites, it becomes feasible to adjust their band gap, thereby enabling control over the emitted wavelength. Consequently, the colors emitted in LEDs can be finely tuned [27].

3. Properties and Types of inorganic Perovskite LED

Inorganic perovskites exhibit several unique properties that make them attractive for various applications, including LEDs. Some of these properties are: Optoelectronic Properties, bandgap Tunability, high Charge Carrier Mobility, stability. Inorganic perovskites have excellent light-absorbing and emitting properties. They possess a high absorption coefficient, enabling efficient light absorption even with thin layers of the material. The composition of the elements can be altered to adjust the bandgap of inorganic perovskites. This allows the emission wavelength of the material to be precisely controlled, making them suitable for different applications, including LEDs with various colors. They typically exhibit higher charge carrier mobility compared to organic materials. This leads to improved electrical conductivity and device performance. They are often more stable than their organic counterparts, especially against moisture and heat. This enhanced stability is desirable for long-term device operation and reliability. These LEDs have gained attention due to their potential for high efficiency and stability. In the following sections, the most important types of inorganic perovskites- LED have been discussed.

3.1 Three-dimensional bulk structure

Perovskites are a group of compounds characterized by their three-dimensional structural framework, denoted by the chemical formula AMX_3 [28]. The A-site within this framework can be occupied by either Inorganic metal cations with a single valence such as Rb^+ or Cs^+ , or organic cations like MA^+ or FA^+ . On the other hand, the M-site is typically filled by divalent metal cations, predominantly Pb^{2+} , however, it is also capable of accommodating various other cations. such as Sn^{2+} , Eu^{2+} , Cu^{2+} , Ni^{2+} , and so on. X represents a halide anion, which can be either Cl^- , Br^- , or I^- . Within the perovskite crystal structure, the M cation is positioned at the cube center, whereas the X anions and A cations are situated at face centers and the vertexes of the cube, respectively [29, 30].

The PbX_6 configuration creates regular octahedra through the central M cation (e.g., Pb^{2+}) and the six halide anions that surround it [31]. This arrangement creates an ideal 3D cubic phase perovskite structure) Figure 3(, where the PbX_6 octahedra share corners in a continuous array. However, if the PbX_6 octahedra are not in perfect cubic alignment, the perovskites adopt a less symmetrical orthorhombic form, as illustrated in Figure 3.

Perovskites exhibit further categorization into two types: organic/inorganic materials, like $MAPbX_3$ and $FAPbX_3$, and all-inorganic perovskites such as $CsPbX_3$ [32]. The initial demonstration of a PeLED, pioneered by Tan et al. [33], The experiment utilized an infrared gadget that produced light at 750 nm through electroluminescence (EL). This gadget was constructed using 3D bulk $MAPbI_{3-x}Cl_x$ as the emitting substance. The PeLED demonstrated a peak EQE of 0.76% and an infrared radiance of $13 \text{ W sr}^{-1} \text{ m}^{-2}$. Notably, in this LED configuration, the perovskite material exists in the form of a bulk crystal.

The structure of perovskite crystals is composed of interconnected networks of metal cations, halide anions, and various other elements [34]. Typically, these three-dimensional perovskite LEDs showcase remarkable optoelectronic characteristics, such as exceptional quantum efficiency and color purity. They have shown great potential in terms of brightness and efficiency. Nevertheless, a significant hurdle faced by 3D perovskites is their long-term stability, particularly when exposed to moisture [35].

3.2. Inorganic perovskite nanocrystals

Another form of inorganic Perovskite LED employs perovskite nanocrystals or quantum dots (QDs). These nanoscale crystals possess a perovskite structure and display optical properties that vary with their size. Consequently, the color produced by these nanoparticles' emission can be adjusted by manipulating their size [36, 37].

Perovskite QD light-emitting diodes (LEDs) present several benefits, including superior color purity, narrow emission line widths, and the ability to be processed in solution [38]. These LEDs have demonstrated potential in achieving both efficient and stable light emission. Furthermore, the nanocrystal structure of perovskite QDs offers enhanced stability when compared to three-dimensional perovskites [39].

Inorganic nanocrystals based on 0D perovskites, specifically colloidal $CsPbX_3$ (where $X = I, Br, \text{ and } Cl$), have appeared as a promising group of materials for light emission. These nanocrystals are synthesized by a reaction between cesium carbonate and oleic acid in a metal halide solution. Notably, Loredana Protesescu et al. [40] were the first to successfully develop the $CsPbX_3$ structure, which exhibits highly adjustable optical properties and a high PLQY. Colloidal perovskite $CsPbX_3$ NCs have adjustable bandgap energies that cover the visible spectrum and emit bright light. The properties of these NCs were observed in toluene under a UV lamp, with different stimulation wavelengths and time-resolved PL decays

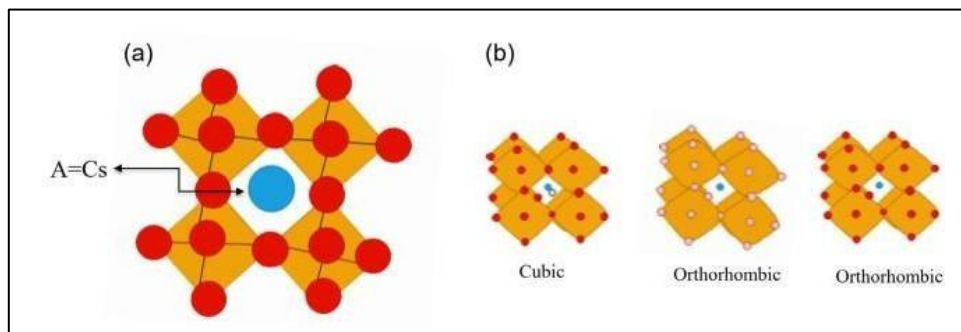


Fig. 3. (a) AMX₃ (A=Cs) inorganic perovskite, (b) Cubic (MAPbBr₃), Orthorhombic (CsPbI₃, CsPbBr₃)

shown in Figure 4. It is important to mention that the smaller-sized nanocrystals demonstrate an increased exciton binding energy, making the photoluminescence emission more likely to occur through exciton recombination rather than bimolecular recombination of free carriers [41].

