

## Advancing Perspectives on Large-Area Perovskite Luminescent Films

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Cite This: *Energy Fuels* 2024, 38, 17343–17354



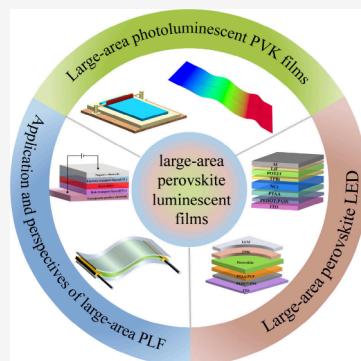
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**ABSTRACT:** The excellent photoelectric properties of perovskite materials are mainly attributed to their high optical absorption coefficients, high carrier mobility, long carrier lifetimes, and adjustable band gaps. The ability of these materials to be engineered into large-area films offers significant advantages for practical applications, particularly in the context of portable and wearable technologies. Their lightweight and flexible characteristics further enhance their suitability for a wide range of innovative uses, from consumer electronics to advanced display technologies. Given the promising potential of large-area perovskite luminescent films (PLF), it is crucial to understand both their underlying properties and the mechanisms driving their luminescence. Therefore, this paper primarily summarizes the luminescence mechanisms of large-area PLF, including electroluminescence, photoluminescence, and mechanoluminescence. It also explores several key fabrication methods in detail. Additionally, the paper highlights the potential applications of these luminescent films, particularly in lightweight, flexible, and wearable technologies, and discusses their prospects in practical applications. By analyzing the current state of research, this paper seeks to underscore the critical role that large-area PLF are poised to play in the future of optoelectronic devices.



### 1. INTRODUCTION

Under the backdrop of rising global demand for renewable energy, as countries and organizations strive to reduce carbon emissions and combat climate change, the new energy and semiconductor industries are experiencing higher prosperity and broader applications for their materials. Perovskite (PVK) materials, as novel light-collecting and light-emitting materials,<sup>1,2</sup> owe their excellent photoelectric properties to high optical absorption coefficients,<sup>2</sup> high carrier mobility, long carrier lifetimes, and adjustable bandgaps.<sup>3–6</sup> They find diverse applications in photovoltaics,<sup>7</sup> light emitting diodes (LEDs),<sup>8</sup> solar cells,<sup>9–11</sup> photodetectors,<sup>12</sup> and various other fields.<sup>13–17</sup> Following, the large-area PVK light-emitting fibers exhibited hold tremendous potential across fields such as lighting,<sup>18</sup> displays,<sup>19</sup> and wearable electronics.<sup>20</sup> Their unique properties and manufacturing advantages position them at the forefront of innovation within the optoelectronics industry, holding promise for continuous advancements and broader adoption in the future. Despite the significant research potential in the development of large-area PLF, comprehensive reviews on this subject remain limited. In response to this gap, we have compiled a summary of recent advances in large-area PLF, aiming to offer valuable insights and guidance for future research in this burgeoning field. This paper provides a detailed overview of the preparation methods for large-area PLF devices and their various applications, with a particular focus on green, blue, and red light-emitting devices (LEDs). We delve into the luminescence mechanisms of perovskite materials, including

electroluminescence, photoluminescence, and mechanoluminescence, and review the progress made in both photoluminescence and electroluminescence devices. Additionally, we explore the latest advancements in fabrication techniques and their implications for device performance and application. Furthermore, this review identifies emerging trends and future directions in the field, offering perspectives on how ongoing research can address current challenges and expand the practical applications of large-area perovskite light-emitting films. By highlighting both recent innovations and potential research avenues, this paper aims to contribute to the advancement of the field and to inspire future developments in perovskite-based optoelectronic technologies.

### 2. LARGE-AREA PHOTOLUMINESCENT PVK DEVICES

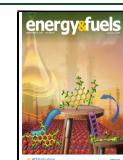
Advances in fabrication techniques, such as solution processing,<sup>20</sup> spin coating,<sup>21</sup> electrospinning,<sup>22–26</sup> and microfluidic methods,<sup>27</sup> have expanded the application potential of PVK to flexible substrates and scalable production. The ongoing research aims to enhance the performance, stability, and

Received: July 3, 2024

Revised: August 24, 2024

Accepted: August 27, 2024

Published: September 9, 2024



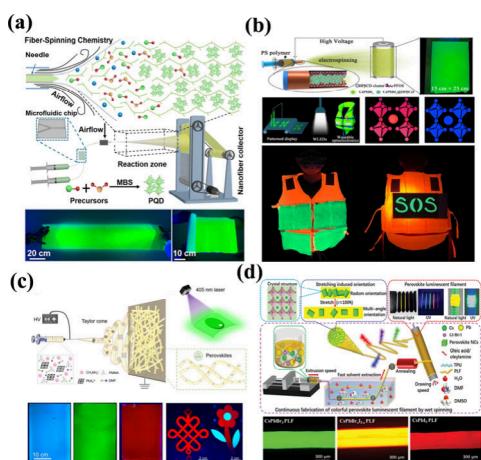
functionality of large-area PVK devices to meet the growing demands of flexible electronics and advanced lighting technologies. One key aspect of these devices is their photoluminescence mechanism. When PVK is photoexcited, the electrons within its band structure are excited by photon energy and transition from the valence band to the conduction band. During this process, excitons are generated, and visible light is emitted when these excitons recombine with other carriers and release energy. This fundamental mechanism underpins the operation of PVK-based light-emitting devices. The ability to modulate the optoelectronic performance of PVK through structural modifications and fabrication processes enables customized design for specific device requirements.<sup>28–30</sup> This chapter will discuss the preparation methods of different PLF and present the advancements in their fabrication, providing a comprehensive overview of the progress and potential in this field.

