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Chapter

Thin Film Deposition Technologies and Application in Photovoltaics

Ning Song and Shuo Deng

Abstract

Renewable energy will play a critical role in reducing emissions to mitigate climate change. Photovoltaic (PV) is one of the most promising and prominent techniques for electricity generation based on renewable solar energy. Thin films play a critical role in PV in Si and thin film solar cells and solar modules. They can be used as an absorber layer, buffer layer, hole/electron transportation layer, passivation layer, transparent conductive oxide and antireflection coating on solar cells or solar modules. This chapter provides an overview of thin film deposition techniques and applications in photovoltaics and highlights techniques that are currently in use or are promising for mass production.

Keywords: photovoltaic, thin films, Si solar cell, thin films solar cells, solar modules

1. Introduction

In the past two decades, human-induced climate change has increased the frequency and intensity of weather and climate extremes around the globe and has cost countries hundreds of billions of dollars and thousands of lives [1]. The world is facing an energy crisis, and the prices of fossil fuels keep increasing, affecting every household. From personal, national and global perspectives, carbon neutrality is necessary. Renewable energy is a predominant term in carbon-neutral roadmaps for every country, and solar photovoltaics (PV) is currently the most affordable, accessible and prevalent technology. It involves the generation of electricity from sunlight shining through the front cover onto solar cells packaged into a solar module.

As of May 2022, global PV installations have reached 1 TW. In the PV market, crystalline-Si (c-Si) solar cells account for 95% and thin film solar cells account for 5% [2]. Thin films (<1 μ m) have an important role in Si solar cells, thin film solar cells and solar modules as absorber, passivation, buffer, electron/hole transport and antireflection coating (ARC) layers on solar cells and modules. Thin films can range from single crystal to amorphous, fully dense to less than fully dense, pure to impure and thin to thick [3].

Any new technology adapted to PV would be suitable for mass production at a reasonable cost. Meanwhile, the industry is shaping itself in the direction of higher conversion efficiency and lower cost, hence new materials and technologies are always under investigation for companies to keep their products competitive.

Therefore, it is significant to evaluate the scalability and cost-effectiveness of fabrication technologies.

The following benefits of thin films make them naturally attractive for the industry [3]:

- Conservation of scarce materials,
- Production of nanostructured coatings and nanocomposites,
- Ecological considerations—a reduction of effluent output and power consumption,
- Improved functionality of existing products,
- Solution to previously unsolved engineering problems by adding functional thin layers and
- Creation of entirely new and revolutionary products such as thin film solar cells.

In materials science, thin film technologies can be divided into gaseous state, solution state and molten or semi-molten state, where gaseous and solution states produce films with a thickness level of nanometers (nm) and Angstroms (Å), which are at the desired level of controllability for photovoltaics. Considering the accessibility and cost, the main thin film deposition techniques used in photovoltaics are physical vapor deposition (PVD), chemical vapor deposition (CVD), chemical solution deposition and sol-gel [3].

2. Crystalline silicon solar cells

As mentioned above, c-Si is dominating the PV industry with a market share of 95%. In c-Si, thin film technologies are commonly applied to fabricate anti-reflection coatings (to reduce surface reflection loss) and passivation layers (to reduce carrier or surface recombination) during c-Si solar cell manufacturing. **Figure 1** shows schematics and fabrication flow steps for mainstream c-Si solar cells: a. Al back-surface field (Al-BSF) solar cell; b. passivated emitter and rear solar cell (PERC); c. n-type solar cell with a tunnel oxide passivating contact (TOPCon); d. silicon heterojunction solar cell (SHJ) contacted on both sides with intrinsic and doped bilayers at front and rear, respectively, and indium tin oxide (ITO). It can be seen that the deposition of ARC/passivation layers is a key step among all four crystalline silicon solar cell configurations.

The technique mainly used for these ARC/passivation layers is CVD, in particular, PECVD, LPCVD and ALD. PVD-sputtering is the main technique that is used for ITO layers in SHJ solar cells. The distinguishing feature between PVD and CVD is the states of vapor. In PVD, the vapor is made up of atoms and molecules that simply condense on the substrate; in CVD, the vapor undergoes a chemical reaction on the substrate which results in a thin film [3]. Films produced by CVD generally have better quality in terms of very high purity and density and better coverage on rough surfaces than those produced by PVD methods, although the process usually involves toxic and/or corrosive gases [7].