The emission wavelength of all-inorganic CsPbX₃ QDs has been adjusted within the range of 410 to 700 nm [42]. Additionally, impressive photoluminescence quantum yields (PLQYs) ranging

from 50% to 90% have been attained. By manipulating the size of perovskite NCs, the emission color can also be regulated through the utilization of the quantum confinement effect. This effect is accompanied by an improved PLQY, which is likely due to the significant exciton binding energy and well-maintained confinement of charge carriers [43].

Table 2. Summary of inorganic perovskite NCs in recent years

Perovskite	EL (nm)	EQEmax (%)	CE ^{max} (cd A ⁻¹)	Lmax (cd m ⁻²)	PLQY (%)	Year	Ref.
CsPbBr ₃	520	1.3	5.2	54	-	2018	[44]
CsPbBr ₃	-	1.70	5.57	1562	-	2018	[45]
δ-CsPbI ₃	682	8.65	-	210	-	2018	[46]
CsPbBr ₃	515	11.6	45.5	55800	61	2018	[50]
CsPbI ₃	692	8.2	0.8	827	-	2019	[51]
CsPbBr	520	8.2	25.7	500	up to 87	2019	[47]
CsPbBr	517	9.7	31.7	2270	-	2019	[52]
CsPbI ₃	671	10.3	-	823	84-89	2019	[53]
CsPbBr ₃	512	12	39.3	7657	38	2019	[54]
CsPbI ₃	689	1.25	-	468	95	2020	[55]
CsPbI ₃	693	10.6	-	981	51	2020	[56]
CsPbI ₃	630	6.4	-	-	-	2021	[57]
CsPbBr _{1-x} I _{3x}	659	8.94	-	2859	15.3	2021	[58]
CsPbBr ₃	522	16.45	-	112 824	-	2021	[59]
CsPbBr ₃	-	19.3	-	1946	-	2021	[60]
CsPbX ₃	-	21.4	-	2000	60.0-87.0	2021	[61]
CsPbI ₃	640	23	-	200	-	2021	[62]
CsPbI ₃	682	8.15	-	584	72.9- 94.3	2022	[63]
CsPbBr ₃	534	11.19	51.0	20388	-	2022	[64]
CsPbI ₃	695	13.8	10.8	7039	73.0 - 59.2	2022	[65]
CsPbI ₃	514	20.79	65.68	584	96.02%	2023	[48]
CsPbI ₃	-	20.65	-	3861	-	2023	[66]
CsPbI ₃	688	22.8	-	15204	93	2023	[49]
CsPbI ₃	664	23	-	-	90	2023	[67]

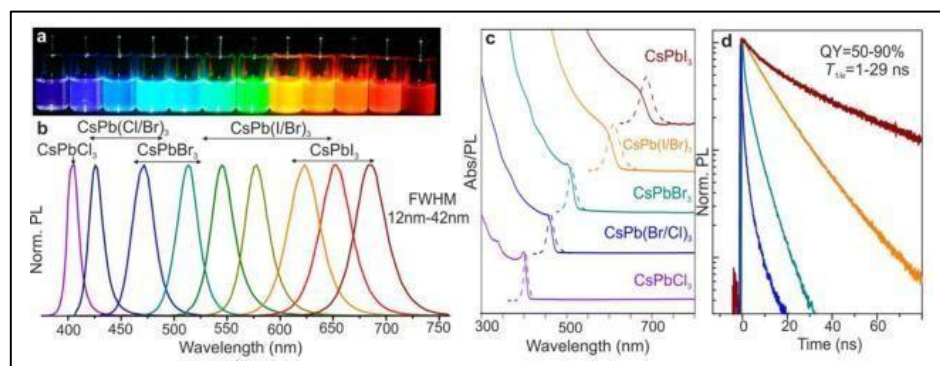


Fig. 4. (a) observation of bright emission in toluene colloidal solutions under a UV lamp ($\lambda = 365$ nm); (b) presentation of representative photoluminescence (PL) spectra with excitation at 400 nm (except for CsPbCl₃ samples, which are excited at 350 nm); (c) display of usual PL spectra and optical absorption; and (d) examination of time-dependent PL decay rates for each sample, excluding CsPbCl₃ [40].

In 2018, Naresh Kumar et al. [44] introduced a novel approach to enhance the efficiency of PeLEDs. They utilized high-quality green emissive colloidal CsPbBr₃ NCs with an average size of 11 ± 0.7 nm. The steady-state photoluminescence (PL) and EL, with FWHM fell within the interval of 20 ± 0.5 nm, indicating a remarkable feature of high color purity in these materials, making them suitable for achieving a wider color gamut. To further optimize the performance of the CsPbBr₃ NCs, the researchers treated a 4-layer film of these nanocrystals with MeOAc. This treatment resulted in a maximum luminance efficiency of approximately 5.2 cd/A, with a mean efficiency of 2.32 cd/A across six devices. Additionally, the electroluminescent quantum efficiency (EQE) reached approximately 1.3% at 6.5 V.

