Processing and Preparation Method for High-Quality Opto-Electronic Perovskite Film

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The key to improving the energy conversion efficiency of perovskite solar cells lies in the optimization of the film morphology. The optical and electrical properties of the perovskite film, such as light absorption, carrier diffusion length, and charge transport, are all directly affected by the film morphology. Therefore, this review starts from the perovskite solar cells structure, and it summarizes the state-of-art perovskite film fabrication technologies and the caused film morphology to the performance perovskite solar cells. The spin coating method has an enormous waste of materials and only a small area of the device can be utilized. It is difficult to be used in commercial manufacturing. However, due to the high efficiency of this preparation method, it is irreplaceable in the initial research and development of perovskite materials, and so this method will be popular for a long time in the laboratory. Chemical vapor deposition and thermal vapor deposition have high technical requirements and a good repeatability of processing and manufacturing, and large-scale production can be realized. It may be the first technology to admit industrial application; the scratch coating method and slot-die have significant technical aspects. The similarity of the roll-to-roll manufacturing technology is also an efficient preparation method. Still, to achieve high-efficiency devices, it is necessary to consider the thickness control of each functional layer, and to find or prepare perovskite paste. Finally, we summarized the various fabrication processes and the prospects for the commercialization of perovskite solar cells. We predict that to achieve the commercialization of perovskite solar cells, the existing fabrication technologies should be optimized and more studies should be conducted.

Keywords: perovskite, fabrication, solar cell, large scale, roll to roll

1 INTRODUCTION

Perovskite has the general formula of ABX₃, where A is an organic or metal cation (e.g., MA⁺, FA⁺, Cs⁺), B is a metal cation (e.g., Pb²⁺, Sn²⁺), and X is a halogen (e.g., Cl⁻, Br⁻, I⁻). Perovskite has superior optoelectronic properties and lower crystallization activation energy, when compared to silicon (Elseman et al., 2019; Selim et al., 2020). Perovskite has gained enormous attention during the past decades because of its superior electrical and optical properties and has great potential in next-generation solar cells (De Wolf et al., 2014; Snaith, 2018). The basic intent behind photovoltaic (PV) products is always to achieve engender energy and its massive scale appropriateness. Nonetheless,
due to the proportions of the PV unit, several challenges are experienced that are not found in smaller devices (Kajal et al., 2018). The perovskite solar cells (PSCs) are first developed from DSSC studies because of the undeniable fact that there could be no element oxide scaffold. The fundamental device architectures are shown in Figure 1. The basic requirement is the presence of interface layers which could be charge selective in nature, considered one of them for holes and one of them for electrons (Sajid et al., 2019). Some renowned interface layers which work much better in the area of PSCs are PTAA, PEDOT, PSS, CuSCN, etc. (Elseman et al., 2020a). On the other hand for electron transports, metal oxides which include tin oxide work thoroughly as the electron transport layer (kour et al., 2018). The charge that is photo-generated gets used in the electron and hole interface layers through the perovskite layer following absorption of light. The typical structure has an absorbing layer comprised of a material that is perovskite with a depth of 300–500 nm (Bansal and Aryal, 2016; Xu et al., 2020). It is tough to regulate the morphology of perovskites over a considerable region. The morphology of perovskite materials could affect the light absorption. The typical structure of a PSC has a flat layer of perovskite and the mild scattering effect on the interface is quite constrained. With the enhancement of the power conversion efficiency (PCE), the various fabrication process have been investigated (Yang et al., 2018a). There is another means to fix the morphology regarding the perovskite layer which could increase the optoelectronic properties. Cheng et al. developed a surface-textured perovskite film utilizing gas-assisted fabrication strategy, a textured CH$_3$NH$_3$PbI$_3$ morphology formed via a thin mesoporous TiO$_2$ layer that is seeding. This textured morphology includes a multilayered nanostructure, which provides enormous upgrades in the charge extraction and light-harvesting via just imprinted procedure with robust microstructure stamp for PSCs. The WG-dependent perovskite films can acquire light trapping by optical feedback and steady absorption. This procedure can likewise quicken electron-hole separation and suppress recombination. The advanced WG device shows negligible J-V hysteresis (Wang et al., 2018). There are also some real issues that need to be dealt with including the quality and thickness of the film during the fabrication (Han et al., 2015; Li et al., 2015). The manufacturing techniques should be chosen cautiously (Yang et al., 2017a).

