Progress of Perovskite Solar Modules

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Perovskite solar cells (PSCs) reached 25.5% of certified power conversion efficiency (PCE) in 2020. A remarkable PCE of PSCs has urged scalable technologies to grow for manufacturing modules. Therefore, scalable technology is rapidly developing though the performance of perovskite solar modules (PSMs) is still far behind that of PSCs. Herein, the recent progress in scalable technologies is reviewed with addressing important parameters in each process. In addition, the performance measurement protocols for PSMs are specifically discussed based on International Electrotechnical Commission to be prepared from the industrial point of view. Finally, the environmental hazard impact by accidental lead leakage and the introduction of green solvent to replace of current toxic solvent are described.

1. Introduction

Perovskite solar cells (PSCs) have emerged as a strong candidate for commercial photovoltaics (PVs) since the first introduction as a solid-type in 2012.[1,2] The certified power conversion efficiency (PCE) of PSC now reached 25.5% in 2020,[3] owing to the sophisticated material engineering based on fundamental understanding of optoelectronic properties of halide perovskite.[4–7] The noticeable growth of PSCs, coupled with their low process cost, urged to study the feasibility of PSCs for a scalable module from the industrial point of view. Accordingly, various scalable technologies have been introduced and adapted to produce a uniform and homogeneous perovskite film,[12–32,33–35] which enables a unit cell to be scaled up toward a scalable subcell to compose a module. Unlike PSCs, the perovskite solar modules (PSMs) are still behind the mature technology, mainly due to a difficulty of using the established technology customized for a unit cell. The conventional PSCs have been minutely engineered based on a spin-coating method, which hinders the perovskite crystals from being grown over large area with ensured film quality.[7] For example, the most widely used method to control the crystallinity of perovskite film is anti-solvent dripping on the spinning film based on the spin-coating method for a substrate area less than 10 × 10 cm in laboratory scale,[8] which is difficult to be imbedded for a scalable substrate, resulting in an inhomogeneous film quality over the large area though the anti-solvent technology still can be applied to PSM using a bathing method with green anti-solvent, such as ethyl acetate and n-butyl alcohol.[9,10] Therefore, the PSM technology is independently under developing, which, however, can be sufficiently accelerated by the aid of the accumulated experience on PSCs. Furthermore, as an interest in PSMs that have been continuously grown from the perspective of industry, it is significantly required to follow international standards for evaluating PV performance and satisfy the criteria, particularly in stability test. For measuring the long-term performance of conventional PSCs, the international standard gets extremely simplified to several conditions, which are then applied for PSCs to monitor their stability.[11–13] The transition from a PSC to a PSM is highly demanding the criteria of performance test that strictly satisfies the international standards to not only provide objective performance results but also compete with other PVs.

In this review, the recent progress in scalable technologies is investigated, ranging from blade coating to slot-die coating, inkjet printing, and vacuum deposition. The technology-dependent efficiencies of scalable PSCs and PSMs are addressed with corresponding device size (or active area). Furthermore, protocols for measuring efficiency and long-term performance are specifically suggested fully based on the relevant standards issued by International Electrotechnical Commission (IEC), followed by discussions about the status of PSMs in terms of PV performance compared with other PV technologies. Finally, lead issue, being considered as a huge obstacle to commercialization, is explored in parallel with strategies to resolve it.

2. High Volume Manufacturing Technology

2.1. Blade Coating

A blade coating method is widely adopted for scalable PSMs due to its facile processability,[14–27] which is indeed regarded as an efficient method in terms of saving the precursor solution.[14,15] A relatively small amount (~20%) of perovskite
A precursor solution is required for the blade coating compared with the conventional spin-coating method. Figure 1a,b shows the schematic illustrations of the blade coating where a D-bar coating is followed (Figure 1a) by a drying process by the aid of an air knife (Figure 1b). It is noted that the rotating D-bar is not necessarily used in the blade coating, as shown in Figure 1a,b. Various types of rod can be applied in the blade coating. A perovskite precursor solution (≈70 μL cm⁻²) was coated on the substrate by rolling with the speed of 5 mm s⁻¹. During the coating, advancing and receding meniscus generated in front of blade (first contact point between the solution and the substrate) and behind blade (the place where the blade has passed) along with coating direction, respectively. The receding meniscus is considered as an important factor determining coating quality, where the surface tension gradient is particularly observed from a mixed solvent system, in the case of perovskite precursor solution, and acts as driving force to form a compact film. Furthermore, the coating speed is found to closely relate with morphology. When the coating speed is low, the evaporation within the receding meniscus region governs film formation, preferring aligned morphology while the high speed is likely to result in isotropic morphology. A removal of the solvent was sequentially enabled by blowing air (N₂ or Ar) with a pressure ranging from 0.6 to 2 MPa at a fixed distance of 70-110 μm (4 mm for the extreme case) between the wet film and the air knife. The air blading, indicating the air knife-assisted drying process, not only helps a uniform dispersion of the precursor-coated wet film but also effectively controls the film thickness. As shown in Figure 1c, the resultant thickness of the perovskite film was in inverse proportion to the air knife pressure. After the drying process by air blading, a post-heat treatment was applied for the film crystallization at around 130 °C. Notably, the evaporation of the solvent plays a key role in determining the morphological quality of the perovskite film. In case of the spin-coating method, being used for the laboratory-scale unit cell, the anti-solvent is generally adopted at the end of spinning for the fast crystallization to induce supersaturation. Meanwhile, the blade coating encourages the precursors to form an intermediate phase, coupled with a residual solvent. Figure 1d represents the scanning electron microscopy (SEM) image of the perovskite film by a blade coating method where the intermediate phase, MA[(PbI₂)₁/₃PbI₃] 2DMSO (methylammonium = MA; dimethyl sulfoxide = DMSO), was well optimized and accordingly led to a larger grain size (~700 nm) while the conventional spin-coating method with anti-solvent formed a smaller size of grains (~300 nm), as shown in Figure 1e.

