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Review

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Key factors influencing perovskite film crystallization in blade-coating processes

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Abstract: Perovskite films have fostered significant progress in the field of optoelectronic devices, particularly in solar cells, due to their excellent optoelectronic properties and cost-effective fabrication process. As a promising technique for large-scale industrial production of perovskite films, the blade-coating method has attracted wide attention because of its low cost, large-area coating capability, and simplicity. However, during the blade-coating process, perovskite films often present challenges such as poor film uniformity, high defect density, and uneven crystallization, which greatly affect the efficiency and stability of the devices. In this review, we summarize the application of blade-coating methods to the fabrication of perovskite films, with a focus on analyzing key factors affecting film quality, including precursor solution formulations, solvent characteristics, evaporation rate, crystallization kinetics, and film thickness. In particular, we discuss the impact of environmental factors on the perovskite crystallization process during blade-coating and explore how optimizing the blade-coating process and precursor solution composition can improve film uniformity and device performance. Additionally, we discuss the main challenges and shortcomings in the blade-coating preparation of perovskite films, including defects during large-area fabrication, differences in solvent evaporation rates, and their effects on crystallization quality. Strategies for addressing these issues are proposed. Finally, the future prospects of the blade-coating method in large-scale production of perovskite films are outlined, emphasizing the importance of a deeper understanding of perovskite film crystallization mechanisms and the development of novel additives to enhance the performance of perovskite optoelectronic devices and accelerate their industrialization.

Key words: Perovskite thin films; Blade-coating; Crystal growth

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1 Introduction

Perovskite materials have revolutionized optoelectronics due to their exceptional optoelectronic properties, including high absorption coefficients, long charge carrier diffusion lengths, and tunable bandgaps (Kojima et al., 2009; Kim et al., 2013; Dong et al., 2015; Saidaminov et al., 2015; Wang et al., 2017). As a disruptive photovoltaic technology, perovskite solar cells (PSCs) within decades have achieved a certified power conversion efficiency (PCE) of 26.95%. It is crucial to produce high-quality perovskite films that not only have controllable crystallinity but also maintain uniform morphology and minimized defect density. The crystallization process of the perovskite film formation directly affects the optoelectronic performance and long-term stability of the devices. Therefore, optimizing the crystallization process during film formation is key to obtaining high-performance devices.

Essentially, the crystallization mechanism of perovskite thin films conforms to the theoretical framework established by the classical LaMer model (Whitehead et al., 2019), which delineates nucleation and subsequent crystal growth. The change in concentration of a precursor solution as a function of time can be illustrated qualitatively by a LaMer diagram (**Fig. 1a**). During solvent evaporation, the solute concentration reaches saturation concentration (Cs) at time t_1, yet nucleation remains inhibited due to

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the existing energy barrier. Upon surpassing the critical concentration (C_c) at time t_2, the system enters the primary nucleation phase (Stage I). When the concentration is higher than C_c (stage II), both nucleation and crystal growth take place. Due to the competition between solvent evaporation and solute consumption by crystal growth, the concentration of the solution then starts to decrease and finally drops below C_c. As the concentration decreases below Cc (Stage III), nucleation ceases while existing grains continue growing until the system reaches sub-saturation concentration (Cs) (Zeng et al., 2020). Therefore, the perovskite film morphologies and properties obtained in this way are significantly determined by its crystallization (Xu et al., 2024).

Spin-coating is the most widely used technique for fabricating perovskite films, as it allows for rapid and uniform film deposition. However, the narrow time window for adding anti-solvent may result in very low tolerance during the device fabrication process, limiting the yield and reproducibility of perovskite devices, especially in large-scale production (Xiao et al., 2014; Yang et al., 2015; Ge et al., 2016; Yang et al., 2016). Additionally, the powerful centrifugal force generated during spin-coating to apply the film and remove excess solution can lead to significant variation in the film thickness across different regions of the substrate (Heo et al., 2014; Ye et al., 2016; Chen et al., 2017). This is undesirable for fabricating large-area devices.

Blade-coating has emerged as a compelling alternative, offering high throughput and exceptional material utilization (Jeong et al., 2019; Park and Zhu, 2020). As illustrated in **Fig. 1b** and **Fig. 1c**, this technique uses a doctor blade to shear-align precursor solutions into uniform wet films, coupled with assisted methods like gas quenching (Huang et al., 2014; Gao et al., 2017; Song et al., 2017; Zheng et al., 2017) to modulate solvent evaporation kinetics. Gas quenching introduces a laminar airflow parallel to the substrate, enabling faster solvent extraction rates than natural evaporation (Kwon et al., 2007; Ghosh et al., 2008; Cho et al., 2009; Zhang et al., 2012). This rapid solvent depletion can potentially induce nucleation and directional crystal growth. Compared to spin-coated films, blade-coated films show enhanced grain uniformity, reduced defect density and low cost. Here, we review and summarize recent progress in blade-coated perovskite solar cells (**Table 1**).

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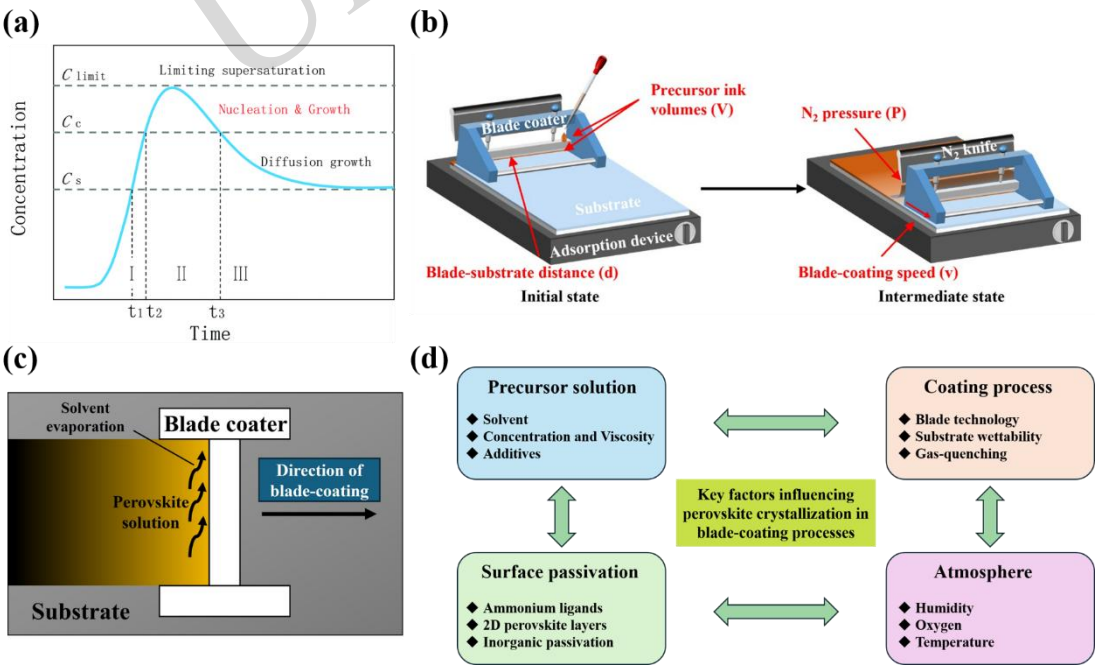

Fig. 1 (a) The LaMer model of nucleation and crystal growth (reprinted from (Zeng et al., 2020), Copyright 2020, with permission from Royal Society of Chemistry); (b) Schematic illustration of the N_2 knife-assisted blade-coating process (reprinted from (Hu et al.,2025), Copyright 2025, with permission from Wiley); (c) Top-view schematic illustration of the blade-coating process; (d) Key factors influencing perovskite crystallization in the blade-coating processes.