In their study, Lin Wang et al. [45] have founded a simple yet effective approach called solvent-assisted surface engineering to enhance the quality of nanocrystal thin films for LED application. By employing solvents of varying polarities, they successfully obtained assemblies of CsPbBr₃ nanocrystals. The efficacy of their strategy was further confirmed through the successful production of CsPbBr₃ LEDs, which exhibited exceptional efficiency. As a result, a significant 34-fold increase in EQE (reaching up to 1.70%) was accomplished in CsPbBr₃ LEDs fabricated using solution processing. Furthermore, Boning Han et al. [46] have reported a promising approach to manipulate phases in CsPbI₃. This involves introducing a long-chain cation such as naphthalene-1-yl ethanamine (NEA) to create stable and high-performing red LEDs based on CsPbI₃. The study showcases an impressive EQE of 8.65% for the distinct red light emission around ≈ 682 nm, marking the highest value reported for Cs-based red PeLEDs up to now. Additionally, the PeLEDs show exceptional stability, with the EQE stays maintained at 90% following 3 months of storage [46]. In another research, researchers successfully synthesized monodisperse CsPbBr₃ nanorods with adjustable the ratio of the span and a pristine surface by converting Cs₄PbBr₆ nanocrystals at the interface. The process of formation was thoroughly investigated. These nanorods exhibited remarkable photophysical properties, such as PLQY reaching 87% and a photoluminescence lifetime of 44 ns. Analysis using ultraviolet photoelectron spectroscopy revealed that CsPbBr₃ nanorods have an elevated valence band peak than nanocubes, suggesting a more effective hole injection rate beneficial for LED applications. To demonstrate the potential of CsPbBr₃ nanorods, a prototype LED was constructed utilizing them as a green light source. Remarkably, the LEDs achieved the EQE of over 8.2% without the need for laborious cleaning steps. This performance is among the best reported for LEDs based on CsPbBr₃ nanocrystals thus far [47].

The effective reduction of defect formation energy in PNCs is achieved by introducing metal acetate salts. The resulting PNCs exhibit exceptional characteristics such as near-perfect photoluminescence quantum yield, remarkable stability, luminance of 1175 cd m⁻², CE of 65.48 cd A⁻¹, EQE of 20.79%, and a wavelength of 514 nm in optimized PNC LEDs. These desirable properties are attributed to the presence of a Cd²⁺ passivator and DDA ligand [48].

In a separate study conducted by Po Lu et al. [49], they utilized a typical triphenylphosphine (TPP) ligand to investigate its impact on the surface of CsPbI₃ perovskites NCs. Additionally, the incorporation of extra benzene rings in the form of 2-(Diphenylphosphino)-biphenyl (DPB) enhanced the delocalized characteristics of the PNC surface and significantly improved the charge transport within the PNCs. Consequently, the DPB passivated CsPbI₃ NCs demonstrated top-emitting LEDs with remarkable performance metrics, including an exceptionally low-efficiency roll-off of 2.6% at a current density of 500 mA cm⁻² and a peak of 22.8%, a maximum luminance of 15,204 cd m⁻². Table 2 summarizes the performance of PeLEDs using various types of perovskite nanocrystals and all-inorganic perovskite materials.

4. Inorganic composite Perovskite LED

Inorganic composite Perovskite LEDs represent a cutting-edge technology in the field of lighting and display applications. These LEDs utilize a unique combination of inorganic materials with Perovskite structures to achieve enhanced performance and efficiency. Perovskite materials, known for their excellent optoelectronic properties, have attracted a lot of focus lately because of their exceptional light emission efficiency, tunable bandgap, and cost-effective fabrication processes. By incorporating these materials into inorganic composite structures, researchers aim to overcome some of the challenges associated with pure organic or inorganic LEDs. One of the key advantages of inorganic composite Perovskite LEDs is their improved stability and lifespan compared to traditional organic LEDs. The inorganic components help enhance the material's resistance to environmental factors such as moisture and oxygen, leading to longer-lasting and more reliable devices.

4.1 Synthesis methods of inorganic-based Perovskite LED

The synthesis techniques for inorganic-based composite Perovskite LED involve combining various inorganic materials to create a highly efficient and versatile light-emitting device. In the past few years, there has been considerable interest in the composite Perovskite LEDs because of their promise for application in advanced lighting and display technologies. Figure 5 illustrates a summary of the synthesis methods for these perovskites.

4.1.1 Hot-injection

One approach that is frequently used for manufacturing perovskite materials is the hot-injection method. By using this technique, different halide compositions, sizes, and forms can be created. The NCs grow extremely quickly, and while reaction time is mostly responsible for determining average size, it is more practical to reduce reaction temperature than growth time. By adjusting the precursor salt ratios, mixed-halide can be produced easily. The band gap and emissions can be adjusted by halide and size modulations [68]. The schematic of the method hot-injection is shown in Figure 6. Novel composites of CsPbX₃@mica with excellent thermal and light radiation stability have been successfully

created through the hot injection strategy by Yanxia Yu et al. [69]. Jinyang Zhu et al. [70] demonstrated perovskite quantum dots (QDs) stability is often compromised, along with the rapid Reaction involving the exchange of anions that occurs among various halide ions. To address this issue, they incorporated red light-emitting CsPb(Br/I)3 QDs and green light-emitting CsPbBr3 QDs into carboxyl-mediated polymethyl methacrylate (PMMA) using the hot-injection method. Consequently, the resulting PMMA/perovskite composites not only maintain a high level of luminescence but also demonstrate excellent resistance to water. Additionally, this treatment effectively prevents the exchange of anions between different halide QD particles when in a solid state. All-inorganic perovskite NCs have gained attention as a viable substitute for hybrid organic-inorganic perovskite in optoelectronic applications due to their distinct electrical and optical characteristics, as well as enhanced chemical stability. The introduction of metal or metal nanoparticles into these all-inorganic perovskite NCs through doping allows for the modification or adjustment of their optical characteristics. In another paper, Ag-CsPbBr3 nanocrystals were successfully synthesized using the hot injection method [71].