**2.1. Electrospinning Large-Area Flexible PLF.** Most PVK films are prepared using methods such as scratch coating,<sup>31</sup> spin coating<sup>21</sup> or inkjet printing<sup>32</sup> integrated on a small area rigid panel, and it is a great challenge to prepare uniform flexible PLF on a large scale with common batch preparation technology. Electrospinning is considered to be one of the best methods for preparing PLF.<sup>22–26</sup> In 2017, Jun Hai et al. successfully synthesized blue, green and red CsPbX<sub>3</sub> quantum dot (QD) codoped flexible film through an electrospinning process, and made it water-resistant with hydrophobic silicone resin coating.<sup>33</sup> In 2019, Lu et al. discussed a fiber spinning chemistry (FSC) method for in situ generation of highly stable halide PVK nanocrystalline (PNCs) fiber thin films.<sup>34</sup> In 2022, Cheng et al. prepared large-area PLF by microfluidic blowing. These films demonstrated excellent optical properties and could remain stable under extreme conditions such as high temperature and water exposure as shown in Figure 1 (a).<sup>35</sup> In the same year, Li et al. achieved continuous mass production without heavy metal waste by

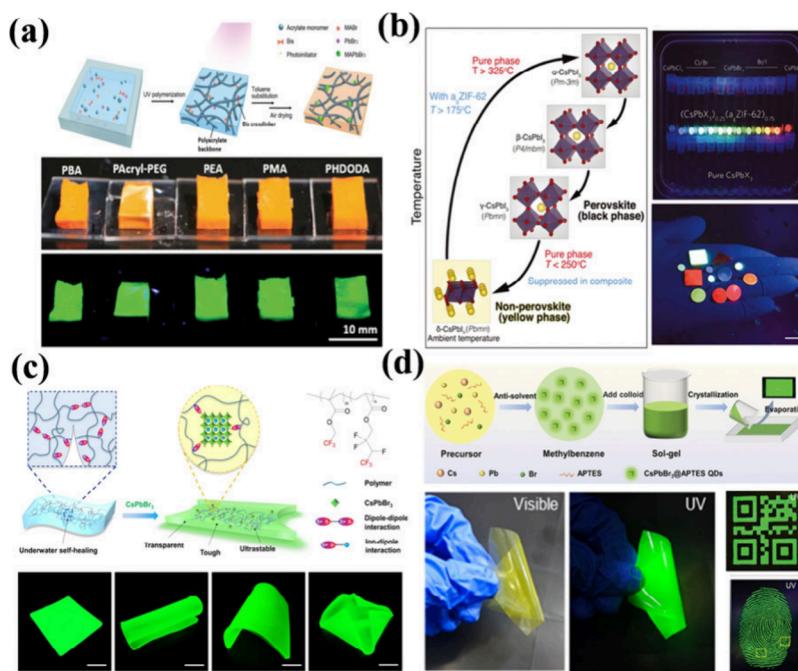
utilizing spinning fibers as reactors. This process allows for simultaneous solidification of polymers and in situ generation of nanocrystals during spinning. The resulting nanofiber films exhibit a tunable emission wavelength, high photoluminescence quantum yield (PLQY) and improved stability over time.<sup>36</sup> In 2023, Tian et al. developed flexible PVK luminescent textiles by electrospinning, they addressed issues such as instability, easy decomposition and toxic lead ion leakage associated with halide PVK materials. The textiles exhibited bright and stable photoluminescence with good resistance to water, ultraviolet radiation, high temperatures, and pressure as shown in Figure 1 (b).<sup>37</sup> That same year, Chen et al. focused on synthesizing large-area PVK-polymer fiber film (PPFM) also using an effective electrospinning strategy. The results showed that it has ultrastable underwater luminescence and strong electrostatic adhesion properties without additional adhesives.<sup>38</sup> Using programmable laser lithography, PPFM patterns for color or three-dimensional displays with self-stitched edges for enhanced mechanical stability<sup>39</sup> can be manufactured (Figure 1 (c)). In addition, Wang's team addressed the inherent instability of PVK materials by encapsulating them in a hydrophobic thermoplastic polyurethane (TPU) matrix. This encapsulation enabled the materials to maintain ultrahigh photoluminescent stability under various harsh conditions. The resulting CsPbX<sub>3</sub> QDs PLF show a wide range of applications in organic dye detection, strain sensing, flexible display, anticounterfeiting and hazard warning systems. The wet spinning technology used for manufacturing allows for the mass production of these PLF as shown in Figure 1 (d).<sup>40</sup>