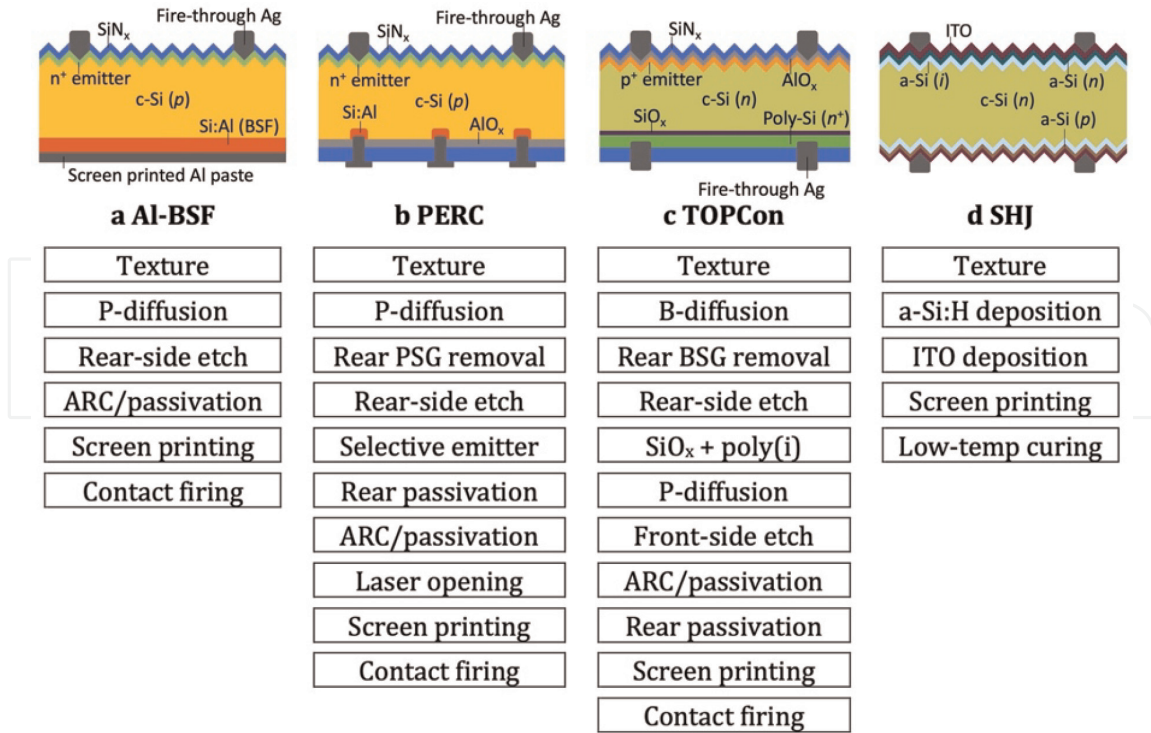


Figure 1. Schematics and fabrication flow for a. Al back-surface field (Al-BSF) solar cell; b. localized rear contacts in the passivated emitter and rear cell (PERC); c. n-type solar cell with a tunnel oxide passivating contact (TOPCon); d. silicon heterojunction solar cell (SHJ) contacted on both sides with intrinsic and doped bilayers (i/n and i/p at front and rear, respectively) and indium tin oxide (ITO) [4–6].

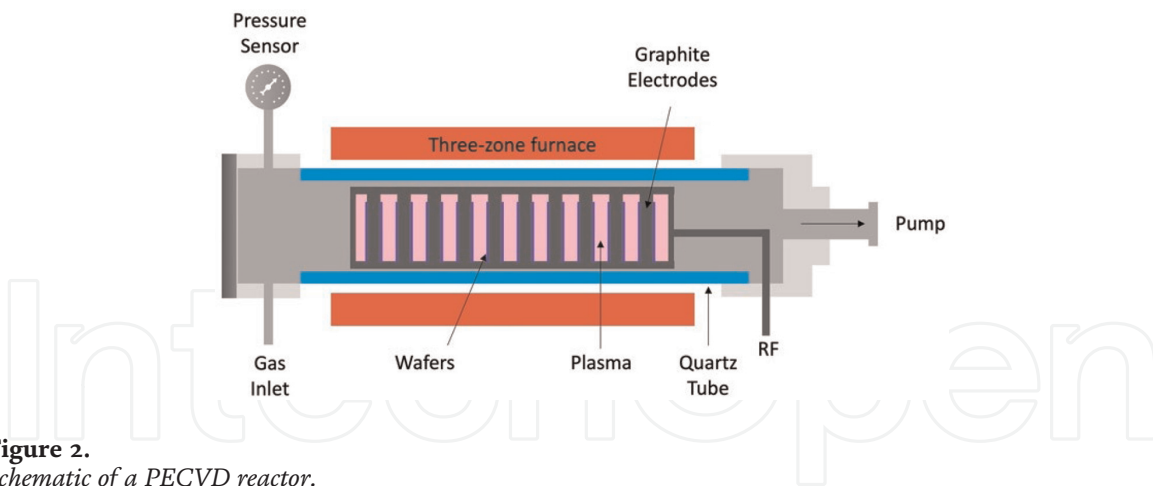


Figure 2. Schematic of a PECVD reactor.

2.1 PECVD

Plasma-enhanced chemical vapor deposition (PECVD) is one of the most commonly used methods to deposit thin films in c-Si solar cell manufacturing. The current fabrication process involves PECVD deposited silicon nitride (SiN_x) used as a front side anti-reflection coating being applied to Al-BSF, PERC and TOPCon solar cells. PECVD SiN_x:H stacked with PECVD or atomic layer deposition (ALD) deposited AlO_x provides excellent passivation and is involved as a standard step in the PERC and TOPCon fabrication processes [8, 9]. Amorphous silicon (a-Si) is another thin film material with excellent passivation property that is commonly deposited by PECVD [10]. **Figure 2** shows the schematics of a PECVD reactor. The PECVD deposition

process normally takes place at a temperature range of 150–400°C and uses RF or DC-generated plasma to initiate the reactant gases into reaction.

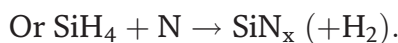
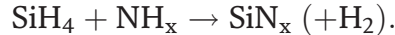
2.2 LPCVD

Compared with PECVD, low-pressure chemical vapor deposition (LPCVD) is another CVD thin film deposition technique but with a higher deposition temperature, lower deposition pressure and typically lower deposition rate. It uses heat to initiate a reaction of a precursor gas on the solid substrate. This reaction at the surface forms a solid phase material. The reactor is kept at low pressure to suppress any unwanted gas phase reactions, which also increases the uniformity. The temperatures can range from 400 to 900°C depending on the process and the reactive gases being used. **Figure 3** shows a schematic of an LPCVD reactor. LPCVD deposited films are typically more uniform, have fewer defects and exhibit better step coverage than films produced by PECVD.

LPCVD dominates in producing poly-Si for TOPcon solar cells in the PV industry. However, the technical difficulties of LPCVD are also required to be overcome to reduce the cost, in terms of higher deposition temperature, lower deposition rate and wrap-around deposition issues which increase the energy consumption for production [5, 11].

Basic CVD (PECVD and LPCVD) chemical reactions with silane gas (SiH_4) are shown below:

Silicon nitride (SiN_x):



Silicon oxide (SiO_x):



Silicon oxynitride (SiON_x):

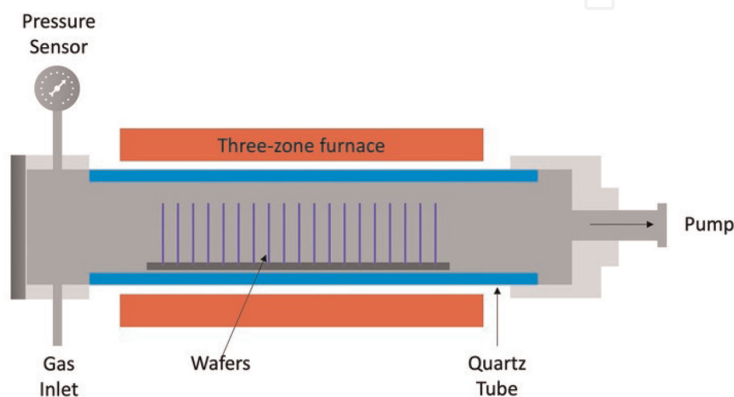
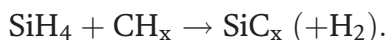


Figure 3.
Schematic of a PECVD reactor.