4.1.2 Room temperature synthesis

The method of creating perovskite crystals at room temperature, usually between 20 and 25 degrees Celsius, is referred to as room-temperature synthesis of perovskite materials. Numerous benefits, including easier reaction conditions and less consumption of energy, come with this method. There are alternate methods that enable room-temperature synthesis in addition to the conventional hot-injection technique that has already been described for perovskite synthesis. Although polar solvents are frequently employed to prepare precursor solutions by injection method, the solvents utilized in room-temperature synthesis methods are typically low polarity or non-polar solvents, such as toluene or chlorobenzene. It's crucial to keep the area dry while mixing the precursor solutions using either method [16]. Li et al. [72] were the pioneers in showcasing the application of a room temperature supersaturated recrystallization technique for creating CsPbX3 nanocrystals.

Mo Qionghu et al. [73] showed the PLQY of CsPbBr3@SiO2 quantum dots (QDs) achieved a remarkable value of 71.6%, whereas it was only 46% for pure CsPbBr3 QDs. Furthermore, the CsPbBr3@SiO2 QDs displayed exceptional stability when subjected to heat. Additionally, an improved two-photon pumped amplified spontaneous emission (ASE) was successfully demonstrated under normal air conditions. Comparatively, the ASE threshold of CsPbBr3@SiO2 QDs films was reduced by 70 $\mu\text{J}/\text{cm}^2$ when compared to those employing CsPbBr3 QDs, utilizing a laser excitation powered by two-photon. As depicted in the schematic presented in Figure 1a, it can be observed the synthetic process was carried out in two steps by Ludan Zhu et al [74]. Firstly, CsPbBr3 nanocrystals were formed, followed by size control through the addition of APTES. The Experimental section provides a comprehensive account of the process. Notably, the sol sample exhibited a vivid green color when exposed to ultraviolet light at a wavelength of 395 nm (Figure 7c). Furthermore, the utilization of an ultraviolet polymerization method enables the preparation of CsPbBr3 nanocrystals/polyaryclic composites (Figure 7b,c).

4.1.3 Ultrasonication

In recent years, the preparation of NP has been documented through the utilization of direct ultrasonication technique, along with the solvent-free mechanical synthesis approach. These methods offer a straightforward, scalable, and efficient approach to synthesizing high-quality colloidal CsPbX3 (X = I, Br, and Cl) perovskite NCs. Notably, this synthesis approach allows for the control of the composition of halide ions and the precursor solutions thickness, all while eliminating the need for polar solvents and incorporating organic capping molecules [37]. Using high-frequency sound waves, usually ranging from 20 to 100 kHz, on a liquid medium that contains the precursor components is referred to

as ultrasonication. Ultrasonication can be employed in the production of perovskite materials to optimize reaction kinetics, regulate the dimensions and structure of the resulting perovskite nanoparticles, and increase precursor material dispersion. Colloidal inorganic perovskite nanocrystals, specifically CsPbBr3, exhibit remarkable optoelectronic properties and enhanced stability compared to their hybrid counterparts.

Nevertheless, the efficiency of blue-emitting CsPbBr3 nanocrystals is still inferior to that of their green counterparts. Moreover, their production usually necessitates elevated temperatures and the use of an inert gas atmosphere. To address these challenges, Xiaoli Xu et al. [75] Suggest a top-down method for the effective production of blue-emitting CsPbBr3 quantum dots in a normal air environment. This technique includes breaking down CsPbBr3 nanoplatelets (NPLs) into spherical QDs through ultrasonication and treatment with hydrobromic acid (HBr). The produced CsPbBr3 QDs show outstanding uniformity, with an average size of around 5 nm, and emit blue light with a wavelength of 460 nm." Furthermore, they demonstrate a high PLQY of 53.2%. This work represents a significant advancement towards achieving the desired synthesis of perovskite NCs with efficient blue emission in air environment. Schematic this method demenstrated in figure 8.

4.1.4 Vapor deposition technique

Without using solvents, perovskites are made using solid-state techniques. This technique has the advantages of solvent-free, being reasonably straightforward, and economical [76, 77].

Using a vapor deposition technique, Ling Yet al. [78] created CsPbBr3/Cs4PbBr6 composites for improved photoluminescence (PL). Dual-source thermal evaporation offers an opportunity to tune the concentration of the component materials in a wide range, which is crucial for understanding the luminous behaviors in these composites. This process is superior to solution synthesis in this regard. Regardless of the ratio between the two phases, the composite films made using this technique have low pinhole-free morphology and surface roughness. Insufficient film coverage and high surface roughness are frequently caused by the low solubility of Cs precursors in solution synthesis (for example, CsBr in the polar solvent) [79, 80].

Efficiency variations can result from different synthesizing techniques. When comparing co evaporated composite thin films to solution-synthesized composite NCs, the PLQY of the second type shows a monotonic increase with increasing Cs4PbBr6 concentration. Due to the interfacial states between the two phases, which contribute to the concentration-dependent PLQY, these variances suggest that alternative preparation techniques can result in varied light emission behaviors. The CsPbBr3/Cs4PbBr6 grain boundaries likely generate a type of shallow trap states in this case that have not yet been seen in halide perovskites and are the cause of the PL increase seen. According to their findings, composites with purposefully created heterogeneous interfaces present new opportunities for researching and enhancing the material's properties and capabilities [78].

Large quantities of materials cannot be obtained by a simple and scalable precipitation synthesis approach. This is due to the fact that conventional methods, such as large organic group confinement crystallization or solvent-induced co precipitation, require several of processing, separation, and precise crystal thickness control [81].