## 2.2. Preparation of Large-Area PLF by Other Methods.

In addition to electrospinning methods, there are other methods for preparing large-area PLF. Likely, Zhang et al. developed homogeneous self-supported luminescent PVK organogels with excellent water resistance stability, which solved the instability problem of PVK quantum dots (QDs) in various environments as shown in Figure 2 (a).<sup>41</sup> Moreover, the physical encapsulation of PVK within stable rigid materials is another excellent strategy for maintaining persistent luminescence stability. For example, Hou et al. successfully prepared PVK composites based on zeolitic imidazolate frameworks (ZIF) glass through liquid phase sintering, which can stabilize the nonequilibrium PVK phase and improve the photoluminescence performance.<sup>42</sup> The resulting composites exhibit high stability under various environmental conditions, indicating their broad prospects for optoelectronics applications,<sup>42</sup> as shown in Figure 2 (b). Research on light-emitting fluoroelastomers has also garnered significant attention due to their shape adaptability and tunable dimensions.<sup>43–46</sup> For example, Liu et al. developed a new light-emitting composite using tough, self-healing fluoroelastomers as a polymer matrix, and then embedding PVK QDs into the polymer matrix to improve stability.<sup>47</sup> The composite is capable of self-healing underwater and maintains its mechanical and optoelectronic properties even after exposure to harsh environments, as illustrated in Figure 2 (c). These materials combine the flexible, stretchable properties of elastomers with the luminescent capabilities of PVK QDs, making them highly suitable for applications in flexible electronics and wearable devices. Thus, the unique properties of fluoroelastomers, such as high resilience, chemical resistance, and excellent mechanical properties, further enhance their potential for use in a wide range of optoelectronic applications.<sup>43</sup> In addition to the above



**Figure 1.** (a) Large area PLF prepared by spinning method;<sup>35</sup> Reproduced with permission from ref 35. Copyright 2022 John Wiley and Sons. (b) A schematic diagram of the preparation process of PLF;<sup>37</sup> Reproduced with permission from ref 37. Copyright 2023 Springer Nature. (c) An illustration of the electrospun perovskite–polymer fiber membranes (PPFMs);<sup>39</sup> Reproduced with permission from ref 39. Copyright 2023 Springer Nature. (d) Schematic illustration of the continuous fabrication of perovskite luminescent filament.<sup>40</sup> Reproduced with permission from ref 40. Copyright 2024 John Wiley and Sons.



**Figure 2.** (a) Schematic of the preparation of perovskite nanoparticles (PNP) gels.<sup>41</sup> Reproduced with permission from ref 41. Copyright 2019 John Wiley and Sons. (b) Preparation PVK composites based on zeolitic imidazolate frameworks (ZIF) glass.<sup>42</sup> Reproduced with permission from ref 42. Copyright 2021 The American Association for the Advancement of Science. (c) Schematic illustration of the stretchable, stable and self-healable luminescent perovskite-polymer material.<sup>47</sup> Reproduced with permission from ref 47. Copyright 2022 Springer Nature. (d) Schematic of CsPbBr<sub>3</sub>@APTES QDs preparation and flexible film for codes and fingerprints application.<sup>51</sup> Reproduced with permission from ref 51. Copyright 2024 John Wiley and Sons.

methods, large-area PLF can also be obtained through a simple casting method.<sup>48–50</sup> For instance, a molecular-level hybridization of bridged polysilsesquioxane (BPSQ) is designed as a matrix to combine the flexibility of organics with the stability of inorganics, enhancing the interfacial compatibility between CsPbBr<sub>3</sub> QDs and the matrix through chemical bond anchoring.<sup>51</sup> The white light emitting diodes (WLEDs) and anticounterfeiting patterns have been successfully fabricated using CsPbBr<sub>3</sub>@APTES@BPSQ, highlighting their broad potential applications in flexible light-emitting devices and information encryption, as shown in Figure 2 (d).<sup>51</sup>

### 3. LARGE-AREA PVK LEDs

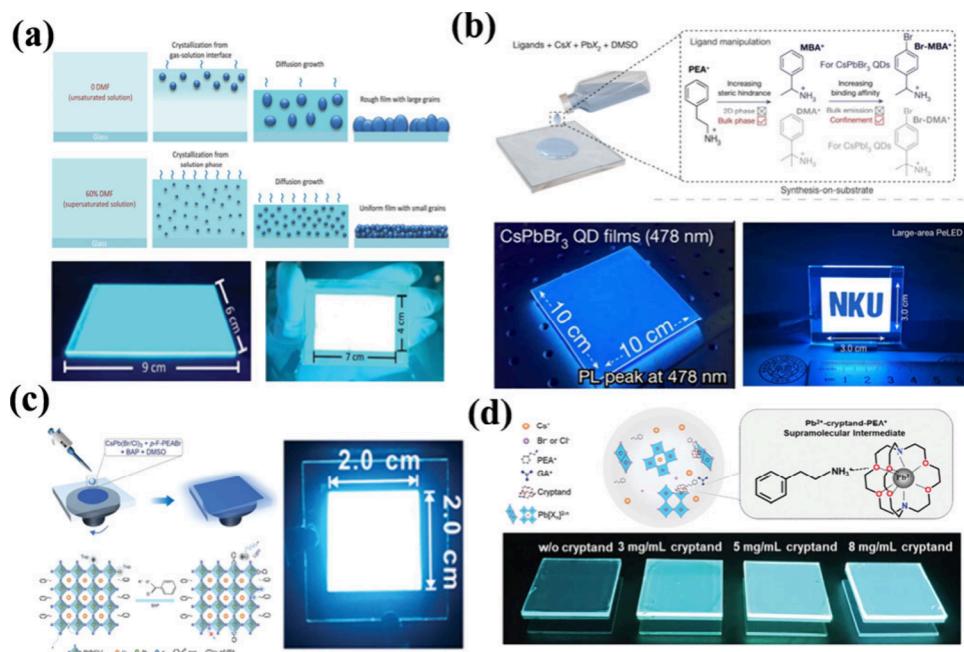
For PVK LEDs, the mechanism involves current injection. When a voltage is applied, current flows through the PVK material, causing electrons to move across the material and recombine with holes. This recombination process results in the release of energy in the form of visible light. The ability to modulate the optoelectronic performance of PVK through structural modifications and fabrication processes enables customized design for specific device requirements. Motivated by excellent optical properties and high luminescence efficiency of PVK materials,<sup>52–54</sup> in the past five years, the external quantum efficiency of red and green light PVK LEDs devices has exceeded 20%, which holds promise for lighting and displaying devices.<sup>55</sup> Therefore, many efforts are devoted to improving the performance and stability of large-area PVK LEDs, which aim to promote their widespread application in smart displays, lighting, sensing, and other advanced technology fields.<sup>56–60</sup> Meanwhile, the importance of large-area PVK LEDs lies in their potential to revolutionize energy-efficient lighting solutions and flexible electronic applications,