Hydrogenated amorphous silicon (a-Si:H):



Silicon carbide (SiC_x):



2.3 ALD

ALD is a widely used deposition technique in the field of solar cells, energy storage, catalysis and semiconductor technology. ALD deposition for AlO_x has been successfully employed by the PV industry for its excellent film quality and process economics [12–14].

In ALD, thin films are built up in cycles, in which the surface is exposed to various vapor or gas-phase species in alternating, separated doses. One cycle typically involves using the precursor dose as the first half-cycle and the co-reactant as the second half-cycle. In each cycle, a sub-monolayer of material is deposited. The precursor molecules and co-reactants react neither with themselves nor with the surface groups created. The products generated during the gaseous reactions, as well as any unreacted precursor or co-reactant molecules, are removed from the ALD reactor in the purge and/or pump steps. This is necessary to avoid reactions between the precursor and co-reactant molecules directly in the gas phase or on the surface, as this could lead to an undesired CVD component. The various steps in a typical ideal ALD cycle are shown in **Figure 4**. As shown, a typical cycle consists of four steps: (i) a precursor dosing step; (ii) a purge and/or pump step; (iii) a co-reactant step, where a small molecule is typically involved, such as water vapor; and (iv) a purge and/or pump step. This figure shows a schematic illustration of the self-limiting surface reactions during the two half-cycles, as well as the saturation of the surface coverage in every step of the cycle. The saturation of both half-cycles leads to a characteristic amount of growth per cycle (GRC) [15].

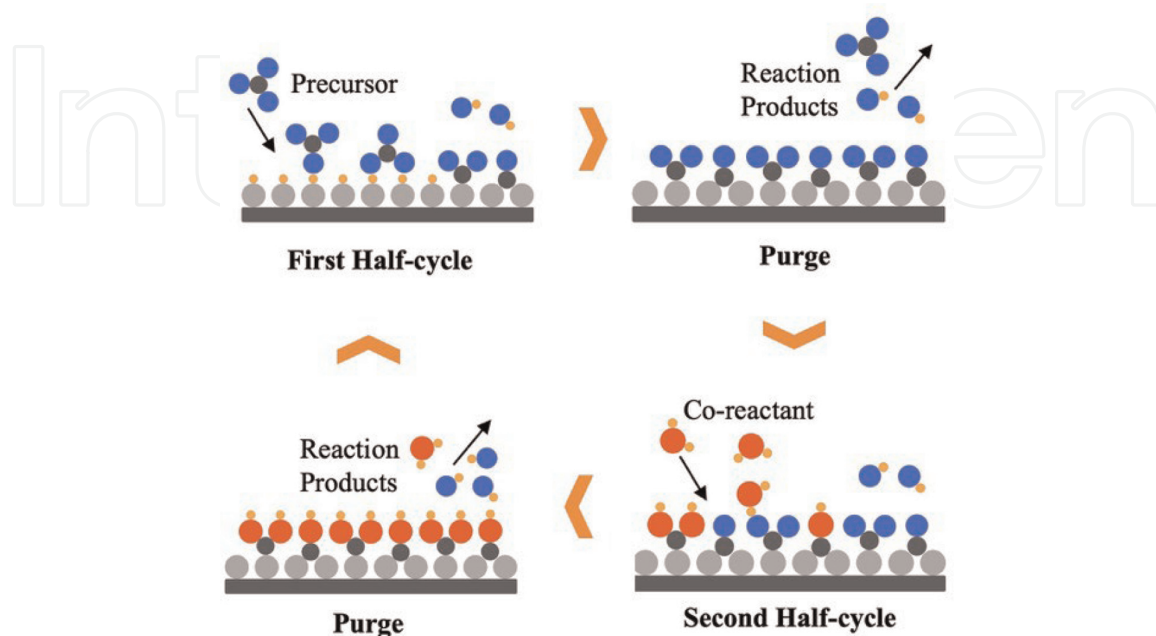


Figure 4.
Illustrations of the ALD cycle process.

3. Thin film solar cells

Even though the photovoltaic module market is dominated by crystalline silicon wafer-based technologies, thin film solar cells have the advantages of being light-weight, flexible, transparent and high temperature and radiation tolerant, making them competitive alternatives for c-Si in applications such as wearable devices, space applications, building-integrated PV and vehicle integrated PV. Thin film solar cells also avoid the massive energy consumption in the fabrication of high-purity silicon ingots in c-Si manufacturing. Moreover, thin film solar cells can be fabricated on Si solar cells to form tandem solar cells, enabling higher conversion efficiency.

Figure 5 shows three different configurations for CdTe, perovskite and CuInGaSe (CIGS) solar cells. The basic structure of CdTe and CIGS (same as $\text{Cu}_2\text{ZnSnS}_4$ (CZTS)) solar cells includes a window layer (TCO), buffer layer, absorber layer and back contacts and substrates, while perovskite solar cells have an n-i-p structure with extra electron/holes transport layers. Different layer stacks are coated on the substrate with thin-film coating methods using either solution-based processes like a chemical bath or an ink-like coating procedure, or using vacuum-based processes like thermal evaporation or sputtering [16].