4.1.5 Others

The adaptability of perovskite nanoparticles is demonstrated by the wide range of factors that can be altered to change their electronic and optical characteristics, such as chemical composition, structure, and particle size. By accurately controlling experimental parameters like the precursor duration, reaction temperature, concentration, and chemical composition, it is possible to tailor the morphology, optical and electronic properties of perovskite nanoparticles with ease. Another various methods have been demonstrated for synthesizing colloidal perovskite NPs, including

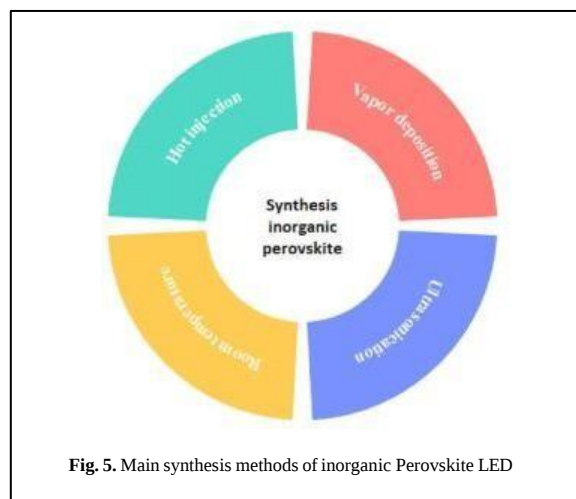


Fig. 5. Main synthesis methods of inorganic Perovskite LED

solvent-induced precipitation, ligand-assisted reprecipitation (LARP), emulsion processes, template-assisted synthesis, and novel synthesis [82, 83]. The dissatisfactory stability of lead halide perovskites, because of their delicate ionic crystal attributes and continually dynamic surface-coordinated states, has limited their applications. However, researchers have discovered that in-situ diphasic structure passivation, involving the application of identical chemical components to protect the perovskites (e.g., passivating CsPbBr₃ with Cs₄PbBr₆), can effectively improve their stabilities while maintaining their high-efficient luminescence properties. In a groundbreaking study, the researchers introduce a novel anion-cation reaction method to synthesize lead halide perovskite nanocrystals with a diphasic CsPbBr₃/Cs₄PbBr₆ structure. Additionally, they observe that the phase transformation between CsPbBr₃ and Cs₄PbBr₆ is temperature-dependent [84].

4.2. Properties inorganic-based composite perovskite LED

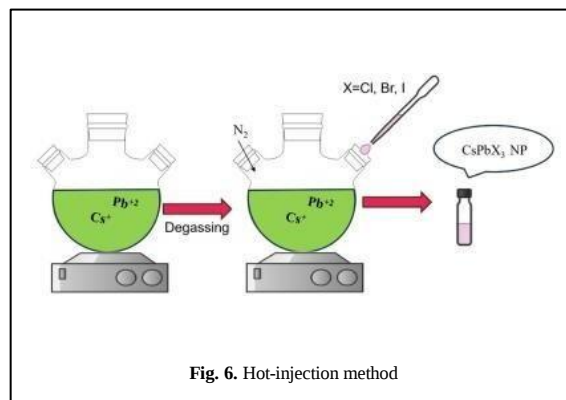
Researchers have suggested numerous solutions to enhance the stability of PeLEDs. Creating a core/shell grain configuration, where 3D bulk grains are enveloped by a shell layer like bulk organic cations, could be a more effective approach to enhance the longevity of PELEDs. This method involves encapsulating the grains with a protective shell, which effectively passivates the grain boundaries in all directions, thereby impeding ion migration [85].

Hu et al. [86] successfully synthesized CsPbBr₃-Cs₄PbBr₆ microcrystals, which exhibit a unique composite structure. These microcrystals demonstrate remarkable luminescent properties and exhibit good stability. Notably, the photoluminescence (PL) intensity of these microcrystals only decreases by 20% after being exposed to ambient conditions for a period of 2 months.

The Cs₄PbBr₆ phase was introduced by Akkerman et al. [87], who also incorporated nanocrystals of CsPbBr₃ within the photoactive Cs₄PbBr₆ to create a core-shell structure known as Cs₄PbBr₆/CsPbBr₃. In this structure, the electrons and holes generated in CsPbBr₃ are confined spatially by Cs₄PbBr₆, leading to a notable enhancement in the luminescence efficiency.

A study on the effects of CsPbBr₃ and Cs₄PbBr₆ on the improvement of PL in composites was undertaken by Ling Y and his colleagues [78]. They discovered that the composites' single light emitter was CsPbBr₃ by varying the material composition over a large range. Surprisingly, the addition of Cs₄PbBr₆ greatly increased the light emission efficiency in the composites despite the fact that it lacked PL characteristics.

As the quantity of CsPbBr₃ decreased in the films, the PL intensity showed an increase. By passivating grain boundaries with luminous shallow states, the inactive Cs₄PbBr₆, in the form of a host-guest system with CsPbBr₃ crystallites, was essential in improving the PL processes. By measuring the PLQY with an



integrating sphere, it was determined that the efficiency of PL improved as the concentration of Cs₄PbBr₆ in the composite thin films increased. Due to the poor sensitivity of XRD in detecting minute levels of luminous components, some materials, notably those dominated by Cs₄PbBr₆, may have been mistakenly identified as luminescent in the past. These findings give designers of composite perovskite materials important information for achieving the best possible light emission performance [87, 88].

In another study, high PLQY and narrow full-width at half-maximum (FWHM), green CsPbBr₃ perovskite nanocrystals are thought to be a potential luminous material for next-generation lighting and displays. Due to their varied electrical structure, zero dimensional Cs₄PbBr₆ perovskite nanocrystals have also been described and created. They are unable to exhibit excitonic and absorption emission in the visible spectrum because of their band gap width exceeding 3.2 eV. [87]. For the purpose of spreading the CsPbBr₃ nanocrystals and creating dual phase CsPbBr₃ - Cs₄PbBr₆ (DP-CPB) composites that emit a bright green light, Cs₄PbBr₆ can act as the framework [88]. The DP-CPB perovskite composites have good solid state luminous performance and the PLQY can reach 95% when compared to the CsPbBr₃ perovskite solid (PLQY 0.1%) [88].