Table 1 Literature survey and summary of recent progress in blade-coated perovskite solar cells. All the efficiency data we collected are the highest values reported by the authors.

Year	Bandgap (eV)	Device type (rigid cell)	Area (cm^2)	PCE (%)	Strategy	Reference
2025	1.53	p-i-n	0.09	24.31	Additives	(Yang et al., 2025)
2025	~1.60	p-i-n	0.08	21.4	Additives	(Feng et al., 2025)
2025	1.52	p-i-n	0.09	24.10	Additives	(Wu et al., 2025)
2025	1.55	p-i-n	0.08	21.74	Solvent engineering	(Hu et al., 2025)
2024	1.68	p-i-n	0.049	20.77	Solvent engineering	(Zheng et al., 2024)
2024	1.78	p-i-n	1.05	19.6	Solvent engineering	(Duan et al., 2024)
2024	1.68	p-i-n	1.05	21.5	Solvent engineering	(Duan et al., 2024)
2024	1.68	p-i-n	N/A	21.31	Additives	(Ge et al., 2024)
2024	1.77	p-i-n	0.07	19.45	Post-treatment	(Pu et al., 2024)
2024	1.77	p-i-n	1	18.71	Post-treatment	(Pu et al., 2024)
2024	1.70	n-i-p	0.1	19.05	Solvent engineering	(Abate et al., 2024)
2024	1.77	p-i-n	0.09	19.28	Surface passivation	(Zhang et al., 2024)
2024	1.77	p-i-n	1	18.08	Surface passivation	(Zhang et al., 2024)
2024	1.67	p-i-n	0.07	22.06	Additives	(Jia et al., 2024)
2024	1.67	p-i-n	1.02	19.63	Additives	(Jia et al., 2024)
2023	1.68	p-i-n	0.07	19.92	Additives	(Zhang et al., 2023)
2023	1.68	p-i-n	10	17.70	Additives	(Zhang et al., 2023)
2023	1.80	p-i-n	0.09	18.92	Solvent engineering, surface passivation	(Tang et al., 2023)
2022	1.65	p-i-n	0.06	21.90	Additives	(Yang et al., 2022)
2022	1.72	n-i-p	46.5	16.07	Solvent engineering, surface passivation	(Tian et al., 2022)
2022	1.72	n-i-p	123.0	13.03	Solvent engineering, surface passivation	(Tian et al., 2022)
2020	1.65	p-i-n	N/A	20.10	Additives	(Chen et al., 2020)

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Furthermore, blade-coating is particularly advantageous for large-area perovskite module fabrication, as it enables scalable and continuous film deposition. However, achieving uniform crystallization across large-area substrates remains a significant challenge. Variation in the coating speed, solution wetting behavior, and drying conditions can lead to non-uniform thickness and defects such as pinholes and grain boundary discontinuities, which ultimately impact module efficiency. Recent studies have focused on optimizing solvent engineering, substrate pre-treatment, and blade-coating parameters to enhance the uniformity and reproducibility of large-area perovskite films. Addressing these challenges is crucial for improving not only module efficiency but also the overall production efficiency of perovskite photovoltaics.

Nevertheless, the crystallization dynamics in blade-coating still faces several challenges. The crystallization process is highly sensitive to factors such as solution properties, process parameters, and environmental conditions. Specifically, variations in precursor formulations, solvent evaporation rates, coating speed, and post-treatment processes such as surface passivation can significantly affect crystallization behavior, leading to uneven crystallization, phase segregation, and defects (Yang et al., 2019; Park and Zhu, 2020; Han et al., 2025). These factors play crucial roles in nucleation and crystal growth, directly influencing the microstructure of the films, crystallization quality, and ultimately, the optoelectronic performance (Yin et al. ; Ding et al., 2019; Park and Zhu, 2020) (**Fig. 1d**). Therefore, systematically analyzing how these factors interact and their specific impacts on film quality is essential for achieving consistent crystallization and high-quality perovskite.

In this review we summarize the key factors

influencing the crystallization of perovskite films in blade-coating, focusing on the optimization of solution properties, process parameters, and environmental conditions to improve film quality. By delving into nucleation and crystal growth mechanisms, we aim to provide theoretical guidance for achieving controlled crystallization, uniform grain growth, and high-quality films. and practical insights for the development of scalable, high-performance perovskite photovoltaics.

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2 Precursor solution

2.1 Solvent

The choice of solvent not only regulates the crystallization process of the film by influencing the volatility, polarity, and boiling point of the solution, but also affects the solubility, nucleation, and crystal growth behavior of the perovskite precursor through its interaction with the precursor. The solvent's polarity and Gutmann donor number (DN) are key factors that influence the crystallization behavior of perovskite films. Studies have shown that the higher the DN value of a solvent, the stronger its coordination ability with metal ions in the perovskite precursor, thereby affecting the solubility and crystallization process of the solution (Hamill et al., 2018; Vidal et al., 2021; Wu et al., 2021; Lee et al., 2022). When the DN value of the solvent exceeds 18 kcal/mol, it can effectively dissolve lead halides, promoting the dissolution and crystallization of perovskite. Dimethyl sulfoxide (DMSO) and dimethylformamide (DMF) can effectively dissolve perovskite precursors due to their strong coordination with Pb^{2+}, facilitating crystal growth (Park et al., 2023). Although DMF has good dissolving ability, its high boiling point and toxicity pose challenges for large-scale fabrication (Abate et al., 2024). Therefore, researchers have begun exploring green solvent systems that do not contain DMF, such as acetonitrile (ACN)(Sun et al., 2021; Wu et al., 2021; Belay Adugna et al., 2022) and 2-methoxyethanol (2-ME)(Deng et al., 2019; Wang et al., 2020a; Wu et al., 2021; Lee et al., 2022), to optimize the crystallization process and reduce environmental pollution caused by solvents.

The physical properties of solvents, such as their

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volatility, vapor pressure, and boiling point, play a crucial role in the crystallization process of perovskite films. The evaporation rate of the solvent directly affects supersaturation during crystallization. Highly volatile solvents like acetonitrile (ACN) and 2-methoxyethanol (2-ME) can evaporate quickly at room temperature. Due to their fast crystallization rates, they often promote nucleation and crystal growth of perovskite, resulting in a more uniform and dense film structure. However, for scalable processing techniques, such as slot-die coating of perovskite thin films (**Fig. 2a)**, the coating window must be extended to deposit a uniform film, as the process involves key steps like precise control of precursor solution flow, coating speed, and substrate temperature, all of which ensure consistent film thickness and uniform crystallization (Whitaker et al., 2018). In contrast, solvents with lower volatility, such as DMSO, have lower vapor pressure and higher boiling points, which typically lead to slower nucleation and crystal growth rates. This can result in more defects in the film, such as pinholes or grain boundaries, negatively impacting the film's uniformity and density.

For large-area perovskite module fabrication, optimizing the solvent evaporation rate becomes even more critical. In large-scale processes, uniform crystallization across extensive substrates is required, and solvent properties need to be carefully controlled to achieve consistent film morphology. Efficient evaporation management, combined with solvent choice, can significantly improve the yield and uniformity of large-area perovskite films, a vital step toward the commercialization of perovskite-based devices.