On the one hand, the composition of precursor can afford to govern the formation of adduct as the intermediate phase using additives, such as lead acetate (PbAc₂), guanidinium iodide (GAI), DMSO, hexamethylphosphoramide (HMPA), tetrabutylammonium bromide (TBAB), thiourea, and Zn(C₆F₅)₂. Therefore, the growth of scalable film with enhanced uniformity and crystallinity was derived by the additives-assisted intermediate phase leading to an increased carrier lifetime. The formation of MA acetate from the precursor solution containing PbAc₂ and MA iodide (MAI) along with the addition of GAI resulted in a beneficial effect on a crystal growth during the drying process. Similarly, the incorporation of TBAB enhanced the crystallinity of perovskite film and also effectively inhibited the organic cations in the lattice from escaping, resulting in a remarkable long-term stability. Thiourea was effective on controlling the crystal morphology with enhanced uniformity on the flexible substrate.

Figure 1. Schematic illustrations of the blading coating composed of a) a D-bar coating process and b) a drying process using an air knife. c) Dependence of perovskite film thickness on the air pressure. High-resolution SEM (HR-SEM) images of perovskite film by d) the blade coating method and e) the conventional spin-coating method with anti-solvent. a,b) Reproduced with permission. Copyright 2019, American Chemical Society. c–e) Reproduced with permission. Copyright 2018, Elsevier.
Furthermore, Zn(C$_6$F$_3$)$_2$ was added in the precursor to reduce the moisture interference during the perovskite coating process at ambient air. On the other hand, the solvent for the precursor solution has a significant impact on the film morphology by affecting the kinetics based on different vapor pressure during the air-blading process. While N,N-dimethylformide (DMF) is widely considered as a solvent for the perovskite precursor solution, its high boiling point (152-154 °C) with a low vapor pressure (0.46 kPa at 25 °C) brought a difficulty in evaporating, which is directly responsible for the resultant inhomogeneous morphology. To this end, 2-methoxyethanol (2ME) with a relatively lower boiling point (124-125 °C) and a higher vapor pressure (0.83 kPa at 25 °C) was applied as a volatile solvent, aiming at facile and controllable solvent evaporation. The replacement of DMF with 2ME improved the perovskite film with high uniformity, resulting in a PCE increase by 13.85% of PSM (area = 16 cm$^2$) with high reproducibility.

### 2.2. Slot-Die Coating

A slot-die coating method can take the overall advantage of the blade coating method due to the compatibility with a blade. Figure 2a shows a schematic process of the slot-die coating, where a perovskite precursor solution is supplied from a container to a slot-die head with a given flow rate by a pump, where a formation of a steady-state meniscus between the slot-die head and the substrate is a key factor to obtain a homogeneous and defectless slot-die-coated perovskite film. Similarly, the gas quenching can be used to control the solvent evaporation for fast crystallization by inducing the instant supersaturation. The deposited perovskite precursors as passing through the slot-die head are sequentially exposed to a gas jet (50 L min$^{-1}$ of N$_2$) through the air knife to induce high uniformity with less pinholes.