Hongting Chen et al. [54] in their research found utilization of NaBr in CsPbBr₃ NPs green LEDs results in a significant improvement in their external quantum efficiency (EQEmax), reaching a remarkable value of 17.4%. This enhancement is evident when compared to the EQEmax of LEDs that do not incorporate NaBr, which stands at 12%. Moreover, even when considering a larger area of 108 mm², the NaBr-based CsPbBr₃ NPs LEDs maintain a commendable maximum EQE of 10.2%. Moreover, to leverage the combined benefits of PEA and PEO, CsPbBr₃ nanoparticles were synthesized in situ, resulting in an average grain size of 32.4 nm. The CsPbBr₃ nanoparticles LEDs fabricated this way achieved a peak EQE of 12%.

5. Strategies for improved properties of perovskites suitable for LED applications

In recent years, Perovskite LEDs have undergone accelerated development and currently, the EQEs of perovskite LEDs is more than 23%. Among the factors that limit the practical use of such devices are the low efficiency of blue-emitting devices, short device lifetimes, insufficient knowledge about how the electric field impacts mobile ions within perovskite materials, a reduction in external quantum efficiency under high current density, and challenges in reaching emission wavelengths beyond 800 nm. [5, 101-103].

Several strategies have been recommended to resolve these problems, such as utilizing stable perovskite materials, optimizing

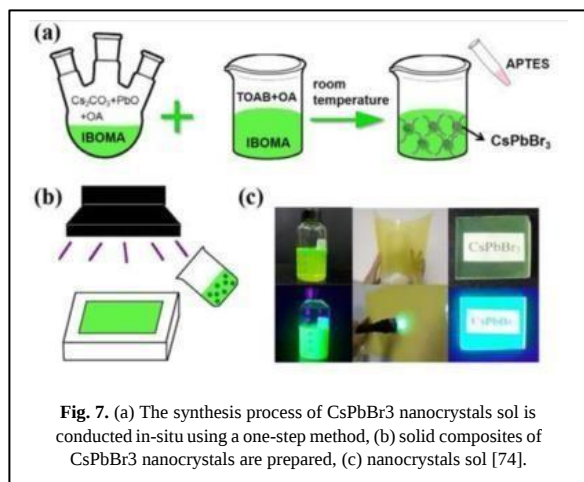


Fig. 7. (a) The synthesis process of CsPbBr_3 nanocrystals sol is conducted in-situ using a one-step method, (b) solid composites of CsPbBr_3 nanocrystals are prepared, (c) nanocrystals sol [74].

of device structures, and the introduction of new fabrication methods. Lessons learned from perovskite solar cells and organic LEDs can be applied to improve the stability and efficiency of perovskite LEDs as well [104, 105].

Perovskite LEDs possess the potential for use in a range of optoelectronic functions. They promise electroluminescent that is both cost-effective and high-performing. Perovskite LEDs with long-term stability and high efficiency will rapidly increase their practical applicability. Additional study is required to overcome the obstacles and enhance the stability and efficiency of perovskite LEDs [106]. In the ultraviolet portion of the electromagnetic spectrum, Perovskite materials commonly emit in the so-called UVA region 320 to 400nm — a range suitable for UV curing and other applications. UV curing is an extensively used, low-cost industrial technique for drying inks, adhesives, and coatings with intense UV light [107-109]. UV light-sensitive materials that contain photo initiator molecules, when exposed to UV light, activation energy is provided to start the reaction. As the chemical reaction initiates, the liquid rapidly solidifies by cross-linking with oligomer and monomer molecules. Although UV-emitting perovskite emitters have been presented, they have not yet been incorporated into LEDs [110]. Due to the necessity of UV radiation of a shorter wavelength, 200 to 280 nm UVC for such applications, the perovskite emitters devised so far are inadequate for applications like purifying drinking water, treating microorganisms, and creating disinfectant. The most significant applications for perovskite LEDs in the range of visible are augmented/virtual reality, high-brightness applications such as solid-state illumination and projection screens, and in display devices. Perovskite LEDs offer numerous benefits as display technology transitions to active-matrix organic LEDs (AMOLED) from liquid-crystal displays. Their easy processing allows seamless integration on thin-film transistor backplanes, enhancing display resolution, brightness, and color gamut. Perovskite emitters, known for superior color accuracy, have the potential to deliver higher resolution. Perovskite materials can be processed on flexible substrates and at low temperatures. Thus, they are ideal options for flexible displays. Particularly, display devices, such as AMOLED-based devices, are manufactured using lithography patterning, a widely used and industrially compatible technique. Lately, perovskite materials have shown compatibility with lithography patterning, enabling size reduction within the 1 to 10 μm range [111, 112].

To prevent the deterioration of perovskite films during liftoff, which often involves high-polarity solvents, researchers have shown that a dry-liftoff method using mechanical peeling of the photoresist material is efficient [111]. In solid-state lighting uses like projection displays, stage lights, indicators, automobiles, decoration, interior lighting and so on, high power efficiency is required. LEDs with a power efficiency ranging from 40 to 100 lm W⁻¹ are deemed

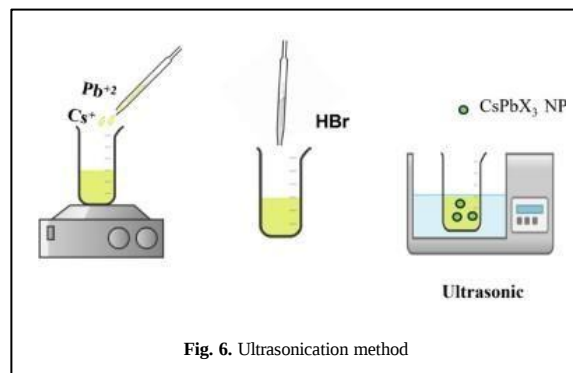


Fig. 6. Ultrasonication method

suitable for solid-state lighting, despite the absence of a clearly defined threshold. In 2017, the luminous efficacy of high-brightness, cutting-edge LEDs made from inorganic materials approached 160 lm W⁻¹ [113], and this increased to 200 lm W⁻¹. A higher lumens-per-watt ratio signifies better energy efficiency, which is the desired characteristic in LEDs. Perovskite LEDs have shown a luminous efficacy greater than 120 lm W⁻¹ [114]. Even though this is still far below the theoretical upper limit of an optimal white-light source (414 lm W⁻¹) [113], there is no basic restriction on the luminous efficacy of Perovskite LEDs.