addressing the increasing global demand for sustainable and adaptable illumination technologies. This chapter will discuss the preparation and development of typical large-area PVK LEDs in three light colors: red, green and blue. A summary of these developments is provided in Table 1.

**3.1. Large-Area Blue-Light PVK LEDs.** PVK LEDs, as a promising candidate material for next-generation display technologies, are receiving more and more attention. Although great progress has been made in this field, the device performance of blue LEDs still lags far behind that of green

**Table 1. Summary of Different Color PVK LEDs**

Emitting color	Peroxskite	EQE	Reference
Blue	CsPbCl <sub>0.99</sub> Br <sub>2.0</sub>	2.4%	62
	CsPb(Br <sub>0.84</sub> Cl <sub>0.16</sub> ) <sub>3</sub>	10.3%	63
	CsPbBr <sub>3</sub>	10.3%	64
	CsPb(Cl/Br) <sub>3</sub>	18.65%	65
	CsPbBr <sub>3</sub>	11.87%	67
	CsPbBr <sub>3</sub>	20.3%	55
Green	CsPbBr <sub>3</sub>	18.7%	78
	CsPbBr <sub>3</sub>	16.2%	79
	FAPbBr <sub>3</sub>	20.1%	81
	CsPbBr <sub>3</sub>	16.8%	82
	CsPbBr <sub>3</sub> /Cs <sub>4</sub> PbBr <sub>6</sub>	8.0%	81
	(PEA) <sub>2</sub> Cs <sub>2</sub> Br <sub>7</sub>	8.24%	84
Red	PEA <sub>2</sub> (FAPbBr <sub>3</sub> ) <sub>2</sub> PbBr <sub>4</sub>	30.84%	85
	CsPbI <sub>2</sub> Br	17.8%	87
	CsPb(Br/I) <sub>3</sub>	23.5%	88
	CsPbBrI <sub>2</sub>	9.32%	94
	CsPbI <sub>3</sub>	23%	97
	CsPbI <sub>3</sub>	9.8%	98



**Figure 3.** Large-area Blue light PVK LEDs. (a) Schematic diagram of crystallization process of unsaturated and large-area blue LED display;<sup>63</sup> Reproduced with permission from ref 63. Copyright 2022 John Wiley and Sons. (b) Photograph of the large-area blue PeLED;<sup>64</sup> Reproduced with permission from ref 64. Copyright 2022 Springer Nature. (c) A photograph of a large-area BAP-modified sky-blue PeLED;<sup>65</sup> Reproduced with permission from ref 65. Copyright 2023 John Wiley and Sons. (d) A PVK film with different concentration of crypt and by 365 nm ultraviolet lamp excitation.<sup>66</sup> Reproduced with permission from ref 66. Copyright 2024 John Wiley and Sons.

and red light. This is mainly due to the high density of the trap and the undesirable redshift of the electroluminescence spectrum under working conditions.<sup>61</sup> To address these challenges, researchers have conducted a series of studies. For instance, Pan et al. successfully synthesized doped-PVK QDs using the room temperature recrystallization method, allowing for the adjustment of the emission wavelength. By doping Ni<sup>2+</sup> ions and adjusting the Cl/Br ratio, they achieved a blue emission of 470 nm with PLQY of 89%, surpassing that of undoped PVK QDs. The blue emission LEDs prepared based on these PQDs demonstrated an external quantum efficiency (EQE) of 2.4% and a maximum brightness of 612 cd/m<sup>2</sup>, outperforming previous LEDs based on blue emission PVK QDs.<sup>62</sup> By adjusting the volume ratio of methyl sulfoxide (DMSO) to dimethylformamide (DMF), Chu et al. obtained a saturated CsPb (Br<sub>0.84</sub>Cl<sub>0.16</sub>)<sub>3</sub> solution for scaling, resulting in a uniform film with smaller particle size, lower trap density, and higher radiation recombination rate.<sup>63</sup> The EQE of the prepared PVK LEDs reached 10.3%, and emitted sky blue light with a wavelength of 489 nm. They successfully fabricated large-area devices covering an area of 28 cm<sup>2</sup> as illustrated in Figure 3(a).<sup>63</sup> By ligand engineering, Jiang et al. successfully synthesized monodisperse QDs with a diameter of less than 5 nm, enabling the preparation of an area of 9 cm<sup>2</sup> of blue PVK LEDs (9 cm<sup>2</sup>) with high EQE, as shown in Figure 3 (b).<sup>64</sup> Furthermore, Zhou et al. utilized bifunctional passivation to inhibit halogen ion migration and passivate lead atoms with insufficient coordination, resulting in an increase in a stable emission wavelength of 483 nm and the EQE to 28.82%, as depicted in Figure 3(c). They found that the introduction of Lewis base benzoate ions and alkali metal cations can alleviate the behavior of halogen ions through optical coupling enhancement.<sup>65</sup> In 2024, Yuan et al. introduced Mill's cage as an additive to regulate the phase distribution in quasi-two-