It is well known that the fabrication process can directly influence the morphology of perovskite films, thus, it is worthwhile to summarize the various fabrication processes and the related film morphology (Elseman et al., 2020a; Elseman et al., 2020b). In this manuscript, we summarized the state-of-the-art fabrication processes and the effects on the perovskite film morphology. Furthermore, the overall device performance was also investigated, including efficiency, and stability. Finally, the prospect of the commercialization of perovskite photovoltaic applications is discussed based on the various fabrication process. The one-step and two-step sequence deposition methods are mostly investigated in lab experiments to achieve higher efficiency and to reveal the superiority of PSCs to improve the development and to gain more public attention. As the basic investigation of PSCs in the lab, the achievements could promote the development of the commercialization of PSCs. Enormous efforts have been made during the past years and great achievements have been made in lab experiments. However, the efficiency of large-scale perovskite photovoltaic devices still falls far behind that of small size (<0.1 cm$^2$) perovskite devices. It is necessary to summarize the recent achievements in large-scale fabrication processes to inspire further investigations. Therefore, we describe the perovskite solar cells in detail from the following aspects: 1) The basic investigation of the perovskite crystallization
process, which includes one-step and two-step methods. 2) This part is the main content of the article. We discuss in detail the preparation methods and processes of large-area perovskites, such as vapor-assisted solution method, thermal vapor deposition, roll-to-roll fabrication techniques, inkjet printing, doctor-blade coating, slot-die coating, and spray-coating. These methods are the mainstream methods in the current research work, and we hope that the summary of these works will be helpful to the work of researchers in the perovskite field.

2 DEPOSITION METHODS

Fabrication strategies are divided into vapor assisted processing and wet chemistry processing. The wet chemistry processing methods are further categorized into one-step deposition techniques and two-step deposition techniques. Vapor assisted processing approaches additionally have exclusive processing strategies which include thermal vapor deposition (TVD) and vapor assisted solution procedure (VASP) (Barrows et al., 2014; Chen et al., 2014). In the following section, we will discuss the various fabrication process and the effects on the film morphology of the perovskite. More importantly, the related device performance is emphasized. We try to elaborate on the advantages and disadvantages of the various fabrication processes and evaluate the prospects for large-scale fabrication to realize the commercialization of PSCs.

2.1 The Basic Investigation of the Perovskite Crystallization Process

The final performance of the perovskite photovoltaic devices was almost totally dependant on the quality of the perovskite film. Thus, to obtain the high-quality perovskite film was highly desirable. In the following part of the text, we will discuss the crystallization process of one-step and two-step fabrication processes of perovskite photovoltaic devices.

2.1.1 One-Step Method

The one-step fabrication process has been widely studied and the investigation of the crystallization mechanism would inspire the large-scale fabrication process. The general crystallization process involves three steps: 1) the supersaturation of solution; 2) the formation of nuclei; 3) the growth of crystal (Jeon et al., 2015; Hsieh et al., 2018). With the assistance of the anti-solvent and the evaporation of the solvent, the solution is supersaturated, then the nucleation process starts, followed by the consumption of solute, and the start of the crystal growth. Previous investigations of the one-step fabrication process give the fundamental cognition of the crystallization process and it is worthwhile to be elaborated (Xiao et al., 2014a; Gardner et al., 2016; Hendriks et al., 2017).

The one-step method is commonly employed to deposit perovskite film because it is not only cheap in manufacturing but also additionally accessible in processing. Before spin coating, organic, and inorganic elements are mixed and dissolved in a polar solvent like dimethylsulfoxide (DMSO) or dimethylformamide (DMF) to form the precursor pigment. In some cases, the perovskite film will be evolved via spin coating of the pigment on a substrate, accompanied by post-annealing at 70–150°C (Roy et al., 2020). Due to the high boiling point and low vapor pressure and the annealing process, the solvent will evaporate and the perovskite structure could be formed. Gratzel and his group tested the results of the annealing temperature at the formation of CH3NH3PbI3 perovskite films by the one-step approach. They observed the coverage of the film increased against annealing temperature ranging 60–100°C (Pellet et al., 2014). The PSCs that annealed on the absorber layer at 100°C yield the highest efficiency (Zuo et al., 2018; Arain et al., 2019). The first PSC was developed by Kojima and their group in 2009, and they investigated the optical properties of methylammonium lead iodide to make mixed halide or iodide only (Kojima et al., 2009). Organic halides (CH3NH3X) and lead halides (PbX2) had been dissolved in GBL, dimethylformamide DMSO or DMF in the nitrogen glove box to prepare a methylammonium lead halide (CH3NH3PbX3) solution as shown in the following equation.