3.1 Solution processing

3.1.1 Spin coating

Since it was first reported in 2009, the power conversion efficiency of perovskite solar cells has increased from 3.8–25% (lab scale) within 13 years. The emerging perovskite solar cells are under massive investigation in both research and industry due to their low-cost and high efficiency, as well as their use in Si-based tandem solar cells with further enhanced efficiency. Solution processing is the traditional fabrication method for perovskite solar cells. Spin coating is the most common fabrication approach used in the laboratory, and it has the advantages of a simple process, with no requirement for expensive and complex vacuum systems. This method could also introduce additives into the perovskite precursor solution to improve device performance [17]. **Figure 6** shows the schematics of the spin coating method for perovskite fabrication. The fabrication of perovskite absorber can be achieved by one-step

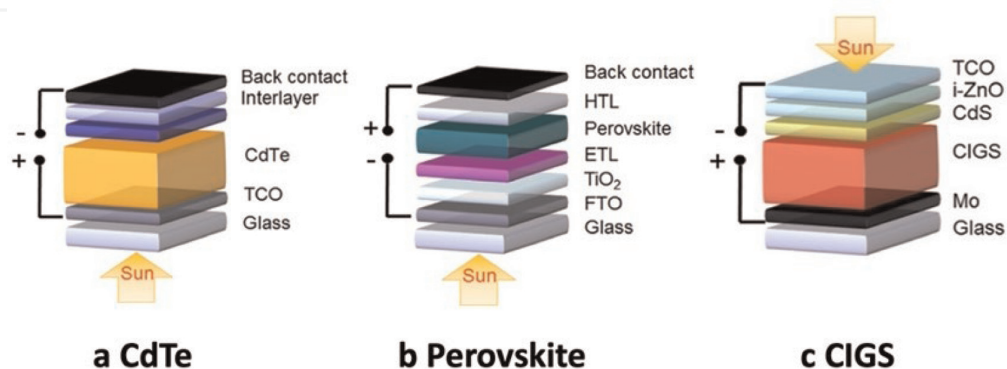


Figure 5. (a) Substrate configuration for CdTe cells; (b) perovskite cells in the n-i-p configuration; c. substrate CIGS cell configuration. Reproduced from [16] with permission.

deposition, two-step deposition or solvent engineering methods with spin-coating deposition and post annealing processes, as shown in **Figure 6**. However, the spin-coating method is generally not suitable for mass production. To translate such PV technology from the laboratory to industrial-scale manufacturing, other techniques need to be explored.

3.1.2 Slot-die coating

One of the most promising techniques for achieving large-scale (roll to roll) perovskite solar cell production is slot-die coating [18–20].

Slot-die coating can achieve highly precise control of material usage and results in very low waste levels of ink compared to other deposition methods such as spin coating or spray and screen printing. **Figure 7** shows the schematics of slot-die coating. By adjusting the ink flow pumped to the die coating head by syringe and the substrate speed, fine control of the deposited film thickness can be achieved, from tens of nanometers to tens of microns. An air knife with nitrogen gas flow would help with drying the perovskite ink, and heating could also be applied for better crystallization [18, 21].

The film formation process can also be controlled by the choice of precursor, solvent and additive. **Figure 8a** shows a schematic of slot-die coating setup with two inks and the influence of the strongly coordinating solvent dimethyl-sulfoxide (DMSO) as an additive in 2-methoxy-ethanol (2-ME) based perovskite ink on film formation. Adding a limited amount of DMSO (11.77 mol%) leads to a denser thin-

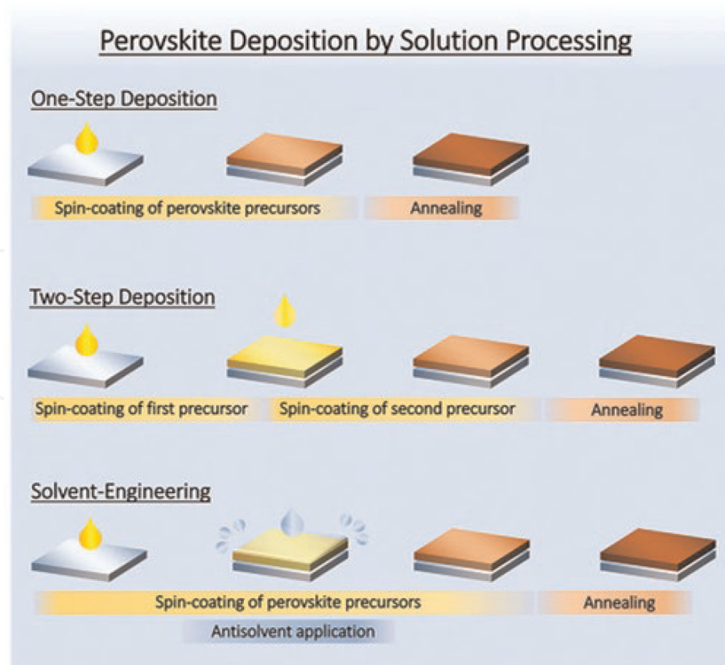


Figure 6. Schematic illustration of the deposition of a perovskite layer by spin-coating and post annealing. In the one-step method, all perovskite precursors are mixed in a single solution, which is deposited onto the substrate; In the two-step approach, the precursors is deposited first, followed by the deposition of a second precursor and thermal annealing; solvent engineering approach is to deposit all perovskite precursors in a single step, during which an antisolvent is applied triggering the crystallization of the perovskite film. Reproduced from [17] with permission.