dimensional PVK films.<sup>66</sup> This intervention improves energy transfer and effective radiation recombination, resulting in EQE enhancement to 10.16% and operational stability of PVK LEDs, as illustrated in Figure 3 (d). To eliminate chlorine vacancies, and improve the PLQY and lifetime of PVK nanocrystals, various types of cations,<sup>67–69</sup> acid<sup>70,71</sup> and alkaline surfactants<sup>72–74</sup> were added into the preparation of PVK LEDs. For example, some teams passivated quantum dots using trifluoroacetic acid and designed mixed hole transport layers to enhance the combination of carrier injection and transport capabilities.<sup>75,76</sup> As a result, the EQE has gradually increased, and the fabrication area has expanded. Looking forward, the stability and fabrication cost of blue light PVK LEDs will be critical factors worth considering.

**3.2. Large-Area Green-Light PVK LEDs.** Compared to blue light PVK LEDs, green light PVK LEDs are typically easier to control and fabricate during preparation. They also exhibit higher stability, which is crucial for industrial production and long-term use. Additionally, the wavelength of green PVK LEDs falls within the higher energy region of the visible spectrum, making them advantageous for attracting broader application demands in areas such as display technologies and lighting applications. Therefore, researching and developing large-area green light PVK LEDs contributes to meeting market demands for stable, easily processed, and high-performance optoelectronic devices. Accordingly, Wei's team achieved a significant breakthrough by surpassing 20% EQE of green PVK LEDs, which incorporated MABr (MA is CH<sub>3</sub>NH<sub>3</sub><sup>+</sup>) into CsPbBr<sub>3</sub> to form a quasi-core/shell structure. This achievement marked the first instance where EQE exceeded 20%, paving the way for industrial applications.<sup>55</sup>

Some interface passivation strategies are used to improve the efficiency and stability of the device.<sup>77</sup> For example, Xu et al. used organic molecules to passivate the top and bottom

interfaces of quantum dot films to improve efficiency and stability. After testing various molecules, the passivated device achieved a maximum EQE of 18.7% and a current efficiency of  $75 \text{ cd A}^{-1}$ , with a 20-fold increase in operating life. These findings highlight the importance of interface passivation for efficient and stable quantum dot-based optoelectronic devices,<sup>78</sup> as shown in Figure 4 (a). In addition, defects can

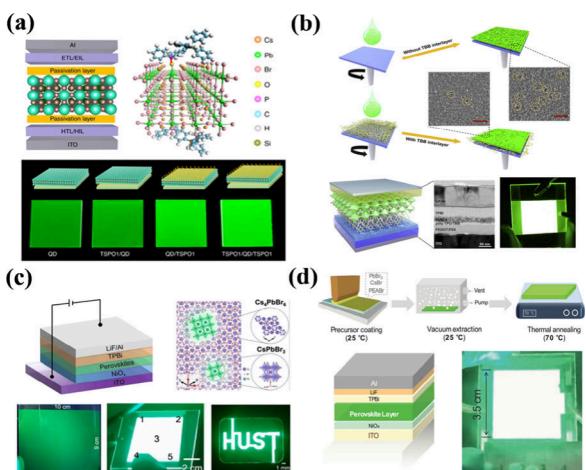
migration, constructing a highly stable and efficient pure green PeLED that approximates the standard green light of Rec.2020.<sup>86</sup> In short, with the efforts of scientific researchers, green PVK LEDs technology has gradually matured and made great contributions to the industrial application of panchromatic LEDs.

### 3.3. Large-Area Red-Light PVK LEDs.

Large-area red light PVK LEDs devices hold significant importance due to several key factors. First, red light is a critical component of RGB (Red, Green, Blue) displays, and high-quality red PVK LEDs can enhance color purity and brightness in screens used for televisions, smartphones, and other display devices.<sup>87,88</sup> And then, red LEDs are essential for various lighting applications, including horticultural lighting, mood lighting, and specialized industrial uses. Furthermore, PVK materials offer the potential for high luminous efficiency, which can lead to lower energy consumption in lighting and display applications.<sup>89–91</sup> Additionally, PVK materials are known for their low-cost raw materials and relatively simple fabrication processes compared to traditional semiconductor materials,<sup>92,93</sup> making large-area red PVK LEDs economically attractive.