Solvent mixtures provide an effective strategy to address this issue,. By precisely controlling the evaporation rate of the solvent, a mixture of volatile non-coordinating solvents and non-volatile coordinating solvents can be used to adjust the solution's evaporation rate and colloid size distribution (Deng et al., 2019; Park, 2021) . For example, mixing DMSO with ACN or 2-ME (Deng et al., 2019; Abate et al., 2024), or combining DMSO with ACN and ethanol (EtOH)(Duan et al., 2024), helps regulate the nucleation rate and crystal growth process, resulting in perovskite films with lower defect density and better optoelectronic properties. The use of EtOH, for example, induces a more uniform distribution of colloidal particles, promoting better film formation and reducing the occurrence of large, irregular crystallites that can impair device performance (**Fig. 2b)**.

The polarity of the solvent and its coordination with the precursor directly influence the solubility and crystallization dynamics of perovskite (Abate et al., 2024). Solvents with low coordination abilities, like ACN, do not excessively coordinate with the perovskite precursor, helping to form smaller colloidal particles and promoting uniform crystal growth. On the other hand, high-coordination solvents such as DMF, while more effective at dissolving the perovskite precursor, may cause excessive coordination, leading to rapid crystal growth and affecting the uniformity and optoelectronic properties of the film.

Note that, in addition to the physical and chemical properties of the solvent itself, the interaction between the solvent and factors such as the substrate, environmental temperature, and humidity can further affect the final quality of the film. For instance, the matching of the solvent evaporation rate with the substrate surface temperature can affect the uniformity and compactness of the film, thereby impacting the crystallization process to variable degrees. Furthermore, while solvent mixtures offer an effective optimization strategy, balancing solvent environmental friendliness and cost-effectiveness while maintaining high-quality films remains a challenge that needs to be addressed. Future research should focus more on the development of green solvent systems and explore how the comprehensive regulation of factors such as solvent properties, solution concentration, and coating speed can further improve the film quality and device performance of perovskite thin films.

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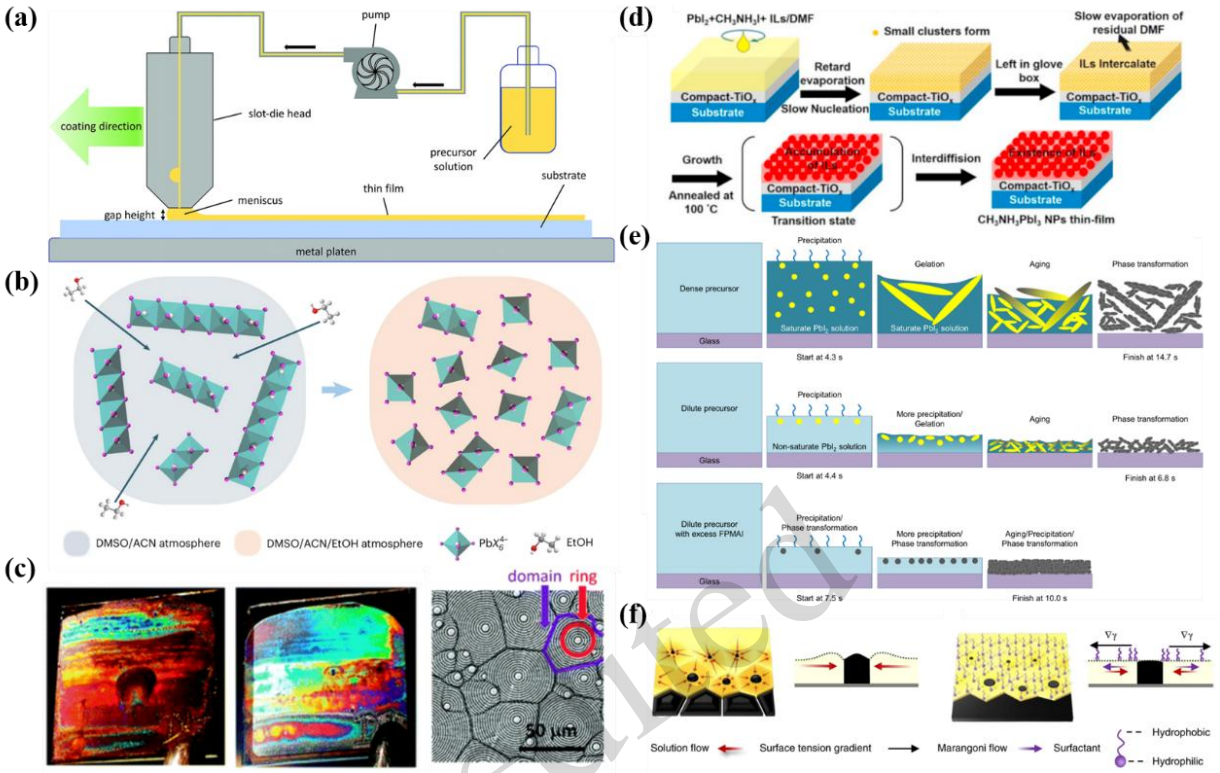

Fig. 2 (a) Schematic illustration of the key steps involved in slot-die coating of perovskite thin films. (reprinted from (Whitaker et al., 2018), Copyright 2018, with permission from Royal Society of Chemistry) (b) Schematic illustration of the colloidal components after adding EtOH to the perovskite precursor solution. (reprinted from (Duan et al., 2024), Copyright 2024, with permission from Springer Nature) (c) Photographs of the prepared colorful organometal trihalide perovskite (OTP) film and the complete device, and a plane view SEM image of a prepared colorful OTP film. (reprinted from (Deng et al., 2015), Copyright 2015, with permission from Royal Society of Chemistry) (d) Schematics of the formation mechanism of $CH_3NH_3PbI_3$ films with ionic liquids. (reprinted from (Shahiduzzaman et al., 2017), Copyright 2017, with permission from Elsevier) (e) Schematics of the sol–gel stages using a dense precursor solution (0.8 M), a dilute precursor solution (0.2 M), and a dilute precursor solution with 50% molar excess FPMAI. (reprinted from (Chu et al., 2021), Copyright 2021, with permission from Springer Nature) (f) Schematic illustration of the directional microscale solution flow towards a perovskite island during ink drying with and without surfactant. (reprinted from (Deng et al., 2018), Copyright 2018, with permission from Springer Nature)

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2.2 Concentration and viscosity

Variation in the solution concentration directly influences the thickness, uniformity, and final crystallization quality of the thin films. A low concentration solution accelerates the solvent evaporation process, causing the perovskite precursor to crystallize quickly during rapid evaporation, resulting in a more uniform film. Deng et al. (Deng et al., 2015) found that when the solution concentration is below a certain threshold, the perovskite thin film forms vibrant, multicolored patterns **(Fig. 2c)**. Such a phenomenon is related to the photonic structures spontaneously formed by crystallizing grains. However, such films may have limitations in crystallization quality and grain size control, with the 'coffee-ring' effect potentially restricting optimization of their optoelectronic properties in practical applications. Nevertheless, low-concentration solutions offer certain advantages in specific applications, such as in flexible or small-area devices, particularly when balancing photoconversion efficiency with ease of preparation, which may provide researchers with new insights.

In contrast, when the solution concentration is high, the film thickness increases, providing more space for the full growth of perovskite crystals. High concentration solutions help increase the wet film thickness, but this also means that the solvent overflow process becomes harder to control.

Excessively high solution concentrations can lead to slower solvent evaporation, impacting the uniformity of the wet film during the drying process and ultimately affecting the crystal growth direction and film quality. Furthermore, high concentration solutions have high viscosities, making it more complex to control the surface tension of the film and the liquid flow during coating. Finding a balance between concentration and viscosity becomes the key to improving film quality.