Perovskite LEDs are also potential for NIR applications [115] including optical switches, telecommunication, and night vision. Here, the specific application is determined by the emission wavelength in the NIR range (700–2,500 nm), such as their use in optical wireless communication and vivo bio imaging, biomedical sensing. Perovskites-based LEDs continue to outshine their NCs counterpart. In 2015, for instance, green-emitting LEDs with the CE of 42.9 CdA⁻¹ were demonstrated using hybrid organic-inorganic perovskites [116]. Nonetheless, since their synthesis has been reported, inorganic (CsPbBr_3) perovskite NCs-based LEDs have demonstrated a remarkable performance boost.

Actually, the initial green-emitting CsPbBr_3 NC LED showcased a current efficiency of 0.43 CdA⁻¹ [117], and in less than a year, LEDs of this type have reached 13.3 CdA⁻¹ [118] in the same spectral range. Due to the expertise gained in the last twenty years in producing LEDs with solution-processed materials, performance is rapidly improving. In fact, the majority of the manufacturing methods utilized in organic (OLEDs) or II-VI and III-V semiconductor QD LEDs are entirely transferable, and a wide range of materials that are capable of being utilized as electrodes or charge transport layers are readily available today, thereby facilitating device optimization. Although there has been a swift enhancement in efficiency and luminance levels, the predominant findings are concentrated within the green spectrum ranging from 500 to 550 nm. In contrast, blue emitting perovskite NCs LEDs continue to trail in terms of efficiency and maximum luminance. In actuality, the stability and overall operational lifetime of perovskite NCs LEDs is a significant concern [117, 119].

Further investigation is needed to uncover the mechanisms of degradation that take place during LED operation, exposure to air, and storage. Previous studies have indicated that the use of mixed halides for color tuning leads to an unstable emission color within a few seconds of operation [119]. In 2014, Prieto and colleagues showcased the initial instance of perovskite nanocrystal LEDs

Composite	Method	Maximum luminance (Lmax)	luminous efficacy	PLQY	Main result	Ref
CsPbBr ₃ @ SiO ₂	Room temperature	-	-	71.6%	The PLQY of CsPbBr ₃ @SiO ₂ quantum dots (QDs) achieved a remarkable value of 71.6%, whereas it was only 46% for pure CsPbBr ₃ QDs.	[73]
Cs ₄ PbBr ₆ /CsPbBr ₃	Room-Temperature	-	-	48.78%	Synthesis of Cs ₄ PbBr ₆ /CsPbBr ₃ mixed crystals on a significant scale, resulting in a high PLQY of 48.78%. During environmental stability testing, it was observed that there was an approximate 20% decline when exposed to ambient conditions with 50% RH for a period of 2 months.	[86]
NaBr /CsPbBr ₃	synergistic	8353	-	17.4%	The utilization of NaBr in CsPbBr ₃ NPs green LEDs results in a significant improvement in their external quantum efficiency (EQEmax), reaching a remarkable value of 17.4%. This enhancement is evident when compared to the EQEmax of LEDs that do not incorporate NaBr, which stands at 12%.	[54]
CsPbBr ₃ @ SiO ₂	all-solid-state process (sintering after grinding)	-	-	15.5%	A straightforward green method was fine-tuned for synthesis, achieving ultra-high stability without the use of solvents. The intensity was sustained at 100% even after undergoing a 196-hour test under harsh double-85 conditions.	[77]
hierarchical CaF ₂ nanospheres loading CsPbX ₃	Hydrothermal/mixing	-	2.7 lm W ⁻¹	50, 82, and 66%	highly luminescent and stable CaF ₂ -PQDs nanocomposites	[89]
silica-coated CsPbBr ₃ QD	sol-gel	-	63.5 lm W ⁻¹	-	Enhanced material durability when exposed to light, heat, and environmental factors such as moisture and oxygen.	[90]
CsPbBr ₃ -Cs ₄ PbBr ₆	single atomic supersaturated recrystallization method	-	-	40.8%	The synthesis process is simple and requires minimal energy, making it suitable for large-scale production and practical applications.	[91]
CsPbX ₃ @mica	hot injection strategy	-	-	38.5%	high thermal stability and light radiation stability	[69]
CsPbX ₃ @α-Al ₂ O ₃	hot injection strategy /simple coating mechanism	-	86.58 lmW ⁻¹	58%	The composite material that was created could produce adjustable emissions ranging from 430 to 636 nm.	[92]
CsPbBr ₃ -Cs ₄ PbBr ₆	chemical solution route without inert gas protection	-	45 lmW ⁻¹	74%	The composite perovskite nanocrystals showed high quantum yield and distinct emission spectra, surpassing CsPbBr ₃ nanocrystals.	[93]
green CsPbBr ₃ @glass and red CsPbBr _{1.5} I _{1.5} @ glass composite	modulating perovskite concentration in glass and heat-treatment temperature	500 000 cd m ⁻²	-	94% and 78%	high PLQY, and excellent stability	[94]
Mn ²⁺ -doped Cs-Pb-Cl	chemical solution route	-	-	77.3%	Improving optical performance ultimately through suppressing the nonradiative transition of the exciton	[95]
CsPbBr ₃ @ Cs ₄ PbBr ₆	solid-state synthesis	-	-	>90%	Improving the overall stabilities of CsPbBr ₃ NCs against heat, moisture, and photoirradiation.	[96]
CsPbBr ₃ :Sr/PbBr(OH)/	high-temperature solid-phase method	-	86 lmW ⁻¹	75%	Strong resistance to heat and water, along with improved stability against the combined effects of blue light and water, or blue light and heat, was observed.	[97]
CsPbX ₃ /SBA-15	in situ growth strategy	-	183 lmW ⁻¹	-	Utilizing Mesoporous SBA-15 leads to diminished surface flaws in CsPbBr ₃ , which consequently lowers the non-radiative recombination rate of carriers, leading to a notable enhancement in luminescence duration.	[98]
CsPbBr ₃ / Cs ₄ PbBr ₆	conventional melting-quenching and heat-treatment technique	remained 80% of the original one	25 lmW ⁻¹	58%	Through refining the raw materials and adjusting the melting parameters, it was possible to achieve a PLQY exceeding 58% while enhancing thermal stability, moisture resistance, light stability, and water resistance.	[99]
CsPbBr ₃ PQD-SiO ₂ and CsPbBr _{1.2} I _{1.8} PQD-SiO ₂	solution route	-	35.4 lmW ⁻¹	-	highly enhanced thermal stability and water stability	[100]