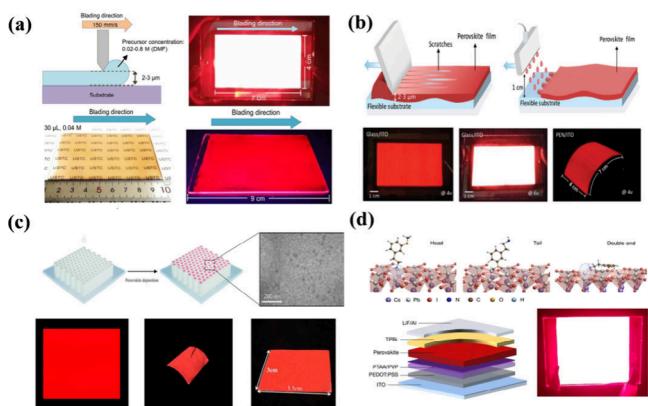
The development of large-area red light PVK LEDs has seen significant advancements, but challenges remain to be addressed. Recent research has focused on improving the EQE of red-light PVK LEDs. Techniques such as doping,<sup>94</sup> dimension regulation,<sup>95</sup> ligands engineering<sup>96</sup> and surface passivation<sup>97,98</sup> have been employed to enhance performance. For instance, Yang et al. developed highly efficient and spectral stable pure red PVK LEDs based on quasi-two-dimensional (quasi-2D) PVK, in which phenylethylamine (PEA) and 1-naphthylmethylamine (NMA) cations were introduced.<sup>99</sup> And Yan et al. discussed the synthesis of a novel 0D manganese-organic-inorganic hybrid PVK with red emission properties and its application to lead-free red light PVK LEDs.<sup>100</sup> However, one of the main issues with PVK materials is their environmental instability. Efforts are being made to improve the operational lifetime and stability of red PVK LEDs through encapsulation techniques and the development of more stable PVK compositions. While small-area devices have shown promising results, scaling up to large-area devices introduces new challenges. Uniformity of the PVK film, defect control, and high efficiency over larger areas are key focus areas. Accordingly, Chu et al. significantly improved the EQE of PVK LEDs by regulating the sol-gel phase of the film formation process.<sup>101</sup> They carried out sol-gel engineering on the low temperature scraping of methylamine-lead iodine ( $\text{MAPbI}_3$ ) PVK film, and obtained a large area of hyperplane film with an EQE of 16.1% and an area of  $28 \text{ cm}^2$ . This method achieves a uniform and efficient PVK led with a luminous layer cost as low as 0.02 cents per square centimeter,<sup>101</sup> as shown in Figure 5 (a). In addition, by incorporating an amino ligand into pure iodine PVK, researchers stabilize the I-Pb-I octahedral structure in  $\text{CsPbI}_3$  PVK, thereby increasing the band gap to achieve pure red emission, reducing iodine migration under electric bias.

Advances in fabrication techniques, such as solution processing,<sup>102,103</sup> template method,<sup>104</sup> vapor deposition,<sup>105</sup> and inkjet printing,<sup>106</sup> are being explored to produce large-area red PVK LEDs with consistent quality and performance. For example, Liu et al. made significant progress in developing large-area flexible PVK LEDs using inkjet printing technology. This approach successfully overcame the challenge of



**Figure 4.** Large-area green light PVK LED. (a) The sectional image of device structure;<sup>78</sup> Reproduced with permission from ref 78. Copyright 2020 Springer Nature. (b) The schematic diagram of preparation a  $20 \text{ mm} \times 20 \text{ mm}$  green PVK LEDs;<sup>81</sup> Reproduced with permission from ref 81. Copyright 2020 Elsevier. (c) The diagram of PVK LEDs structure and the schematic diagram of  $\text{CsPbBr}_3$  embedded in the  $\text{Cs}_4\text{PbBr}_6$  matrix;<sup>83</sup> Reproduced with permission from ref 83. Copyright 2021 Springer Nature. (d) A schematic structure of the PVK LEDs with an emitting area of  $3.5 \times 3.5 \text{ cm}^2$ .<sup>84</sup> Reproduced with permission from ref 84. Copyright 2021 John Wiley and Sons.

be passivated by introducing other substances.<sup>79,80</sup> Such as, Wang et al. employed precise control over film formation dynamics and introduced bromine (Br) species to eliminate pinholes, passivate defects, prevent short circuits, and reduce nonradiative recombination. This approach significantly enhanced charge injection and transport, leading to the fabrication of  $20 \text{ mm} \times 20 \text{ mm}$  green PVK LEDs with an EQE exceeding 16%,<sup>81</sup> as depicted in Figure 4 (b). Han et al. used methylene bisacrylamide cross-linking methods to passivate defects and inhibit ion migration, thereby improving the external quantum efficiency (EQE) and operational stability of PeLEDs.<sup>82</sup> In the following year, Du et al. developed efficient and large-area PVK LEDs by conducting space-confined enhanced radiative recombination and inhibition of nonradiative recombination in vacuum-deposited  $\text{Cs-Pb-Br}$  films,<sup>83</sup> as shown in Figure 4 (c). Also this year, Chaoran Chen et al. used a vacuum process to control crystallization dynamics, and by separating precursor deposition and thermal annealing, they prepared dense and uniform PVK films, achieving efficient energy transfer. The quasi-two-dimensional cesium lead chloride tribromide layer prepared by this method achieves high external quantum efficiency of 8.24% and 6.12% over different effective areas,<sup>84</sup> as shown in Figure 4 (d). In 2023, Bai et al. achieved a record EQE of 30.84% at  $6514 \text{ cd m}^{-2}$  brightness by optimizing the device structure and using the  $\text{Ni}_{0.9}\text{Mg}_{0.1}\text{O}_x$  film as the hole injection layer.<sup>85</sup> This year, Zhang et al. synthesized a ligand (IMiligand) that inhibits ion