The viscosity of the solution not only affects the operability of the coating process but also determines how the solution is distributed on the substrate. Shahiduzzaman et al. (Shahiduzzaman et al., 2017) found that the solution's viscosity is closely related to the aggregation process of the perovskite precursor. In the absence of ionic liquids (ILs), the rapid evaporation of DMF during spin-coating causes fast crystallization, resulting in non-uniform morphology (**Fig. 2d**). However, ILs, with their high boiling points and low vapor pressures, slow down crystallization, leading to more controlled nucleation and uniform perovskite films. High viscosity solvents hinder the diffusion of the perovskite solute, whereas low viscosity solvents allow faster dispersion of solute, promoting collisions and aggregation between perovskite molecules, resulting in the formation of uniform nanoparticles and enhancing the crystallization quality of the film. During coating, the flowability and surface tension of the solution directly affect its spreading speed and uniformity on the substrate. Chen et al. (Chen et al., 2022) suggested that high viscosity IL solvents improve the quality of blade-coating and printing because they limit the movement of the perovskite, thereby suppressing the deprotonation of MA/FA (Wang et al., 2020).

The viscosity of the solution not only affects the thickness and uniformity of the film but also plays a crucial role in the grain morphology and growth direction. High viscosity solutions can effectively maintain a high concentration of perovskite solute, promoting crystal growth (Huang et al., 2024a). However, how to avoid excessive aggregation leading to overgrowth of crystals, and how to adjust the grain size and morphology of the film through proper control of viscosity and concentration, remain challenges that need to be addressed.

While many studies have revealed the effects of concentration and viscosity on film quality, how factors such as solution volatility, solvent polarity, and surface tension interact in the actual coating process still requires further investigation. Future research directions should focus on the systematic optimization of solution formulations, exploring new film-forming methods based on solution concentration and viscosity adjustment, especially in terms of achieving efficient and uniform coating in the blade-coating process to optimize the crystallization process of thin films.

2.3 Additives

In the process of preparing perovskite thin films by blade-coating, additives influence the crystallization process of the films through various mechanisms, including regulating the precursor solubility, extending crystallization time, controlling the nucleation process, improving the interface quality, and enhancing the stability of the films.

Solubility regulators, such as lead thiocyanate ($Pb(SCN)_2$)(Ke et al., 2016; Ding et al., 2019) and 1,8-diiodooctane (DIO)(Liang et al., 2014; Chueh et al., 2015; Yang et al., 2015), can enhance the solubility of the perovskite precursor solution (Jang et al., 2022), extending the crystallization time. The prolonged crystallization time allows better control of the blade-coating process, avoiding issues caused by rapid crystallization, such as uneven crystallization, incomplete growth, and solvent residues. The increased solubility provides more time for the precursor molecules to arrange in an ordered manner, which helps achieve a smoother surface and improved crystal quality. In this way, additives can effectively regulate the solvent evaporation rate and solute migration, offering possibilities for optimizing the film's crystal morphology.

Nucleation regulators, especially organic ammonium halide additives (Chu et al., 2021), can effectively suppress excessive grain growth by increasing the density of nucleation centers in the precursor solution. After adding an appropriate amount of organic ammonium halides, the grains of the perovskite thin film typically change from rod-like to spherical, indicating that the grain growth has been effectively controlled, thus avoiding unevenness between grains (**Fig. 2e)** (Chu et al.,

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2021). These additives also shorten the film formation time, preventing excessive grain growth during film formation, further improving the uniformity and density of the film. This regulation of the nucleation process enhances not only the appearance of the film but also its optoelectronic performance.

The interface quality of the film plays a crucial role in its optoelectronic performance, especially at the buried interface of the perovskite layer (Chen et al., 2021a; Chen et al., 2021b; Ni et al., 2022; Zheng et al., 2022). To reduce issues such as non-radiative recombination at the interface, researchers have proposed using additives with strong coordination abilities. These additives accumulate at the interface during the film crystallization process, effectively suppressing harmful interfacial reactions. By adjusting the deprotonation of organic cations in the precursor solution, especially using additives with a pK_a value close to neutral, unordered crystallization can be avoided, thereby optimizing the interface quality of the film (Gao et al., 2024).

Surfactants (Deng et al., 2018; Liu et al., 2020) are another important class of additives. They improve the adhesion between the film and the underlying charge transport layer by altering the film's drying kinetics, thus enhancing the uniformity and smoothness of the film. The presence of surfactants can guide the solution flow in a more controlled manner, helping to prevent the formation of islands and promoting a more homogeneous film structure during the drying process **(Fig. 2f)**. L-α-phosphatidylcholine (LP) is commonly used as a surfactant (Deng et al., 2018). It can effectively passivate charge traps and reduce the formation of island structures, making the film surface smoother and more uniform.

In addition to morphological and performance regulation, stability is also a key consideration in the preparation of perovskite thin films. By introducing stability-enhancing agents such as formamide and MACl (Jang et al., 2022) , issues like voids and wrinkles in the film can be addressed. Formamide can slow the solvent evaporation rate, maintaining the uniformity of the solution and avoiding defects and non-uniformity caused by rapid crystallization of the film. However, due to formamide's high boiling point and its miscibility with DMSO, excessive use may lead to the formation of voids, so its amount must be precisely controlled. To further reduce voids, MACl as an additive can volatilize during annealing, promoting perovskite recrystallization and grain coarsening, ultimately resulting in a dense, void-free film (Williams et al., 2014; Zhao and Zhu, 2014; Zhou et al., 2015; Kim et al., 2019; Odysseas Kosmatos et al., 2019).

Furthermore, ammonium salt additives (Lu et al., 2017; Wang et al., 2017a; Zhang et al., 2017; Wu et al., 2019) not only improve grain growth and increase film density but also enhance the bending resistance and long-term stability of the film through hydrogen bonding interactions between the perovskite particles. Particularly in practical applications, bis-alkylamine (BAA) additives (Wu et al., 2019) can significantly enhance the durability of the film, providing greater assurance for the commercialization of perovskite thin films.

In the context of large-area perovskite module fabrication, additives also play a crucial role in ensuring uniform crystallization across large substrates. By tuning the crystallization rate and enhancing film uniformity, additives enable the production of high-quality, defect-free films that meet the stringent requirements for industrial-scale perovskite device manufacturing.

3 Coating process

3.1 Blade technology

The blade speed directly determines the crystallization behavior of perovskite and the final film structure by affecting the coating characteristics of the solution (Noblesse et al., 2006; Noblesse et al., 2008; Tsai et al., 2015). Fig. 3a and Fig. 3b show that two distinct deposition regimes are identified as a function of blade speed. At lower blade speeds (typically below 4 mm/s), the perovskite thin film enters the evaporation mode (Le Berre et al., 2009; He et al., 2017; Deng et al., 2018). In this mode, the solution rapidly evaporates after coating, and the deposition process of the perovskite precursor is controlled mainly by solvent evaporation. This method effectively prevents excessive wetting of the film surface and solution flow. The lower supersaturation of the solution slows down the

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nucleation process and promotes grain growth (Lee et al., 2020) , resulting in a higher quality perovskite thin film. However, due to the slower coating speed, this mode has low production efficiency and is not suitable for large-scale production (He et al., 2017).

As the blade speed increases (above 20 mm/s), the coating process enters the Landau-Levich mode (Le Berre et al., 2009; Deng et al., 2018). In this mode, after the blade is lifted, the wet layer remains moist, and the solution does not dry quickly, leading to uneven crystallization and large "island" structures on the film surface. This structure not only affects the uniformity of the film but can also form pinholes and defects on the surface, reducing the overall performance of the film. Therefore, while increasing the blade speed can accelerate the coating process, it is crucial to strictly control the drying rate after coating to avoid excessive solution flow and a decline in film quality. During this process, optimizing the blade speed requires finding an appropriate balance between fast coating and film crystallization quality.