\[ \text{PbX}_2 + \text{CH}_3\text{NH}_3\text{X} \xrightarrow{\text{Organic Solvent, Temp, Stiring}} \text{CH}_3\text{NH}_3\text{PbX}_3 \]

The perovskite film was deposited via spin coating at the top of TiO2 layer as shown in Figure 2 (Ahmadian-Yazdi et al., 2016; Jamal et al., 2018).

There is a drawback in the one-step method on a large area is better film coverage and uniform thickness of the layer. Anti-solvent precipitation is a well-known technique in chemistry to crystallize a material. The best thing is that this method is applied to the manufacturing of perovskite solar cells by using an anti-solvent dropping method. This approach became first reported through Jeon and his crew; they observed that using an anti-solvent in the fabrication of perovskite film produced first-class and dense films (Jeon et al., 2014). However, the anti-solvent preparation process should be conducted at right time and the volume should be precisely controlled. Besides, this anti-solvent step requires high efficiency. Furthermore, uncontrolled crystallization can form pin-holes which could damage the efficiency and stability of the devices (Zuo et al., 2018; Arain et al., 2019).

In addition to the anti-solvent method to accelerate the crystallization, the gas-assisted crystallization process was also investigated. Nie et al. (2015) developed the hot-casting process to accelerate the evaporation of the high-boiling solvent to render a high-quality perovskite film and a tunable perovskite morphology by tuning the temperature of the substrate. A vacuum-assisted solvent evaporation method was elaborated by Li et al. (2016). After the spin-coating process, the substrate was placed in the chamber under a vacuum environment to achieve the rapid evaporation of the solvent followed by the annealing process. Finally, a certified PCE of 19.6% was achieved under the active area over 1 cm². However, the rapid evaporation process resulted in a faster nucleation rate thus decreasing the grain size. A deeper understanding of the crystallization mechanism should be sought to balance the solvent evaporation process and the crystallization process.

Though the one-step fabrication process delivers a feasible fabrication technique and high efficiency, the simple one-step
spin-coating is not suitable for large-scale fabrication since a uniform and homogeneous perovskite film cannot be obtained. Luckily, the one-step fabrication process is popular in lab-scale investigations because of the simple preparation, and less requirement could deliver a good performance device which is beneficial for the future commercialization of PSCs.

2.1.2 Two-Step Method
To solve the uncontrollable film morphology fabricated by the one-step fabrication process, the two-step method was investigated. Usually, the one-step deposition method uses a combination of MAX and PbX2 that ends in the out-of-control precipitation of the perovskite, which leads to significant morphological variation that prevent the possibilities for commercial applications. Consequently, low-temperature two-step deposition methods become more advanced (Bi et al., 2013; Burschka et al., 2013). The two-step deposition approach is an excellent technique to clear up issues resulting from the dipping approach. In a two-step deposition approach, an inorganic solution is deposited on a substrate with the spin coating technique (Jiang et al., 2016). Consequently, the coated substrate is immersed into a second organic solution accompanied by annealing (Bi et al., 2014; Yang et al., 2017b). Take the well-studied perovskite MAPbI3 for instance: firstly, the PbI2 was spin-coated at the substrate; secondly, the resulting film was spin-coated with MAI solution or immersed in MAI solution. Followed by the annealing process, a high-quality perovskite film could be achieved. Furthermore, the deposition of methylammonium iodide (MAI) solution via spin coating over a PbI2 film has been employed followed by means of annealing as shown in Figure 3 (Im et al., 2014).

Though the sequence deposition process can deliver the full coverage film and large grain scale, however, the resultant film has a high surface roughness which will cause surface recombination loss (Yang et al., 2015; Yang et al., 2018b). Some researchers found that incomplete conversion of perovskite is a major issue in the two-step deposition method. The transformation speed of PbI2 to MAPbI3 is quite suitable after the film is immersed into the solution, due to the structural association of heavy metal halides that are inclined to engage with small molecules. Consequently, a compact perovskite layer is shaped on the pinnacle of PbI2 and the movement of MAI to the lowest layer is difficult, which becomes a cause of partial perovskite transformation (Coleman et al., 2011; Olaleru et al., 2020). Wu et al. solve this problem by introducing recently developed techniques which include solvent engineering and adding precursors to some additives like DMSO or H2O that can prevent the formation of a compact perovskite layer on the pinnacle of PbI2 (Wu et al., 2014; Wu et al., 2015; Olaleru et al., 2020). The strong coordination effect was observed between DMSO and Pb2+ ions to slow the crystallization of PbI2 in forming the amorphous PbI2 film, which could accelerate the complete conversion of the perovskite phase.