**Figure 5.** Large-area red light PVK LED. (a) A schematic of the doctor-blading process and photo image of ultra large-area PVK LEDs with a device area of  $4 \times 7 \text{ cm}^2$ .<sup>101</sup> Reproduced with permission from ref 101. Copyright 2021 Springer Nature. (b) Schematics of the blade coating fabricated PVK LEDs.<sup>107</sup> Reproduced with permission from ref 107. Copyright 2023 John Wiley and Sons. (c) Schematic illustrations of the PVK deposition process on porous alumina membranes (PAM) template;<sup>108</sup> Reproduced with permission from ref 108. Copyright 2023 Springer Nature. (d) Adsorption models and device structure of PVK LEDs.<sup>109</sup> Reproduced with permission from ref 109. Copyright 2024 Springer Nature.

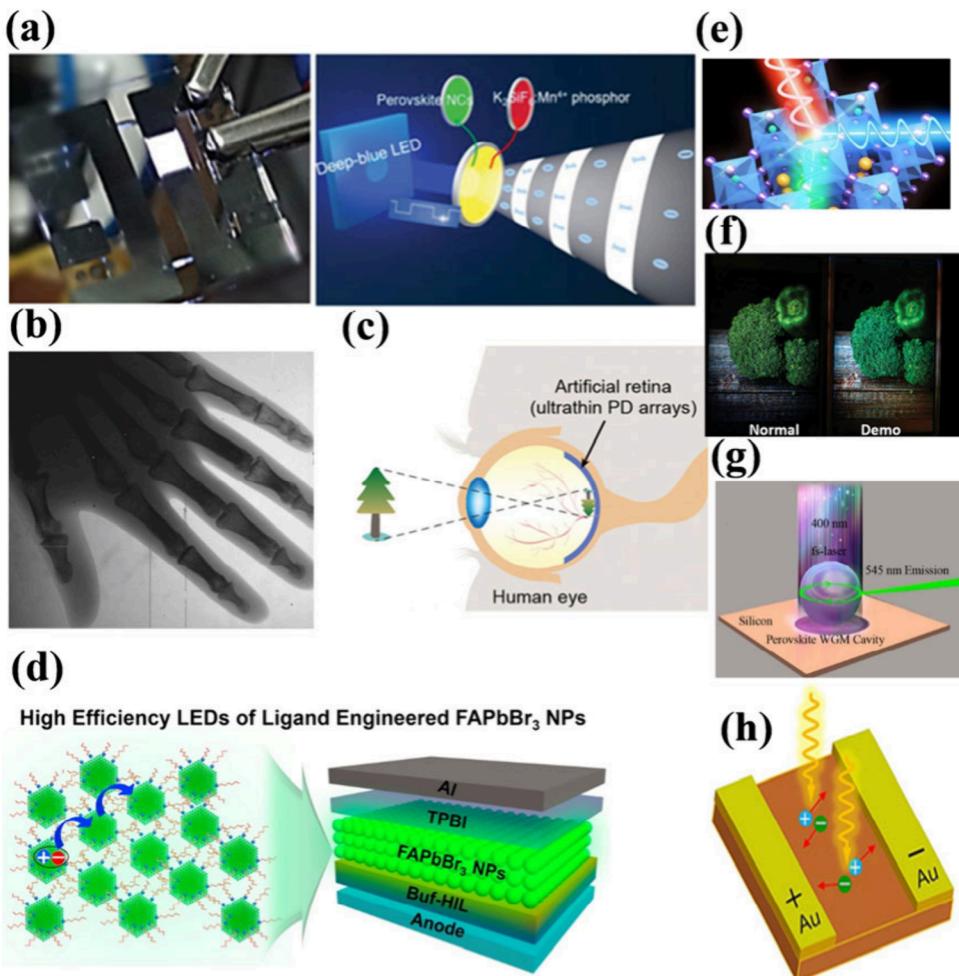
depositing uniform perovskite films on flexible substrates,<sup>107</sup> as shown in Figure 5 (b). Their work highlighted the potential of inkjet printing for creating high-quality flexible PVK LEDs, opening new avenues for wearable and flexible electronic applications. By using porous alumina films as templates, Cao et al. successfully fabricated high-density and homogeneous crystalline all-inorganic PVK quantum wire arrays achieving high PLQY of pure red PVK quantum wire arrays,<sup>108</sup> as shown in Figure 5 (c). This year, another significant advancement involves the synthesis of PbCl<sub>IX</sub>-modified CsPbI<sub>3</sub> QDs using a nucleophilic reaction. This method results in stable electroluminescence spectra and improved efficiency decay and half-lifetime, demonstrating the most advanced pure red PVK LEDs to date,<sup>109</sup> see Figure 5 (d). The development of large-area red PVK LEDs has seen remarkable progress, driven by innovations in fabrication techniques and material engineering. These advancements address critical challenges in scaling production and improving device performance. The ongoing research efforts are poised to enhance the practical application of red PVK LEDs in various fields, including displays and lighting.