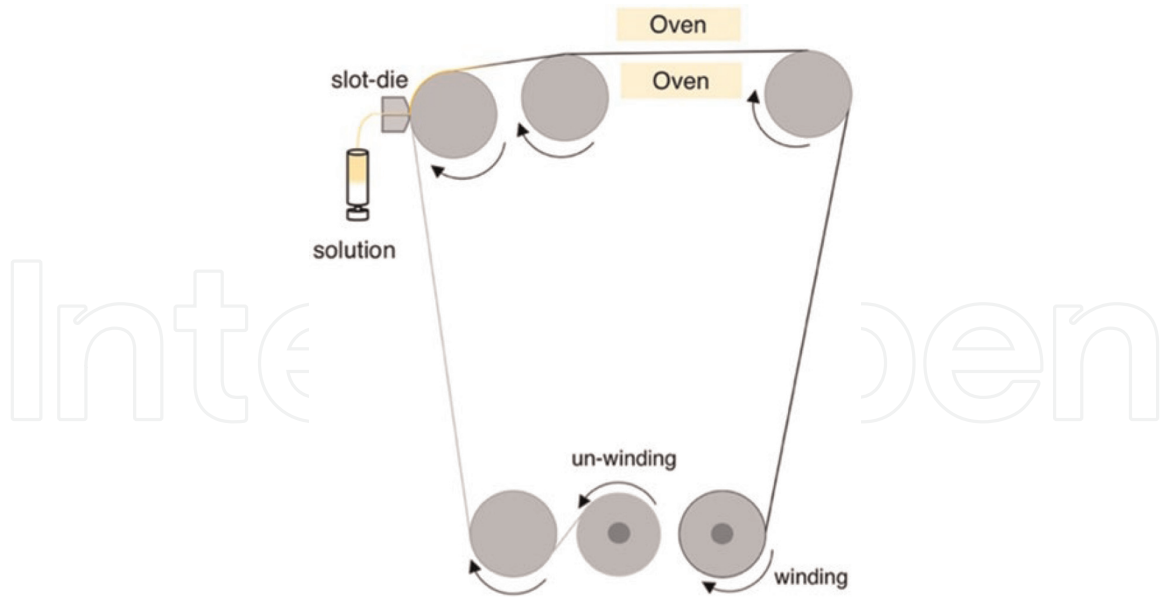


Figure 7. Schematic of roll-to-roll slot-die setup. Reproduced from [20] with permission.

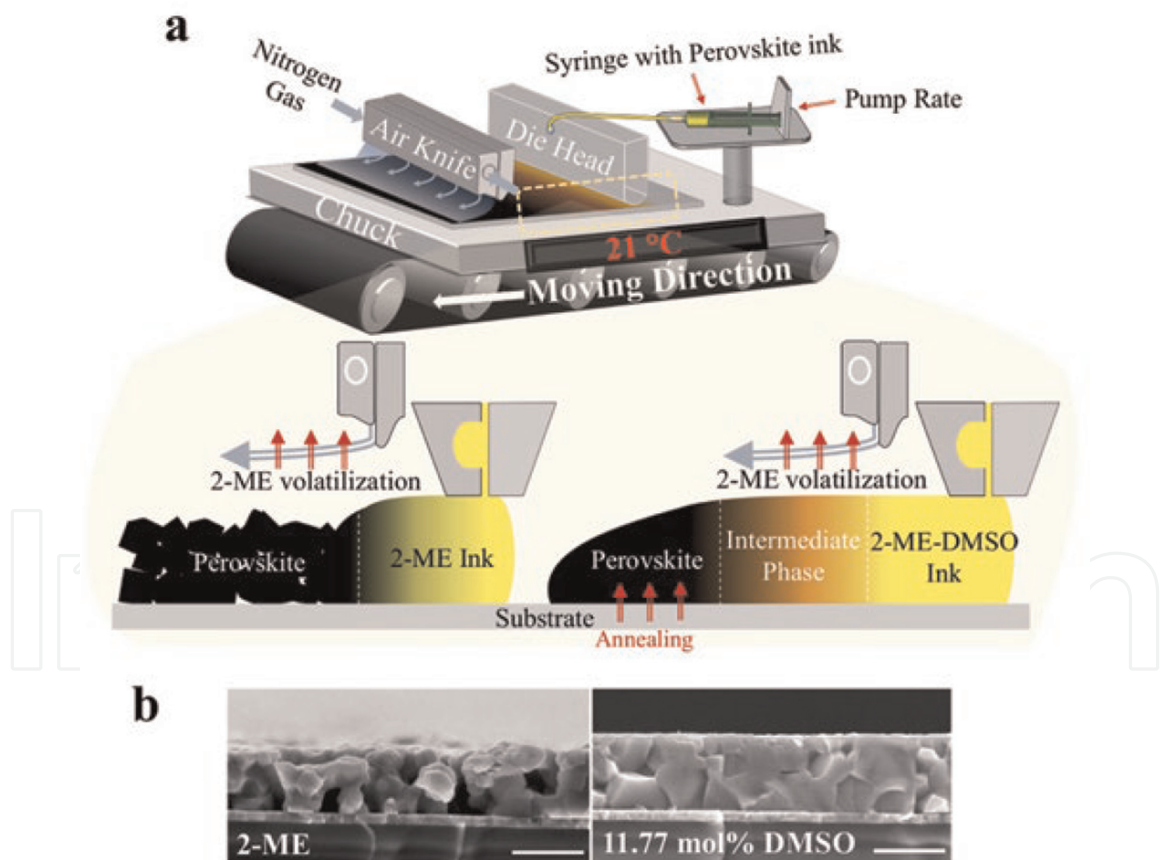


Figure 8. (a) Schematic of slot-die coater setup with 2-ME ink and 2-ME-DMOS ink. (b) SEM cross-sectional images of the coated films without and with DMOS. Reproduced from [22] with permission.

film without pinholes and large columnar crystallites as shown in the SEM cross-section images in **Figure 8b** [22]. Research on the film morphology and optimization of perovskite ink and chemicals is still ongoing.

3.1.3 Inkjet printing

Inkjet printing is another promising large-scale production method for perovskite solar cells, as it offers several favorable properties. Ink-jet printing of perovskite can achieve high-resolution film formation with customized patterns under ambient conditions.

A schematic of inject printing is shown in **Figure 9**. Inkjet printing of perovskite is carried out by a drop-on-demand (DOD) printing approach, which by definition, enables the generation of a single droplet when required, hence enabling high precision of material usage. The location of film formation can be finely controlled by motions of the inkjet printhead nozzles and the substrate. The ejection of printing materials (perovskite ink) is forced out of the nozzle by regular pressure pulses caused by contractions of ink chamber volume in the nozzles. The pressure pulse can be generated by either the mechanical deformation of a piezoelectric transducer or the collapse of thermal bubbles which involves resistive localized heating in the ink chamber. Due to the technical challenge of generating vapor bubbles in high-vapor pressure fluid, and the fact that a piezoelectric inkjet printing system can control droplet size and velocity by simply adjusting the actuation pulse, piezoelectric DOD inkjet printing would be more suitable for large-scale production of perovskite solar cells. **Figure 9** demonstrates the schematic of an inkjet printing nozzle, which an industry-scale printhead would have several hundreds of, enabling rapid and high-resolution printing at a low cost [23, 24].