In blade-coating, if the solution does not dry immediately, it will flow in different modes driven by surface tension or evaporation, leading to unevenness and discontinuity in the film (Deng et al., 2019). As mentioned earlier (Deng et al., 2018), this solution flow can lead to defects on the film surface and uneven crystallization. To address this issue, one method is to significantly reduce the blade speed, allowing the solution to dry immediately after the blade is lifted. Although this method effectively prevents solution flow and film defects, it limits the coating speed of the blade, which impacts production efficiency (Deng et al., 2019). Alternatively, meniscus-assisted solution printing (MASP) (Fig. 3c) provides a solution to this challenge (He et al., 2017). This technique involves loading metal halide perovskite ink between two nearly parallel plates, with a fixed gap of 300 μm. The capillary forces at the meniscus edge assist in forming large perovskite grains.

Another possible solution is to lower the solution viscosity, as described in Section 2.2, to form a thinner coating film. By adjusting the coating speed and solution concentration, the film thickness can be increased (Deng et al., 2019). This approach makes the coating process more flexible and allows the optimal balance between crystallinity and film thickness to be found according to the needs of the film. Therefore, while focusing on the effects of blade speed and pressure on film crystallization, it is also important to explore the synergistic effects of the process. Optimizing the physical and chemical properties of the solution (such as viscosity and volatility) can help adjust the blade speed and pressure, ultimately achieving the best film quality.

3.2 Substrate wettability

The wettability of the substrate surface directly influences the spreading behavior of the liquid film, the diffusion of the solution, and the nucleation behavior of the thin film, thereby determining the uniformity and optoelectronic performance of the film. The tendency of the liquid film to dewet is closely related to the contact angle of the substrate (Deng et al., 2022). The evolution of the precursor solution droplets on these three hole transport layers (HTLs) shows how the contact angle influences the spreading behavior of the solution over time **(Fig. 3d)**. This dynamic process further highlights how the interaction between the precursor solution and substrate determines the film's uniformity and coverage as the droplet spreads, affecting the final perovskite film morphology. The contact angle of the substrate determines how the liquid film spreads, which in turn affects the crystallization quality of the film. If the substrate has poor wettability (i.e., a high contact angle), the liquid film may dewet quickly (Vachaparambil and Einarsrud, 2018; Deng et al., 2022), leading to uneven films and impacting the uniformity of crystallization and the orientation of the grains. Changes in the contact angle significantly affect the rate at which the liquid film dewets. According to Edwards et al. (2016), the rate of dewetting of the liquid film is sensitive to the equilibrium contact angle (**Fig. 3e)**. In particular, under surface tension, substrates with poor wettability make it easier for the solution to dewet, causing unevenness during the crystallization process. Therefore, by adjusting the wettability of the substrate, the behavior of the liquid film during the coating process can be optimized, improving the uniformity and crystallization quality of the film.

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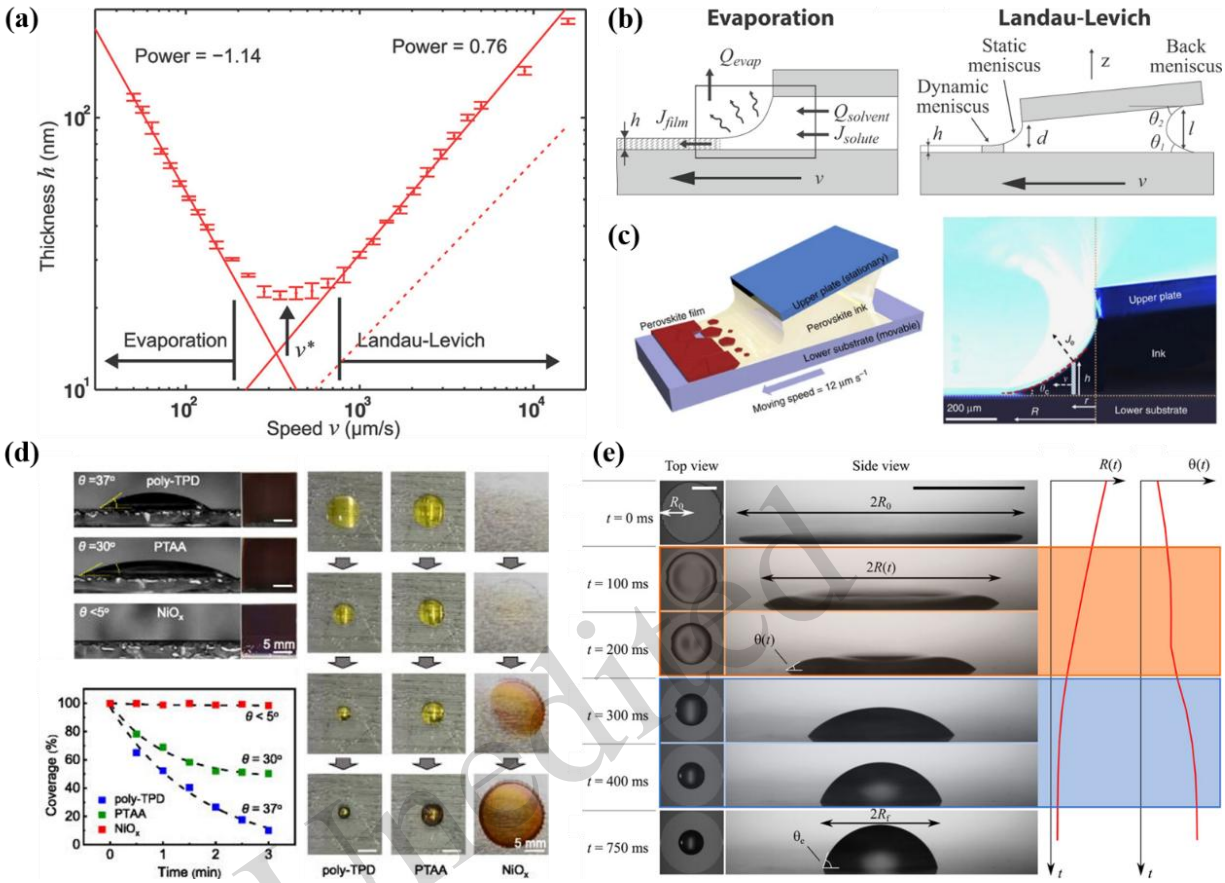

Fig. 3 (a) Measured film thickness *h* as a function of deposition speed *v*. Two regimes are identified as a function of *v*: the evaporation and Landau−Levich modes. (reprinted from (Le Berre et al., 2009), Copyright 2009, with permission from American Chemical Society) (b) Schematic of the two deposition regimes. (reprinted from (Le Berre et al., 2009), Copyright 2009, with permission from American Chemical Society) (c) Schematic illustration of the meniscus-assisted solution printing (MASP) of large-grained perovskite films and optical micrograph of the side-view meniscus ink. (reprinted from (He et al., 2017), Copyright 2017, with permission from Springer Nature) (d) Contact angle of precursor solution on PFN-Br treated poly-TPD, PTAA, and NiO_x HTLs, and corresponding photographs of perovskite films. Evolution of precursor solution droplets on the three HTLs, and the corresponding coverage of droplets against time. (reprinted from (Deng et al., 2022), Copyright 2022, with permission from American Institute of Physics) (e) Experimental imaging of the dewetting of a liquid droplet from a smooth solid surface. (reprinted from (Edwards et al., 2016), Copyright 2016, with permission from AAAS)