#### 4. CURRENT CHALLENGE AND PERSPECTIVES OF LARGE-AREA PLF

PLF has shown immense potential in many applications based on their unique optical and electronic properties. Below, we discuss some cutting-edge applications of PLF. For example, in flexible electronics large-area PLF could revolutionize flexible electronics, Cs<sub>4</sub>PbBr<sub>6</sub>/CsPbBr<sub>3</sub> nanocomposites containing polymer films with enhanced water resistance and light stability have been prepared by a simple *in situ* preparation method, which can be used in ultrawide color gamut displays.<sup>108</sup> The Cs<sub>4</sub>PbBr<sub>6</sub> matrix provides a protective coating for the easily degraded CsPbBr<sub>3</sub> nanocrystals and improves their stability. The room temperature saturated recrystallization method minimizes the contact time between CsPbBr<sub>3</sub> nanocrystals and water/oxygen,<sup>110</sup> further improving stability

including flexible displays<sup>56,111</sup> and wearable devices.<sup>112–116</sup> In the field of advanced lighting, the researchers used an electric field deposition technique to fabricate a dense, smooth NP-based film. The (PEA)<sub>2</sub>PbBr<sub>4</sub> NP films exhibit dark blue emission at 410 nm and have excellent stability under environmental conditions. This highly efficient dark blue LED can be used as an excitation light source to achieve white light emission, showing potential in optical communication applications (Figure 6 (a)).<sup>116</sup> With their high PLQY and tunable emission spectra, PVK films are ideal candidates for next-generation lighting solutions. These could include energy-efficient, color-tunable LED<sup>8,117–121</sup> lighting systems for both residential and commercial applications. At the same time, the exceptional light absorption and charge transport properties of PVK materials make them highly suitable for X-ray imaging, medical devices and LEDs applications (Figure 6 (b-d)).<sup>15,120,122–128</sup> It is also widely used in lasers,<sup>129,130</sup> photodetectors<sup>131–134</sup> and light-emitting transistors optoelectronic devices. The PVK lasers demonstrate excellent performance in terms of cost-effectiveness, low threshold, high coherence and multicolor tunability (Figure 6 (e-h)).<sup>135–137</sup> For example, by adjusting the halogen composition and size of the microspheres, researchers can continuously tune the wavelength of the single-mode laser from red to purple (425–715 nm),<sup>136</sup> as shown in Figure 6 (g). The finely controlled synthesis of cesium lead halide PVK nanospheres/microspheres provides an alternative way to fabricate widely tunable miniaturized single-mode lasers. In short, PLF has different application potential in various fields, but it is still a great challenge to industrialize and generalize it. For the future development of large-area PLF, minimizing capital investment costs, particularly for equipment and fixed assets, is crucial. This requires a thorough understanding of the production equipment needed for high-efficiency, large-area PLF while focusing on cost reduction. Selecting cost-effective production equipment and optimizing production processes are essential for lowering production costs. Additionally, using durable and low-maintenance equipment will help reduce long-term operational expenses. Properly sizing production areas to avoid resource waste is also a key cost-reduction strategy. Technological innovation, exploration of new processes and advanced materials, and optimization of equipment configurations are vital to achieving cost-effectiveness. By addressing these aspects, it is possible to enhance the efficiency and lifespan of PLF devices while reducing costs, thereby boosting their competitiveness and facilitating their widespread application across various fields.

In conclusion, this review offers an in-depth examination of the preparation methods and diverse applications of photo-excited PLF and electro-excited large-area perovskite LEDs in red, green, and blue colors. It also explores the utilization of perovskite films across various sectors, including lighting, display technology, anticounterfeiting, and medical applications. In addition to presenting these advancements, the paper critically addresses the current challenges facing the development of large-area PLF, such as cost reduction, efficiency optimization, and production scalability. It also discusses prospects and potential breakthroughs in this rapidly evolving field. By providing a comprehensive overview of both achievements and ongoing obstacles, which aims to offer valuable insights and guidance for researchers and practitioners seeking to advance the field of PLF and expand its practical applications.



**Figure 6.** Application of large-area PVK film. (a) Photograph of a white PVK LED.<sup>116</sup> Reproduced with permission from ref 116. Copyright 2019 John Wiley and Sons. (b) A hand phantom X-ray image obtained from a polycrystalline MAPbI<sub>3</sub>(MPC) detector.<sup>13</sup> Reproduced with permission from ref 13. Copyright 2017 Springer Nature. (c) A schematic illustration of the ultrathin device for artificial vision sensing application;<sup>15</sup> Reproduced with permission from ref 15. Copyright 2021 John Wiley and Sons. (d) High-efficiency LEDs of ligand-engineered FAPbBr<sub>3</sub> NPs.<sup>121</sup> Reproduced with permission from ref 121. Copyright 2017 Elsevier. (e) Schematic diagram of PVK semiconductor laser;<sup>135</sup> Reproduced with permission from ref 135. Copyright 2021 American Chemical Society. (f) The stacked polymer films;<sup>110</sup> Reproduced with permission from ref 110. Copyright 2020 John Wiley and Sons. (g) Schematic of individual CsPbBr<sub>3</sub> microspheres on;<sup>136</sup> Reproduced with permission from ref 136. Copyright 2017 American Chemical Society. (h) The X-ray detector based on the (BDA)PbI<sub>4</sub> crystal.<sup>137</sup> Reproduced with permission from ref 137. Copyright 2020 John Wiley and Sons.

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### Notes

The authors declare no competing financial interest.

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## ACKNOWLEDGMENTS

This work was supported by the National Natural Science Foundation of China (NSFC 22305215), Guangxi Key Laboratory of Advanced Structural Materials and Carbon Neutralization (2023 Open Funding, GXAMCN23-7).

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