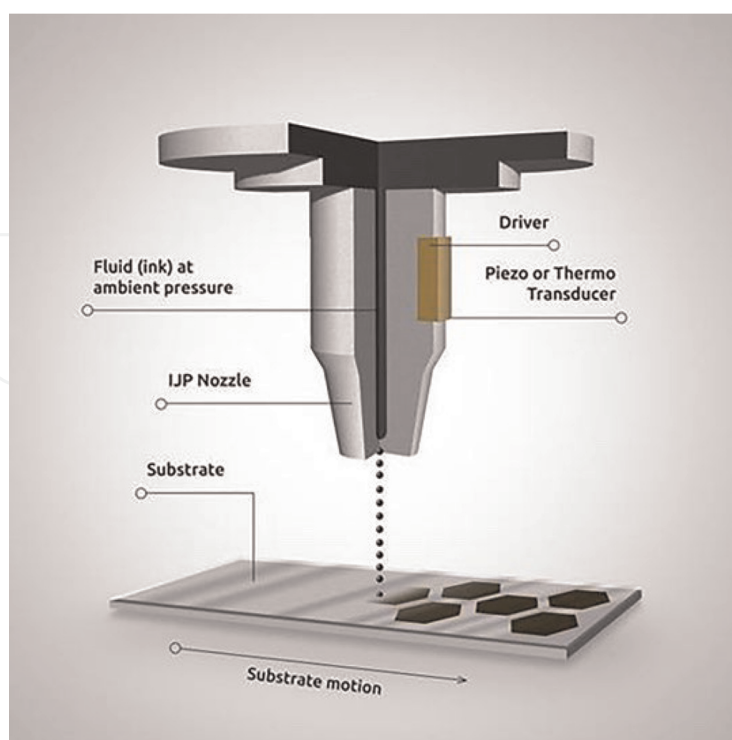


Figure 9. The schematic of an inkjet printing nozzle (Saule technologies). Reproduced from [23] with permission.

3.2 Vacuum-based processing

3.2.1 Evaporation

Among vacuum-based processing techniques, evaporation is a widely used deposition process for the formation and growth of thin films in PV. The process is beneficial in a contemporary environment and extensively applicable in the laboratory and industrial manufacturing for the deposition of thin films. A schematic of the evaporation coating is shown in **Figure 10a**.

The basic sequential steps for the evaporation process are given below:

- A vapor is created from subjecting the target material to a very high temperature by subliming or boiling (thermal) or bombarding with a beam of electrons (E-beam).
- The ejected vapor from the target material is transported to the substrate through a vacuum.
- Condensation of the vapor takes place to form a solid thin film on the surface of the substrate and further repeats of the deposition cycles result in thin film growth and nucleation.

The evaporation process has been reported to be performed using different configurations, including molecular beam epitaxy, reactive evaporation and activated reactive evaporation [3].

Snaith and his team first demonstrated that perovskite can also be deposited by thermal evaporation, reaching an efficiency of 15.4% in 2013 [25]. A recent study by

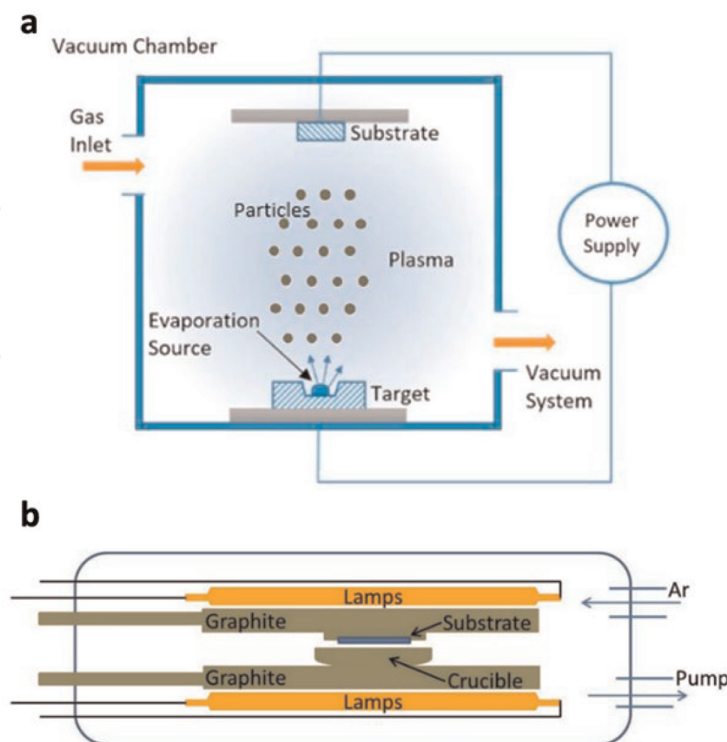


Figure 10. Schematics of (a) evaporation coating methods. Reproduced from [38] with permission. (b) Close space sublimation (CSS) system. Reproduced from [37] with permission.

Bruno and co-workers demonstrated thermally evaporated perovskite mini-modules with an active area of 21 cm² and an impressive efficiency of 18.13% [26]. These results are superior to all other reports of large-area devices in the literature, among which the highest is a blade-coated perovskite solar cell with a maximum efficiency of 16.4% [27]. Notable progress was made in 2016 by Momblona *et al.*, who demonstrated a fully evaporated planar perovskite solar cell with an efficiency above 20%. It is worth noting that the charge carrier-selective layers, the MAPbI₃ absorber and the metal electrode were all sequentially evaporated, demonstrating an all vacuum-based process for the first time [28].

Thermal evaporation offers the unique possibility of depositing multilayers of perovskite materials. Moreover, the fact that perovskite layers can be formed without the need for thermal annealing makes thermal evaporation particularly suitable for flexible optoelectronics application, in which low processing temperatures are required. Finally, this process avoids the use of the toxic solvents and allows the deposition of perovskite films without the risk of damaging the underlying layers of tandem devices [17].

Thermal evaporation is also employed in fabrication processes for commercial CIGS and CdTe solar cells [29–32]. CIGS absorbers are usually fabricated by thermal co-evaporation of the constituents, using the so-called 3-stage process. During this process, In, Ga and Se are evaporated in the 1st and 3rd stages, while Cu and Se are deposited in between, leading to the so-called double gradient of the In and Ga concentrations [33].