During the coating process, the diffusion of the solution is closely related to the wettability of the substrate. For example, in large-area perovskite film preparation, especially on TCO (transparent conductive oxide) substrates, the solution often first fills the nanoscale valleys on the TCO surface, leading to multilayer stacking and affecting the uniformity of the coating (Phung et al., 2022; Ren et al., 2023). Wang et al. (Wang et al., 2024) improved the wettability of the substrate using poly-DBPP and DBPP, enhancing the diffusion of the solution and improving the distribution of perovskite ink, which ultimately improved the uniformity and optoelectronic performance of the film. The strong bonding between poly-DBPP and the ITO substrate significantly reduced the contact angle, further enhancing the interaction between the substrate and the solution, thereby improving the crystallinity of the film. Note that substrate wettability can lead to the coffee-ring effect (Yu et al., 2019), which can impact the uniformity of the film and device performance. The coffee-ring effect is caused by capillary flow during the evaporation of a droplet, which typically leads to the accumulation of solutes at the edges of the film, forming a ring-shaped deposit. By improving the wettability of the substrate (Liu et al., 2024),

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particularly using substrates with stronger hydrophilicity, this effect can be reduced, allowing solutes to be more evenly deposited on the substrate, thereby improving the uniformity of the film. The coffee-ring effect can also be suppressed by altering the solvent system and lowering the solution viscosity (Hu and Larson, 2005; Abo Jabal et al., 2017; Malinowski et al., 2018; Yu et al., 2019; Hu et al., 2020; Wilkinson et al., 2021; Yang et al., 2022).

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To further optimize the quality of perovskite thin films, researchers have recently proposed several methods to improve wettability through substrate surface treatments. Introducing self-assembled monolayers (SAMs) or other interface modification layers on the substrate can significantly adjust the substrate's wettability, thereby controlling the spreading and crystallization behavior of the liquid film. These surface modification techniques, by adjusting the surface energy of the substrate, can promote the oriented growth of perovskite crystals, reduce defects, and enhance the crystallization quality and device performance of the film.

3.3 Gas-quenching

The main challenge of the blade-coating method lies in the slow solvent evaporation rate, which prevents the solution from quickly reaching supersaturation and initiating crystallization (Yu et al., 2021). Auxiliary crystallization methods, such as anti-solvent quenching (Deng et al., 2015a; Zeng et al., 2020), vacuum flash evaporation (Li et al., 2016; Zeng et al., 2020), and gas-quenching, can rapidly remove solvents to reach supersaturation, thus promoting nucleation and crystal growth. Among these, gas-quenching, as a widely applied crystallization assistance method, has shown significant potential for industrial-scale production (Wu et al., 2018; Cheng et al., 2019; Sun et al., 2019; Zhang et al., 2019; Szostak et al., 2021). Gas-quenching induces nucleation and crystal growth by extracting solvents from the wet precursor film through strong convection. This method offers notable advantages, especially as it does not require complex equipment or toxic chemicals, showcasing good stability and reproducibility. After gas-quenching treatment, the blade-coated films exhibit a more uniform surface, larger grain size, and significantly reduced defect density, thereby greatly improving the film quality and optoelectronic performance (Razza et al., 2015; Ke et al., 2016; Song et al., 2017a; Deng et al., 2019; Ding et al., 2019).

The key to controlling gas-quenching crystallization lies in the distance between the air knife and the liquid film. If the air knife is too close to the liquid film, the solvent evaporation rate on the liquid film surface will be too fast, leading to an uneven perovskite film. On the other hand, if the air knife is too far from the liquid film, solvent diffusion will be slow, causing the solution to reach supersaturation at a slower rate, and the liquid film may fail to dry completely (Yu et al., 2021). Therefore, optimizing the gas-quenching process depends on precisely controlling the airflow velocity and the distance between the air knife and the liquid film to achieve supersaturation in a short time, promoting high-quality nucleation and crystal growth. Gao et al. (2017) used a multi-stream air knife (MAK) to compensate for the limitations of a single air knife (Gao et al., 2017a) . By optimizing the airflow velocity and temperature, they were able to obtain more uniform and dense perovskite films (Gao et al., 2017b). This improvement not only enhanced the quality of the films but also provided a more stable and efficient crystallization control method for industrial-scale production.

However, further research is needed to enhance the adaptability of the gas-quenching method to different solution formulations and substrate surfaces, as well as to optimize the film formation process by

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precisely controlling airflow parameters. For example, during the gas-quenching process, local over-drying or cooling may occur, affecting the overall film quality. To address this issue, future studies should focus on optimizing airflow distribution, temperature control, and airflow direction. Additionally, combining gas-quenching with other film formation methods (such as anti-solvent quenching or vacuum flash evaporation) may offer more effective strategies for improving the crystallization quality of perovskite films and their device performance.

4 Surface passivation

Passivation techniques play a crucial role in improving the crystallization quality of perovskite films, reducing surface defect densities, and enhancing their overall stability. The effects of passivation are not limited to increasing grain size but extend to optimizing the film structure by reducing surface defects, improving crystal order, and minimizing non-radiative recombination losses (Stolterfoht et al., 2019; Wang et al., 2019; Caprioglio et al., 2023). In the blade-coating method, surface passivation of perovskite films does not necessarily require the development of a specific passivation technique for the fabrication process.

One of the main approaches to tuning energy levels in perovskites is the incorporation of lithium fluoride (LiF) or magnesium fluoride (MgF_x) into both the perovskite and the electron transport layers (ETLs), which helps reduce non-radiative recombination (Stolterfoht et al., 2018; Peña-Camargo et al., 2020; Liu et al., 2022). This approach effectively aligns the energy levels between the perovskite and the ETL, facilitating better charge extraction and reducing recombination losses (**Fig. 4a**). Interfacial capping layers, such as ammonium ligands or functional reactive compounds, are commonly used to passivate surface defects of perovskite films (Li et al., 2020; Li et al., 2022; Zhang et al., 2022; Chen et al., 2023; Zhou et al., 2023; Huang et al., 2024b; Liu et al., 2024b). These capping layers serve to protect the surface, effectively mitigating defect-related issues that could negatively impact device performance.

For enhancing the humidity stability of perovskite films, passivating agents like the MABr/MACl dual-passivation system (Kim et al., 2023) have shown significant effects. This dual-passivation system notably suppresses interfacial defects, especially in environments with fluctuating humidity levels, and effectively reduces the negative impact of humidity on the film. It also plays a role in improving the interface stability, which is essential for enhancing the overall longevity and performance of perovskite-based devices.

Introducing two-dimensional (2D) perovskite layers onto the surface of the film, especially to passivate the surface defects of three-dimensional (3D) perovskite layers, can further enhance both the stability and optoelectronic performance of the film (Tian et al., 2022; Teale et al., 2024). The formation of 2D perovskite layers can either directly or indirectly cover the surface of the 3D perovskite. Through passivation, this process reduces the surface defect density, thereby improving the film's stability and photonic response. Two common methods for 2D perovskite layer passivation include direct and indirect approaches (Teale et al., 2024). The direct method typically involves physically stacking or using solvent-selective dissolution techniques to transfer the 2D perovskite layer onto the surface of the 3D perovskite. The indirect method, on the other hand, involves applying passivating ligands to the surface of the perovskite, followed by an annealing process that gradually forms the 2D perovskite layer (Quan et al., 2016). This method allows for the continuous adjustment and formation of a more ordered 2D perovskite layer at the surface of the perovskite's dimensionality by varying the n value (**Fig. 4b**). This effectively reduces surface defects and improves film quality thereby enhancing the stability and optoelectronic properties of the material.