The one-and-a-half-step method combines the advantages of both the one-step and two-step deposition methods. In the first step, the preliminary perovskite phase was acquired via mesh
assisted grid technology, and then the deposited perovskite grid was dipped into formamidinium iodide-isopropanol to transform it into the proper perovskite segment, which is termed the "half step." The devices fabricated by using the one-and-a-half-step process display stronger performance than those fabricated through the one-step technique because of their advanced crystal quality, crystal size, absorbance, and stability (Rai et al., 2018). A novel fabrication process called the intramolecular exchange process (IEMP) was investigated by Yang et al. The larger grain size and more homogeneous film deliver a PCE of over 20% (Yang et al., 2015).

Though the two-step sequence deposition process could produce a superior performance to the one-step fabrication process, this method still needs a more complicated fabrication process and annealing environment, and the second spin-coating process or the immersion step should be precisely controlled to get a high-quality perovskite film. High-performance perovskite devices are mostly based on the two-step sequence deposition process, and this method gets more attention in lab experiments. However, it is still not suitable for large-scale fabrication processing because of the complicated fabrication process accompanied by the waste of raw materials (Xiao et al., 2014b; Fan et al., 2017).

The one-step fabrication process and the two-step sequence deposition method aim to achieve superior performance of PSCs, and they usually do not account for the cost, furthermore, the traditional fabrication method is not suitable for the large-scale preparation. Thus, the above-mentioned methods are indicated as the groundbreaking investigation to be the foundation for commercial applications in the future of PSCs (Huang et al., 2016; Rahimnejad et al., 2016).

Due to the low formation energy of the perovskite structure, low-temperature large-scale fabrication processes could be realized according to investigations from lab experiments. In the following sections, we will discuss the large-scale fabrication processes and their advantages and disadvantages, respectively.

The one-step fabrication technique renders the simplest fabrication process and facilitates the understanding of the perovskite crystallization process. However, the uncontrollable crystallization process and relatively low efficiency limited its future investigation. The two-step fabrication process could deliver high efficiency, however, the issue of incomplete conversion of the precursor still needs further study.

2.2 Large-Scale Fabrication Process
As mentioned above, we have briefly summarized the fabrication process and the crystallization dynamics process which was beneficial and enlightened for the followed investigation. To realize the commercialization of the perovskite photovoltaic devices, the large-scale fabrication process was essential and unignorable. Great efforts have been made to explore the large-scale fabrication method. Herein, several advanced large-scale fabrication processes are summarized.

2.2.1 Vapor-Assisted Solution Method
Though the two-step sequence deposition fabrication process has made great achievements, most high-performance perovskite device is based on this method. However, the intrinsic drawbacks still limit the enhancement of performance for perovskite devices. The long-term conversion process of perovskite structure and the incomplete conversion of PbI₂ would be harmful to the perovskite device. To solve the drawbacks of the two-step sequence deposition process (Zhu et al., 2018) the vapor-assisted deposition process was developed.

The vapor-assisted solution technique (Figure 4) is the further modification of the two-step deposition approach wherein MAI is deposited through a vapor deposition approach (Chen et al., 2014). This technique is based on the kinetic reactivity of MAI and the thermodynamic stability of perovskite throughout the growth process. The perovskite layer developed by this method offers excessive grain size, large crystal size, homogenous coverage, negligible surface roughness, and the full transformation of the perovskite (Hao et al., 2014). The fabricated devices generated an outstanding power conversion performance of 10.6% with excessive reproducibility showing a negligible deviation of 0.1%. Liu et al. fabricated the mixed halide perovskite film through the co-evaporation process by simultaneously evaporating the MAI and PbCl₂, followed by the annealing process. The high-quality perovskite film was obtained with a uniform and full coverage. The sequential vaporization fabrication process was also investigated (Wang et al., 2014). Abbas et al. fabricated the PSCs by evaporating the PbI₂ and MAI sequentially. The sequential evaporation process could render a more sufficient conversion of PbI₂ (Zheng et al., 2020).