Close-spaced sublimation (CSS) is the fastest and simplest deposition process for both the CdTe and CdS semiconductors used in CdTe thin film solar cells, permitting high-speed in-line production [34]. The current CdTe thin film solar cell record is 21% on an approximately 1cm² cell on glass made by First Solar [35].

Figure 10 b shows a schematic of a CSS system, in which the CdTe is deposited at a pressure between 1 and 100 mbar in argon or nitrogen. The substrate and crucible are kept a few centimeters apart for vapourization of CdTe granulate and condensation (crystallization) on the substrate. Because the substrate is kept at a temperature range from 450–600°C for high-quality crystallization, a relatively high pressure is applied to suppress re-evaporation of the material [34, 36, 37].

3.2.2 Sputtering

Sputtering is another widely used PVD technique in PV that can be upscaled. In sputtering processes, a magnetron is positioned near the target. The ionic gas is introduced in an accelerated way into the vacuum chamber, blasting the target, releasing atomic-sized particles to be deposited, which will be violently projected onto the substrate. A schematic of sputtering deposition is shown in **Figure 11a**. Sputtering deposition has become a generic name for a variety of sputtering processes. These processes are named based on their source and the orientation of the process. Variants of sputtering include diode sputtering (cathode or radio frequency), reactive sputtering, bias sputtering, magnetron sputtering and ion-beam sputtering with a DC or RF power source [3]. Because of the mechanism nature of sputtering, it is ideal for the deposition of doped materials with multiple target sources. With gas inlets, the sputtering process could be used for the deposition of thin film metal or nonmetal oxides, and with hydrogen gas, hydrogenated thin films as well. Sputtering enables the production of smooth surfaces using lower temperatures, presenting excellent mechanical and tribological properties and having very good adhesion to the main materials used as substrate.

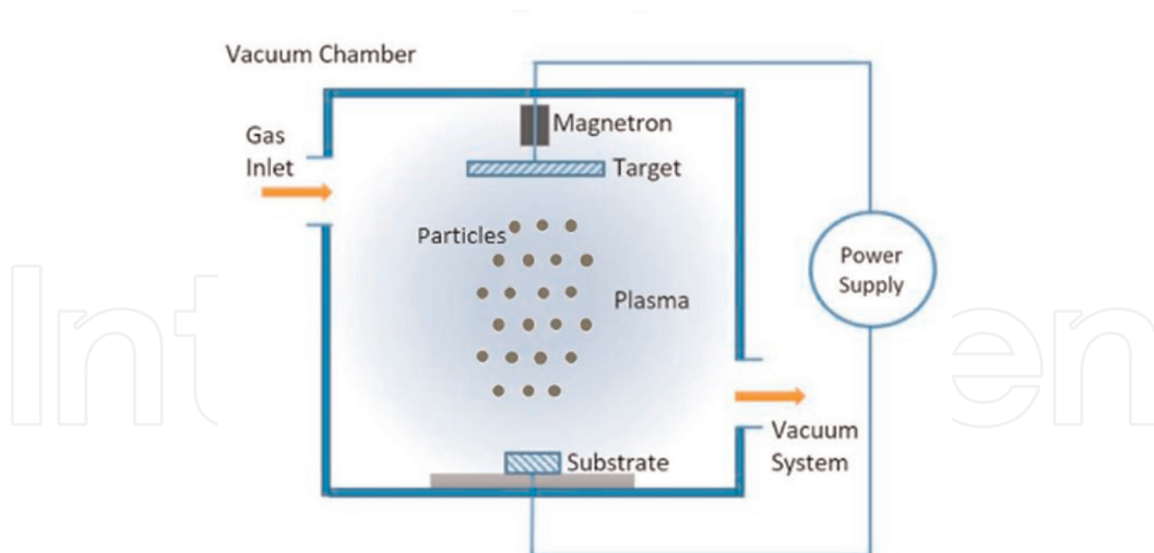


Figure 11. Schematic of sputtering deposition. Reproduced from [38] with permission.

Sputtering has been used in the deposition of CIGS and CZTS absorber layers [39, 40]. A record efficiency of 23.35% in CIGS thin film solar cells was achieved with a sputtered absorber layer [40]. Sputtering is also commonly used for TCO, buffer layers and ETL/HTL with materials such as ITO [41], FZO [42], MoO_x [43], AZO [44], TiO_x [45], and other metal oxides as interlayers, passivating thin film solar cells [46–48]. A commercialized all-sputtering system was developed by Midsummer for CIGS production with process sequence completed in different sputtering chambers for diffusion barrier, absorber, buffer, window and TCO layer deposition [49]. The sputtering method could also be employed for the deposition of poly-Si contacts and passivation for heterojunction solar cells [50–53].

3.2.3 MOCVD (MOVPE)

III–V thin film solar cells are widely used in aerospace applications, due to the high energy conversion rate, wide operating temperature range and high radiation resistance [54]. The record efficiencies for III–V multijunctions are 38% for a five-junction cell (bonded) and 37.9% for InGaP/GaAs/InGaAs solar cells. The record efficiency for a GaAs single-junction cell is 25.1%, held by Alta Devices [35]. One key deposition method used in III–V thin film solar cell fabrication is metalorganic chemical vapor deposition (MOCVD), also called metalorganic vapor-phase epitaxy (MOVPE). Metal–organic CVD (MOCVD) is a CVD process for growing epitaxial films and is done by flowing precursor gases over the substrate. In III–V semiconductors, the metallic element is carried by an organic gas such as $\text{Ga}(\text{CH}_3)_3$ and $\text{In}(\text{CH}_3)_3$ along with AsH_3 or phosphine (PH_3). The gases are allowed to decompose due to pyrolysis on the heated substrate surfaces to produce the desired film. A schematic is shown in **Figure 12**. Commercially available MOCVD tools are designed to produce traditional III–V semiconductor devices for electronic and optoelectronic applications. Relative to solar cells, these devices have complex layered structures that require extremely precise control of thickness, composition and doping profiles, and each fabrication typically takes multiple hours to complete. The simpler structure and relatively wide process windows of solar cells present an opportunity to use correspondingly simpler