The passivation of perovskite films using inorganic materials such as phosphate and sulfate ions has also been shown to be crucial in improving film quality (Yang et al., 2019). The in-situ formation of a lead sulfate ($PbSO_4$) top layer on the perovskite surface plays a key role in protecting the material (**Fig. 4c**). This inorganic passivation layer stabilizes the surface of the perovskite by reacting with it, reducing defect densities, and significantly enhancing the film's resistance to moisture and chemical

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degradation in humid environments. As a result, these inorganic passivation layers contribute to the long-term stability of the films, making them more suitable for practical applications.

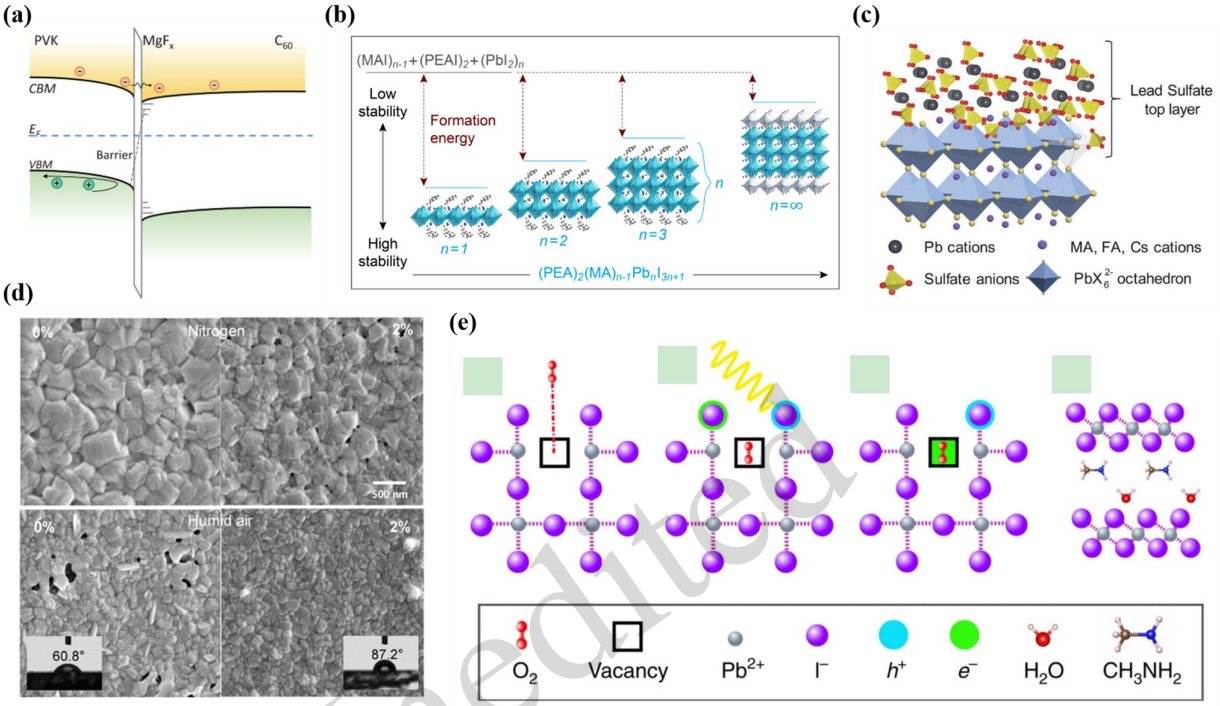

Fig. 4 (a) Energy level diagram of the perovskite/C_{60} interface with an MgF_x insertion layer. (reprinted from (Liu et al., 2022), Copyright 2022, with permission from AAAS) (b) Unit cell structure of perovskites with different n values. (reprinted from (Quan et al., 2016), Copyright 2016, with permission from American Chemical Society) (c) Schematic illustration of the protection of perovskites through in situ formation of a lead sulfate top layer on the perovskite surface. (reprinted from (Yang et al., 2019), Copyright 2019, with permission from Springer Nature) (d) SEM images of perovskite films with 0% and 2% [bvbim]Cl (the passivate) molar content prepared in nitrogen or humid air conditions, and contact angle measurement observed for films prepared in humid air. (reprinted from (Xia et al., 2020), Copyright 2020, with permission from Wiley) (e) Schematic representation of the reaction steps of O_2 with $CH_3NH_3PbI_3$. (reprinted from (Aristidou et al., 2017), Copyright 2017, with permission from Springer Nature)

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In conclusion, passivation remains one of the most effective strategies for enhancing the performance and durability of perovskite films. By carefully selecting and applying passivating agents, the structural and functional properties of the films can be optimized, leading to higher-quality, more stable perovskite-based devices that are better suited for large-scale, commercial applications.

5 Atmosphere

The blade-coating method for perovskite film preparation in industrial-scale production is typically carried out in ambient conditions, as solution processing within a glovebox is not feasible for commercial applications (Ko et al., 2015; Yang et al., 2015c; Lee et al., 2017; Song et al., 2017b; Fu et al., 2022). However, environmental factors such as humidity, temperature, and oxygen levels can significantly influence both the crystallization quality of the perovskite films and the performance of the resulting devices. Therefore, extra attention must be given to the impact of these environmental conditions on the crystallization process during blade-coating.

Perovskite materials, especially organic halides like methylammonium iodide (MAI) and formamidinium iodide (FAI), are highly hygroscopic (Bass et al., 2014; Christians et al., 2015; Leguy et al., 2015; Yang et al., 2015a; Zhang et al., 2015; Wang et al., 2016; Niu et al., 2017; Xu et al., 2018). In the blade-coating process, variations in humidity directly

affect the stability of the perovskite precursor solution. When humidity is high, moisture can promote hydrolytic reactions of the perovskite, leading to the degradation of organic halides and the formation of PbI_2(Niu et al., 2014; Niu et al., 2015; Yang et al., 2015c; Huang et al., 2017; Xia et al., 2020). **Fig. 4d** shows the impact of humidity on the film morphology. Films exposed to humid air show increased defects and uneven grain structures, which negatively affect the crystallization quality. In particular, during the crystallization process, the presence of water can cause volume defects in the perovskite crystals (Jang et al., 2022), which compromises the film's crystallization quality. The impact of water causes instability in the crystallization speed, leading to smaller grain sizes and an increase in defect density within the perovskite films.

Humidity is closely linked to the evaporation rate of the solvent during the blade-coating process. In a high-humidity environment, the slower evaporation of the solvent results in uneven crystallization on the surface of the perovskite film. This unevenness can create defects such as pinholes and bubbles in the film, which further degrade its optoelectronic performance. In contrast, in low humidity conditions, blade-coated perovskite films generally exhibit better uniformity and low surface defect densities, leading to improved crystallization quality and device efficiency (Yang et al., 2015c).

Temperature is another critical factor influencing perovskite film crystallization during the blade-coating process. Elevated temperatures generally aid in the evaporation of the solvent, thereby accelerating the crystallization process of the film. However, if the temperature is too high, it can lead to rapid solvent evaporation, causing the solid-liquid interface to form prematurely at the solution's surface, which results in uneven vertical growth of the film. Additionally, this can leave solvent residues behind, negatively affecting the crystallization quality of the film. Therefore, precise temperature control is essential to ensure uniform grain growth, particularly during the blade-coating process, where managing temperature is vital to reduce crystallization defects.

Light exposure and the presence of oxygen also affect the crystallization of perovskite films during blade-coating. Perovskite materials are highly sensitive to light and oxygen, especially methylammonium-based perovskites ($MAPbI_3$), which are prone to degradation under the combined effects of light and oxygen (Aristidou et al., 2015; O'mahony et al., 2015; Bryant et al., 2016). When films are exposed to strong light or oxygen during the blade-coating process, the perovskite material may undergo photooxidative degradation, forming peroxides, which can further deteriorate the quality of the material (Bryant et al., 2016). Particularly during the crystallization of the precursor solution, oxygen and light can facilitate the deprotonation of the organic cations in the perovskite, leading to the formation of PbI_2 and consequently compromising the integrity and stability of the perovskite crystals (**Fig. 4e**).