Though the vapor-assisted solution method can relieve the disadvantages of the two-step sequence deposition process, this method is still not suitable for large-scale fabrication processes since this method still needs the spin-coating method to fabricate the first deposited film. The vapor-assisted solution technique can be divided into various detailed fabrication processes: 1) evaporate the material on the surface of the as-formed film; 2) deposit the film through the hot flow. The vapor-assisted solution method could also act as the post-treatment process to decrease the trap density and enhance the perovskite film quality.

2.2.2 Thermal Vapor Deposition
The thermal vapor deposition can also be divided into two versions: 1) the sequence deposition process; 2) the co-evaporation process.

The vapor deposition strategies are generally carried out by means of a high vacuum process. MAX and PbX₂ are deposited consecutively or simultaneously at the TiO₂ substrate through thermal evaporation. TVD for the perovskite absorber layers was reproduced by Liu et al. A MAPbI₃ absorber was deposited via a dual-source evaporation system in a nitrogen-filled glove box. The primary advantage of the TVD is to supply a noticeably uniform layer and the coverage of the total surface area, but controlling the vapor phase reaction is decisive. On this approach, PbX₂ and MAX were simultaneously evaporated from special sources at a precise molar ratio under an excessive vacuum. The vapor-deposited perovskite film is uniform, flat, and provides barely large crystal capabilities (Liu et al., 2013).
The co-evaporation process has drawbacks because the deposition rate is hard to control. The sequence evaporation method draws researchers’ attention, in which PbI₂ was firstly evaporated, followed by the evaporation of MAI to fabricated the high-quality perovskite film.

Since the evaporation rate of organic material is hard to control, the thermal vapor deposition is very suitable for the fabrication of all-inorganic perovskite devices. Duan et al. fabricated the high-efficiency perovskite light-emitting devices by evaporating the CsPbBr₃ (Chen et al., 2020). Wang et al. reached the record PCE of Cs₂AgBiBr₆ PSCs by the evaporation process. They claimed that the PSCs fabricated by thermal vapor deposition exhibited better performance than those fabricated by the solution spin-coating process (Igbari et al., 2019). Adachi et al. used the perovskite layer (MAPbCl₃) fabricated by the thermal vapor deposition as the hole transport layer in organic light-emitting devices (OLED), and ultra-thick OLEDs could be achieved.

In the meantime, the thermal vapor deposition method is suitable for large-scale fabrication processes, which is inspired by the OLED fabrication process. The controllable thickness and the homogenous film quality render the possibility of the commercialization of perovskite applications.

### 2.2.3 Roll-To-Roll Fabrication Techniques

Roll-to-roll (R2R) is a promising solution processing strategy used for the production of PSC. The R2R fabrication technique has some basic necessities and enforces limits on the substrates and materials used: for high-speed production the substrate should be flexible, which requires low processing temperatures (<140°C) (Etgar et al., 2012). R2R includes exceptional coating techniques which are used to shape thin film on a substrate through the usage of rotating rolls as shown in Figure 5. These techniques could be used to manufacture perovskite solar cells on a flexible substrate.

Essentially, a low crystallization energy barrier lets perovskite films be readily prepared by means of a low-temperature fabrication technique like R2R, which sets up a bridge between research and commercial applications. The slot-die coating approach is the appropriate approach for the deposition of HTL and ETL layers due to its highly uniform film deposition. R2R deposition is the continuous process of depositing layers on the substrate for making solar cells through the usage of various kinds of R2R compatible strategies. The excellent aspects of this form of deposition are that it is faster, exceptionally scalable, and cost-effective (Deng et al., 2015; Galagan et al., 2015).

### 2.2.4 Inkjet Printing

Inkjet Printing has precise control of patterns and ink consumption, and it is controlled by a computer program which provides a superior advantage in that the substrate has fewer limits. Inkjet printing can be used in flexible substrates with desired patterns, which represents the future development direction of flexible devices (Peng et al., 2017; Huang et al., 2019). Wei et al. firstly demonstrated the superiority of inkjet printing by fabricating perovskite devices using inkjet printing (Wei et al., 2014). However, the PbI₂ layer was still fabricated by spin-coating. Further investigation was conducted by Song et al., who investigated the various parameters (e.g., substrate, ink character, precursor, and so on) which would affect the final perovskite films. By cautiously controlling the fabrication parameters (e.g., solvent, substrate) and combining with the
MAI vapor post-treatment, the PCE of 17.74% could be achieved (Li et al., 2018a). This achievement inspired the following studies. The mechanism of the inkjet printing is shown in Figure 6.