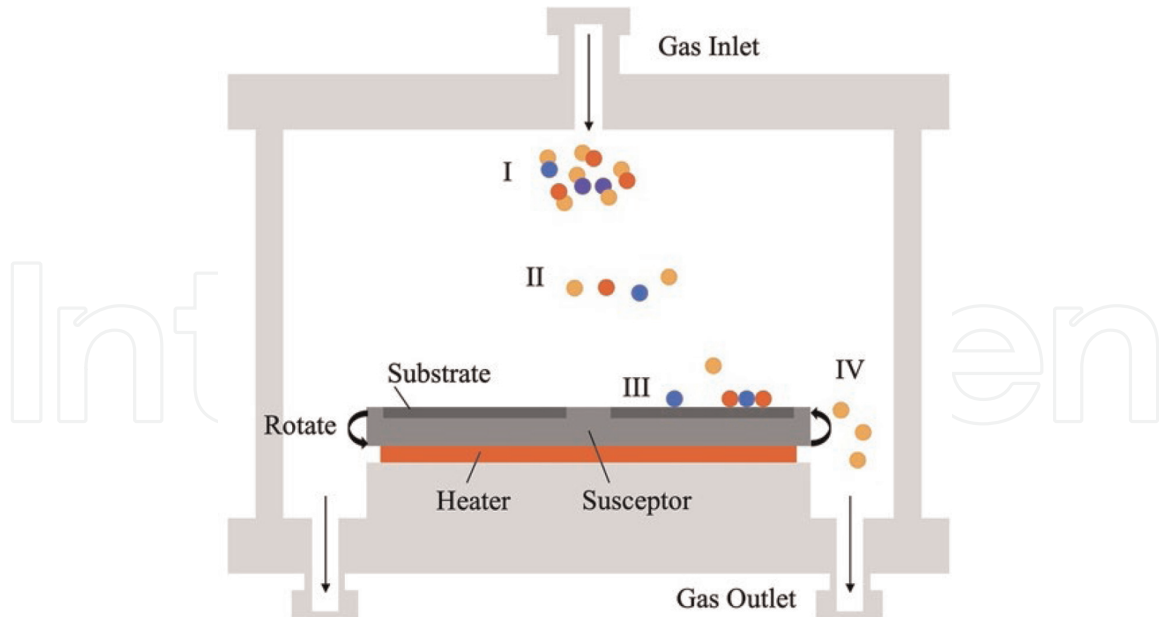


Figure 12. MOCVD reactor basic scheme and fundamental working principle: I—Precursor, II—Cracking, III—Deposition and IV—Removal of residual gases.

and less expensive equipment. With customized MOCVD, combining automated substrate loading and unloading, fast temperature ramping, high growth rate and the elimination of pressure cycling, the fabrication process time could be reduced to 15 min, which makes a significant contribution to fabrication cost reduction [55].

4. Solar modules

Another important application of thin films in PV is the antireflection coating (ARC) on the surface of solar glass where the light first reaches the solar panels. Currently, single-layer antireflection coated solar glass has a dominant market share of 95% compared to glass with other coatings or no coating, for Si PV modules [2]. This ARC results in an efficiency gain of 2–3%; that is, 2–3% more light can enter the solar modules to be

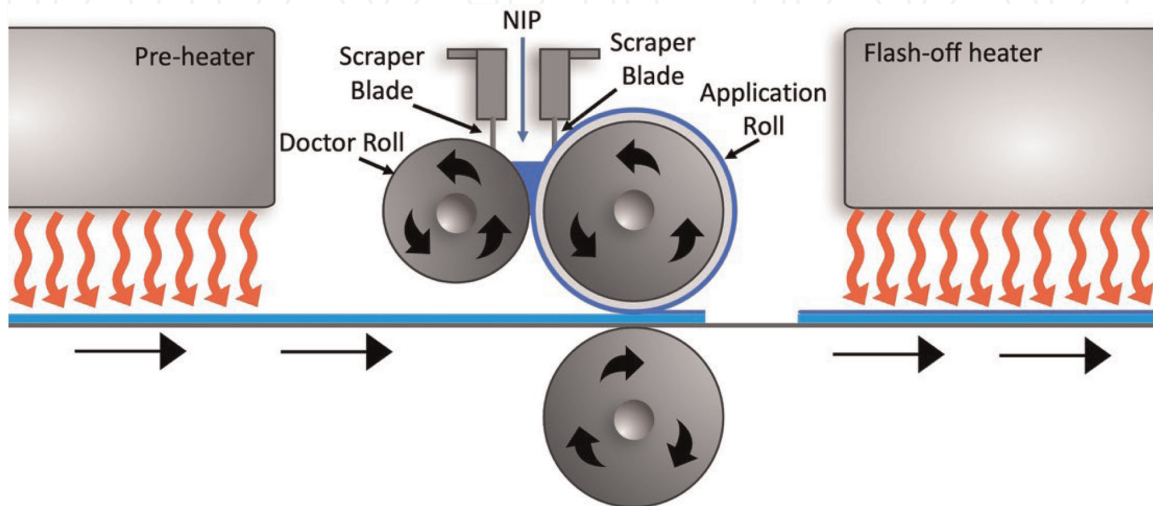


Figure 13. Schematic of roller coating process.

absorbed by the solar cells and converted into electricity. The most common PV ARC consists of a ~ 100 nm single layer of nano-porous silica deposited onto the solar glass cover via sol-gel roller coating, followed by a high-temperature sintering and tempering process. The roller coating is one type of sol-gel method that is scalable and low cost. A schematic of a roller coating process is shown in **Figure 13**. Other chemical methods such as slot-die coating, dip coating, spin coating, spray coating, as well as PVD methods like sputtering multilayer ARC are also under investigation.

5. Conclusion

This chapter reviewed thin film deposition techniques in the PV industry and research and highlighted those which are suitable for mass production including vacuum-based techniques such as CVD, evaporation, sputtering and non-vacuum based like slot-die coating, roller coating and inkjet printing. Thin films are important components for solar cell and solar module devices which enable high solar energy to electricity conversion efficiency. The increasingly enormous deployment of PV and rapid development of the PV industry has placed higher requirements for fabrication technologies and equipment in terms of good film properties and low cost for mass production.

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Conflict of interest


The authors declare no conflict of interest.

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