emitters, free MAPbBr₃ NCs with a 20% quantum yield (QY) employed, and only low brightness and insufficient device performance reported. Zeng et al. [120] improved the brightness of perovskite NC-based LEDs through using bright CsPbBr₃ NCs (PLQY greater than 85%) as the emitters, which reaches 946 cd m⁻² at a voltage of 8.8 V. Due to the instability of iodine-based NC materials, it is more difficult to produce effective iodine-based perovskite NC LEDs than bromine-based perovskite NC LEDs. Initial research on CsPbI₃ NC LEDs was focused on their performance [117].

Using a cross-linking method based on trimethylaluminum (Al₂(CH₃)₆) vapor, the CsPbI₃ nanocrystal film was enhanced in capturing electron-hole pairs, leading to the development of red LEDs with a peak EQE of 5.7% [119]. Zhang et al. [121] Demonstrated that a simple treatment of CsPbI₃ nanocrystals with polyethylenimine effectively reduced surface defects, leading to a significant enhancement in electroluminescence efficiency to 7.25%. Following this, the researchers started to consider the device's stability. By entirely epitaxially growing PbS semiconductor on the CsPbI₃ NCs surface, Zhang et al. [122] Created a technique to enhance both the stability and optical characteristics of CsPbI₃ nanocrystals while preserving their semiconductor properties intact. By introducing PbS capping, the CsPbI₃ nanocrystal film transitioned from n-type to almost ambipolar behavior, enabling the fabrication of LEDs with p-i-n structures. These LEDs exhibited enhanced performance and longevity, along with an EQE of 11.8% [122]. Using SrCl₂ as a co-precursor while synthesizing CsPbI₃ NCs, Lu et al. developed a method to enhance both the EL and photoluminescence (PL) efficiencies in order to make LEDs more efficient and stable. As a consequence, NCs with Cl surface passivation and simultaneous Sr doping, were produced, and devices employing these emitters demonstrated improved stability and EQE of 13.5% [123].

Utilizing Zn-alloyed CsPbI₃ NCs to act as emitters allowed for a subsequent increase in the device's EQE to 15.1%. CsPb_{0.64}Zn_{0.33}I₃ is the element found in the most effective and stable NCs. In addition to NC LEDs based on pure Bromine or Iodine component NCs, mixed halogen perovskite NC devices have also been investigated. Mixed halogen NCs offer more choices on the emitting color, which is promising in high-purity color display [124].

Even while luminance and efficiency for green-emitting LEDs have significantly increased in just over a year, considerable work still has to be done to better understand the processes that cause NCs to degrade, create efficient red and blue LEDs, stabilize their color, and optimize the device architecture. While chemists all over the world are continuously manufacturing novel perovskite NCs, all these constitute significant obstacles [125].

The presence of lead in the crystal continues to fuel the exploration for new structures and compositions of perovskite NCs. The expectation is that the knowledge acquired in producing LEDs with solely inorganic or hybrid lead perovskite nanocrystals can eventually be applied to their environmentally friendly versions. The toxicity of lead-based perovskite remains a major hindrance to their practical use. Also, All-inorganic perovskites made of cesium and lead halides have shown great potential as semiconductor materials in various fields such as lighting, display technology, light detection, and lasers. However, the durability of lead halide perovskites poses a significant issue because of their delicate ionic crystal framework and the presence of surface-coordinated states that are highly dynamic. To address this issue, the CsPbBr₃@Cs₄PbBr₆ composite nanocrystals have been optimized to exhibit excellent light emission performance and enhanced stability against atmospheric conditions, moisture, and heating. Leimeng Xu et al [84]. were able to achieve a remarkable 135% enhancement in the NTSC color gamut by employing anion exchange. The resulting composite NCs, characterized by their exceptional emissive properties and enhanced stability, hold

significant promise for various optoelectronic applications in the future.

6. Conclusions

Inorganic perovskites possess alluring optical and electronic attributes, making them highly promising for optoelectronic uses and sparking considerable research attention. However, the unfavorable structural and chemical stability of these materials is the greatest hindrance to dependable device performance. Review of the research conducted on inorganic perovskite composites in an effort to improve their stability and efficacy as LED. The optical properties and efficacy of composites comprised of inorganic perovskite were discussed. According to the findings of recent studies, composites of inorganic perovskite can increase chemical stability. Additionally, the quantum efficiency and optical properties of LED based inorganic perovskites have been enhanced through doping and formation of composites. The proportion of composites' constituent materials, in the optical properties of LEDs are crucial, so it is recommended to use new, cost-effective, and scalable materials to obtain the best LED performance.

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