The as-mentioned inkjet printing was based on the two-step fabrication process. In 2016, Quintilla and co-workers demonstrated a direct fabrication of perovskite devices by inkjet printing (Mathies et al., 2016). By combining the vacuum annealing process, they achieved a PCE of 11.3%. This work was a milestone for the commercialization of PSCs by the inkjet printing fabrication process.

The inkjet printing process has proved that scalable and reproducible devices could be achieved. However, the efficiency still falls far from the spin-coating process and the uniform film is hard to conduct. Unlike the perovskite devices fabricated in a glovebox, inkjet printing is mostly conducted in an atmospheric environment, there are so many parameters that would affect the quality of the obtained perovskite film, the detailed effects are still unclear, and the coffee ring effects still perplex the enhancement of perovskite device. However, by combining the investigations conducted on the spin-coating process and the deeper understanding crystallization process, the inkjet printing fabrication process still has the potential for large-scale fabrication. Furthermore, more efforts should be conducted in inkjet printing to solve the above-mentioned issues and to pursue the higher performance of perovskite devices fabricated by inkjet printing.

2.2.5 Doctor-Blade Coating

Doctor-blade coating is a scalable and simple fabrication process. Unlike inkjet printing, doctor-blade printing does not need expensive equipment and is easy to execute (Chen et al., 2009; Tian et al., 2012). The thickness of perovskite film can be easily controlled by the gap between blade and substrate, and the concentration of the solution can also affect the thickness of the obtained perovskite film. The perovskite precursor solution was purged on the substrate or flowed through the blade, followed by the swiping of the blade and the evaporation of the solution, and then the perovskite film can be obtained. In 2015, the doctor-blade coating was firstly demonstrated by Huang et al. However,
the quality of the perovskite film was poor (Deng et al., 2015). The subsequent investigations explored the effects of substrate temperature on the crystallization process. The hot plate accelerated the nucleation process and the growth of the crystal. With MACl as the additive, an enlarged grain size could be achieved with a PCE of 19.3% (Li et al., 2018b).

The well-studied additive strategy in the spin-coating process can also be applied in doctor-blade coating. Inspired by the previous investigations, the adhesion between substrate and precursor solution was also investigated since the speed of the blade was so fast, to obtain a uniform and fully covered perovskite film (Bi et al., 2018). By adding a small amount of surfactants, L-α-phosphatidylcholine (LP), the wettability was enhanced between the substrate and the precursor solution. Through this strategy, a compact and uniform perovskite film could be obtained. The PCE of a large-scale perovskite device (33 cm²) reached 15.3% (Deng et al., 2018).

The doctor-blade coating process has the advantages of a low cost and an easy fabrication process. By utilizing the additive strategies of the well-established spin-coating process, accompanied with the precisely controlled fabrication parameters (e.g., substrate temperature, concentration of the solution, speed of blade), doctor-blade coating has a great potential for large-scale applications. The mechanism of the doctor-blade coating is shown in Figure 7.

2.2.6 Slot-Die Coating

Slot-die coating is similar to the doctor-blade coating, however, with the ink control system, a more precise control could be achieved, which means a better perovskite film morphology. The only drawback is that more perovskite precursor solution is needed (Krebs, 2009; Sandström et al., 2012). In 2015, Vak et al. first demonstrated that by combining the 3D print technique with slot-die coating to control the position of the coating tip. By using the two-step sequence deposition process through slot-die coating to fabricate the PbI₂ film, followed by the vapor annealing, the high-quality perovskite film could be achieved, and the PCE of 11.96% could be achieved (Hwang et al., 2015). Byrkitt et al. (2018) fabricated the full slot-die coating device (except electrode), and this achievement points out the superiority of the slot-die coating. The additive strategy was also investigated, with the addition of NH₄Cl as the additive, followed with the optimization of substrate temperature and airflow system, and a perovskite device with a PCE of 15.57% could be achieved (Jeong et al., 2019).