To mitigate the negative impact of these environmental factors on the crystallization process, many researchers are exploring controlled environment blade-coating methods. For instance, performing blade-coating in a dry atmosphere can effectively prevent degradation due to moisture. Additionally, using UV-blocking materials to shield the perovskite film from direct light exposure can significantly improve the crystallization quality and stability of the films. These measures help create a more stable environment for the perovskite crystallization process, ensuring better film quality and enhanced device performance in industrial applications.

In conclusion, understanding and controlling environmental factors during the blade-coating process are essential for improving the crystallization quality and stability of perovskite films. By mitigating the effects of humidity, temperature, and light exposure, it is possible to achieve high-quality films with better long-term performance and reliability, paving the way for large-scale industrial production of perovskite-based devices.

6 Conclusions and Perspective

Blade-coating, holds significant potential as a promising large-scale and low-cost technique for fabricating perovskite films but faces several key challenges, particularly in controlling the

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crystallization process and ensuring uniform film quality. As discussed in this review, various factors that influence perovskite film crystallization—such as the precursor solution, coating process, substrate conditions, and environmental factors—need to be optimized to minimize defect density and achieve uniform and consistent film morphology on large-area substrates.

During blade-coating, the crystallization of perovskite films is affected by multiple parameters, with uneven evaporation rate being one of the main issues. Differences in evaporation rates across different areas can lead to non-uniform film thickness and may result in island-like perovskite growth (Deng et al., 2015a; Han et al., 2025). If not properly controlled, these islands can cause pinholes that allow direct contact between the ETL and HTL, which in turn generates leakage currents and degrades device performance (Ye et al., 2016; Hu et al., 2017; Deng et al., 2018; Rong et al., 2018). The crystallization dynamics vary significantly between perovskites with different composition, such as $MAPbI_3$ and FACs perovskites. $MAPbI_3$-based films are relatively more mature for large-area coating, but $MAPbI_3$ is more sensitive to environmental factors. In contrast, FACs perovskites, where the iodide (I^-) is partially replaced by bromide (Br^-), have a higher diffusion rate of Br^- in solvents than I^-(Park, 2016), which leads to different crystallization rates and phase segregation, affecting the stability and long-term performance of the films.

As the size of perovskite solar cell modules increases, efficiency losses are influenced by multiple factors, including the increase in series resistance, decrease in shunt resistance, large-area coating non-uniformities, and dead zones (non-active areas) in busbars and interconnection electrodes (Li et al., 2018). During large-area coating, variations in precursor solution concentration, differences in solvent evaporation rates, and temperature gradients further exacerbate non-uniformities in the crystallization process, making it increasingly difficult to prepare high-quality films.

The main factors influencing perovskite film crystallization include solvent properties, solution concentration, viscosity, and the use of additives. Solvents, through their polarity and volatility, regulate the crystallization dynamics, affecting nucleation and grain growth. Solution concentration and viscosity impact film thickness, uniformity, and grain morphology. High-concentration solutions promote thicker films but may result in slower evaporation and more defects, while low-concentration solutions offer better control over crystallization but can lead to insufficient film formation. Additives are also critical in modulating the crystallization process, controlling nucleation rates, improving film morphology, and enhancing stability by regulating solubility, interface quality, and grain size. A balanced combination of these factors is essential to achieve optimal perovskite film quality and long-term performance.

Despite these challenges, significant progress has been made in recent years in enhancing the scalability and uniformity of perovskite films. By optimizing precursor solution formulations, including the use of additives and solvent combinations, researchers have been able to exert more control over the crystallization process, improving film morphology, reducing defect density, and enhancing the stability of perovskite films. However, the selection and concentration of ideal additives remain heated topics in current research, as the effects of additives can vary depending on the solvent system and perovskite composition.

The key to improving blade-coating technology lies in a deeper understanding of the crystallization mechanism of perovskite films. Future research should focus on revealing the kinetics of perovskite film formation, particularly how solvent evaporation and precursor concentration gradients affect the crystallization process. In addition, developing advanced in-situ monitoring techniques to track the crystallization process in real-time will be critical for optimizing blade-coating processes and achieving more reproducible results. Research into novel perovskite compositions, such as mixed-cation or mixed-halide systems, may also help develop films with superior crystallization characteristics and stability, which is crucial for advancing the commercialization of perovskite solar cells.

Moreover, transitioning to more stable and efficient perovskite compositions still requires further optimization of the blade-coating process to develop universal coating methods that can adapt to their different crystallization dynamics and solvent

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requirements. Precisely controlling the crystallization and growth of these perovskite films on large-area substrates will be a key step toward achieving commercial viability.

In addition to improving film quality, developing reliable post-treatment techniques such as annealing and drying strategies and passivation can effectively reduce solvent residues, minimize defects, and ensure the long-term stability of perovskite solar cells. As blade-coating technology continues to evolve, integrating these advancements with large-scale, automated production lines to achieve competitive manufacturing of high-quality perovskite films will be a crucial area for future research.

In conclusion, the blade-coating method holds immense potential for the fabrication of perovskite films, especially in large-scale production. However, to successfully realize this potential, several key challenges, including film uniformity, defect control, and scalability, must be addressed. With continued advancements in solution chemistry, process optimization, and a deeper understanding of crystallization mechanisms, the efficiency and stability of blade-coated perovskite photovoltaics will significantly improve, paving the way for the large-scale application of perovskite solar cells and other perovskite-based optoelectronic devices.

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Acknowledgments

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Author contributions

Jingyi SUN and Borui LEI wrote the first draft of the manuscript. Jingjing XUE, Jingyi SUN and Borui LEI revised and edited the final version. All authors approved the final manuscript.

Conflict of interest

Jingyi SUN, Borui LEI, and Jingjing XUE declare that they have no conflict of interest.

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中文概要

题　目：影响刮涂过程中钙钛矿薄膜结晶的关键因素

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概　要：钙钛矿薄膜由于其优异的光电性能和低成本的制造工艺，在光电器件领域，特别是太阳能电池领域取得了重大进展。作为一种很有前景的钙钛矿薄膜大规模工业生产技术，刮涂法因其成本低、大面积涂布能力强、操作简单等优点而受到广泛关注。然而，在涂布过程中，钙钛矿薄膜往往存在薄膜均匀性差、缺陷密度高、结晶不均匀等问题，极大地影响了器件的效率和稳定性。本文综述了刮涂法在钙钛矿薄膜制备中的应用，重点分析了影响薄膜质量的主要因素，包括前驱体溶液配方、溶剂特性、蒸发速率、结晶动力学和薄膜厚度等。特别地，我们讨论了环境因素对刮涂过程中钙钛矿结晶的影响，并探讨了如何优化刮涂工艺和前驱体溶液组成来提高薄膜均匀性和器件性能。此外，我们还讨论了刮涂法制备钙钛矿薄膜面临的主要挑战和缺点，包括大面积制备过程中的缺陷，溶剂蒸发速率的差异以及它们对结晶质量的影响，并提出了解决这些问题的方法。最后，我们展望了刮涂法在钙钛矿薄膜大规模生产中的应用前景，强调了深入了解钙钛矿薄膜结晶机理和开发新型添加剂对提高钙钛矿光电器件性能和推动其产业化的重要性。

关键词：钙钛矿薄膜；刮涂法；晶体生长

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