Compared with the doctor-blade coating, the slot-die coating provides a more precise control of the deposition process with the optimization of substrate temperature, the width between the blade and the substrate, and the speed of the fabrication process. The potential of slot-die coating can be emphasized. The mechanism of slot-die coating is shown in Figure 8.

2.2.7 Spray-Coating

Spray-coating has been widely investigated in organic photovoltaic devices and the technique has drawn researchers’ attention in the field of perovskite devices (Susanna et al., 2011; Lehraki et al., 2012; Jung et al., 2018). The basic process is as follows: 1) the perovskite precursor solution was dispersed to mist; 2) the mist was spread to the substrate by gas flow; 3) after the annealing process, the perovskite film was finished (Bishop et al., 2018). The first demonstration of the fabrication of perovskite film by spray-coating was conducted by Barrows et al. They investigated the various parameters (e.g., the temperature of the substrate, the annealing temperature, and time) which would affect the quality of the perovskite film. Finally, high-quality perovskite films could be produced and a PCE of 11% was achieved (Barrows et al., 2014). Further investigation was conducted by Ishihara et al. They studied the relationship between the surface tension and the quality of perovskite film. By using NMP and DMF as the solvent, a full-coverage perovskite film could be produced and a PCE of 14.2% was achieved (Ishihara et al., 2016). Inspired by the use of anti-solvent which was usually conducted in the one-step fabrication process, Yun et al. demonstrated a novel fabrication based on spray-coating, and the spray-antisolvent (SAS) technique was conducted. By using a metal mesh as the

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FIGURE 7 | Schematic of perovskite film fabrication through doctor-blade coating.

FIGURE 8 | Schematic of perovskite film fabrication through slot-die coating.
electrode, a large-scale (16 cm²) PSCs model was demonstrated with a PCE of 12.1% (Kim et al., 2017). The mechanism of spray-coating was shown in Figure 9.

Spray-coating shows great potential in the future large-scale application of perovskite devices, however, the deeper mechanisms should be studied such as the properties of the substrate and perovskite precursor solution, the speed of spray-coating, and the annealing process. More effects should be conducted on spray-coating to realize cost-effective perovskite devices. Table 1 shows the important parameters of perovskite solar cells through the various fabrication process.

### 3 CONCLUSION

In this manuscript, we summarized the various fabrication process of perovskite devices, including the well-established one-step and two-step deposition processes which can render high-performance PSCs. One- and two-step spin-coating processes were the precursory investigation to explore their properties for future commercial applications. Though the record PCE was exhilarating, the spin-coating process was not suitable for commercialization. The vapor-based deposition process could render the high-quality film and accurate thickness. However, vapor-based fabrication needs more energy consumption. As for the roll-to-roll fabrication process, it is suitable for flexible perovskite photovoltaic devices. The precise control of thickness and the film morphology is its bottleneck. The doctor-blade coating, slot-die coating, and spray coating also have the same problems as the roll-to-roll fabrication process. Thus, we consider the inkjet printing process as the most promising technique for further application in large-scale perovskite devices. Inkjet printing could precisely control the droplet size and the trajectory, and has the virtues of a precise

### TABLE 1 | The perovskite solar cells through the various fabrication process.

| Active layer | Structural | Deposition methods | Active area [cm²] | PCE [%] | References |
|--------------|------------|--------------------|------------------|--------|------------|
| CsₐFAᵡ₋₁PbI₃ | Compact, (regular structural) | HCVD | 12 | 14.6 | Jiang et al. (2018) |
| FAPbI₃Brₓ | Compact, (regular structural) | HCVD | 12 | 14.7 | Jiang et al. (2019) |
| FAPbI₃Clₓ | Compact, (regular structural) | HCVD | 15.4 | 5.8 | Leyden et al. (2016) |
| CsₓFAPbI₃Brₓ | Compact, (regular structural) | HCVD | 41.25 | 12.24 | Lu et al. (2018) |
| Csₓ(FAPbI₃Brₓ)₀.₁ | Planar, (regular structural) | HCVD | 82.6 | 10.37 | Gao et al. (2019) |
| MAPbI₃ | Compact, (regular structural) | Roll-to-roll | 2.25 | 10.56 | Gong et al. (2020) |
| [CsₓFA₉Pb₁₋ₓBr] | - (regular structural) | Inkjet | 1 | 17 | Schackmar et al. (2021) |
| MAPbI₃ | Compact, (regular structural) | Inkjet | 2.02 | 17.74 | Li et al. (2018a) |
| MAPbI₃ | - (inverted structural) | Inkjet | 2.3 | 12.4 | Abzieher et al. (2019) |
| MAPbI₃ | Mesoporous/Compact, (regular structural) | Inkjet | 1.5 | 9.1 | Verma et al. (2020) |
| MAPbI₃ | - (regular structural) | Inkjet | 4 | 13.27 | Liang et al. (2018) |
| Csₓ₀.₅₁MA₁₋ₓPb₁₋ₓBrₓ | Mesoporous, (regular structural) | Inkjet | 4.04 | 14.5 | Li et al. (2020) |
| MAPbI₃ | Compact, (regular structural) | Blade | 26.04 | 14.6 | Yang et al. (2018a) |
| MAPbI₃ | Compact, (regular structural) | Blade | 57.8 | 14.9 | Yang et al. (2018a) |
| MAPbI₃ | Compact, (regular structural) | Blade | 100 | 4.3 | Razza et al. (2015) |
| MAPbI₃ | Compact, (regular structural) | Blade | 22.4 | 7.2 | He et al. (2019) |
| MAPbI₃ | -(inverted structural) | Blade | 1 | 19.41 | Wang et al. (2020) |
| MAPbI₃ | -(inverted structural) | Blade | 10.5 | 15.5 | Wang et al. (2020) |
| MAPbI₃ | Compact, (regular structural) | Slot-die | 17.6 | 10.6 | Cai et al. (2017) |
| MAPbI₃ | Compact, (regular structural) | Slot-die | 36 | 15.6 | Bk et al. (2019) |
| MAPbI₃ | -(inverted structural) | Slot-die | 47.3 | 4.56 | Vak et al. (2015) |
| MAPbI₃₋ₓClₓ | Compact, (regular structural) | Slot-die | 151.875 | 11.1 | Di Giacomo et al. (2018) |
| MAPbI₃ | -(regular structural) | Slot-die | 40 | 0.39 | Hwang et al. (2015) |
| MAPbI₃₋ₓClₓ | -(regular structural) | Spray | 40 | 15.5 | Hec et al. (2016) |
| MAPbI₃₋ₓClₓ | -(regular structural) | Spray | 108 | 13.14 | Lee et al. (2019) |
| MAPbI₃ | planar, (inverted structural) | Spray | 1.02 | 16.2 | Chen et al. (2015) |
| MAPbI₃ | Compact, (regular structural) | Spray | 1.3 | 16.42 | He et al. (2019) |
| MAPbI₃₋ₓBrₓ | -(regular structural) | Spray | 6.25 | 15.6 | Guo et al. (2021) |
| Csₓ₀.₈ₓFAₓ₋₁MA₁₋ₓPb₁₋ₓ(½Br₀.₈ₓ) | Compact, (regular structural) | Spray | 16.8 | 12 | Taheri et al. (2021) |
patterning ability and material-conserving characteristics. This fabrication process is suitable for various substrates. The large-scale fabrication process for commercialization is elaborated, and the advantages and disadvantages are discussed. Though great achievements have been made for large-scale fabrication processes during the past years, there are still some parameters that should be optimized:

1) The treatment of the substrate should be emphasized since the substrate temperature, the wettability of the substrate, and the cleanliness of the substrate can affect the obtained perovskite film.

2) The importance of the perovskite precursor solution should be elaborated since the crystallization process is heavily dependent on the solution composition, concentration, and types of solvent.

3) The fabrication process of large-scale devices should be optimized. The fabrication parameters should be widely investigated and carefully adjusted to obtain high-quality perovskite film.

4) To expand the large-scale fabrication process to various functional layers to shorten the preparation time of the entire device.

We predict that by combining the well-established one-step and two-step fabrication processes with the optimization of the novel large-scale fabrication process, the forecast of large scale applications is hopeful. Except for the high efficiency and the large-scale fabrication processes, the powerful encapsulation technique should be on the agenda to prolong the lifetime of the entire device.

Finally, with the efforts all made we, the dawn of the commercial perovskite devices will surely come.

**AUTHOR CONTRIBUTIONS**

ZC proposes writing ideas and completes the writing of the main part of the article and PH participates in the writing part. DW draws the pictures, CC designed the main frame of the article. MM helped the author investigate the data. YL provided writing advice, YD made critical comments and revisions to the article.

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Conflict of Interest: The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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