Various affecting parameters are carefully optimized to maintain the steady-state meniscus. Figure 3a–c shows the effect of correlation between the flow rate and the coating speed on the morphology of perovskite films. The flow rate is generally given as 10–1000 µL min$^{-1}$, whereas the coating speed is set around 1 m min$^{-1}$. When the flow rate is too low for a given coating speed, the deficient precursor supply breaks the meniscus and results in the inhomogeneous coverage, as shown in Figure 3a with a thinner film thickness (400 nm at F) at the end point compared with the initiation point (600 nm at S). The excessive precursor supply by a high flow rate also leads to a poor film morphology with a variation of film thickness (680 nm at S and over 1 µm at F, Figure 3b). The thick precursor coated on the substrate by the high flow rate has trouble in controlling a uniform removal of the solvent, leaving pronounced defects even with cracks on surface. On the other hand, the balanced flow rate and coating speed enable the steady-state meniscus to maintain during the whole coating process, which is responsible for a comparably uniform thickness across the film (580 nm at S and 560 nm at F, Figure 3c) with a homogeneous grain morphology (Figure 3d,e). When the balanced flow rate and coating speed are properly set, the perovskite film thickness is governed by a height of the slot-die head and a concentration of the perovskite precursor solution. The gap height (distance) between a substrate and the slot-die head is kept as 30-200 µm similarly with the blade coating method. As the gap height was increased from 40 µm to 50 µm and 60 µm, the perovskite film became thicker from 457 nm to 563 nm and 647 nm, respectively. The PCE of PSCs using the different gap heights decreased from 15.7% for 40 µm to 12.5% for 50 µm to 9.4% for 60 µm (Figure 3d) mainly due to a gradual reduction in the open-circuit voltage ($V_{OC}$) and fill factor (FF). In the meantime, the variation in the concentration of the perovskite precursor can affect the film thickness, showing an increased film thickness from 336 nm for 20% w/w to over 1 µm for 40% w/w. The overall high PCEs were observed between 431 nm- and 549 nm-thick film, which was obtained from the concentration of the perovskite precursor solution between 23% w/w and 30% w/w (Figure 3e). Furthermore, the compositions of the perovskite precursor solution and the solvent are highly important to determine the film morphology in the same manner with the blade coating method. The use of a mixed precursor for the lead source of PbAc$_2$ and PbCl$_2$ is beneficial to the crystal growth in the slot-die coating and, thus, results in the uniform morphology with larger grains. A perovskite precursor based on 2ME suffers from uncontrollable crystallization, which can be effectively suppressed by adding 12 mol% DMSO to form a dominant crystal intermediate phase with DMSO over 2ME, leading to a high efficiency with great reproducibility.

![Figure 2](image_url). Schematic image of a slot-die coating method. Reproduced with permission. Copyright 2018, Royal Society of Chemistry.
The slot-die coating method aims for the high-throughput commercial production with large scale. A high compatibility of the slot-die coating with industry standard including roll-to-roll process, furthermore, encourages a study on the ink formulation of each layer for commercialization.\cite{verma2020fully} Figure 4a shows an architecture of PSM with a perovskite-infiltrated c-TiO$_2$ (compact-TiO$_2$) and m-TiO$_2$ (mesoporous-TiO$_2$) as an electron transport layer, m-ZrO$_2$ as a spacer, and carbon electrode. PSMs exhibit discrete stripes for a serial connection, which can be facilitated either by a sequential line scribing (noted as P1, P2, and P3 in Figure 4a) or by delicate layer depositions in the presence of a spatial offset with high accuracy. It is noted that the whole metal oxide layers, c-TiO$_2$, m-TiO$_2$, and m-ZrO$_2$, were sequentially slot-die coated and sintered together once at 450°C for 1 h, which was followed by another slot-die coating of carbon electrode with a sintering at 400°C for 30 min (Figure 4b). The perovskite precursor solution was then dropped on the carbon electrode and simply infiltrated through the sintered stack.\cite{verma2020fully}

2.3. Inkjet printing

An inkjet printing method has been tried to PSMs owing to the merits of cost-efficiency, low material consumption, and noncontact deposition for complicated patterns.\cite{wang2018inkjet, chen2018inkjet, zhang2018inkjet} Nevertheless, the performance of the PSC prepared by the inkjet printing method is usually lower than that by other scalable technologies due to stringent requirements for the process. The engineering of the solvent composition is necessary to make a controllable and printable ink. Furthermore, controlling the interaction between the ink and the substrate is particularly challenging to form a uniform wet film from individual ink droplets. The drying process after the printing is indeed important, because a removal of the solvent is directly involved in nucleation and crystallization. Figure 5 shows the key steps of the process chronologically from 1) ink preparation to 2) inkjet printing, and 3) a drying and annealing process.\cite{zhang2018inkjet}

The recent ink composition generally resembles the conventional precursor solution where multiple cations and double halides are mixed with DMSO to form an intermediate phase to improve the film morphology.\cite{zhang2018inkjet} However, the solvent is more carefully engineered not only for making a controllable droplet by the piezoelectric response but also for inducing a homogeneous solvent evaporation during the drying process. A combination of γ-butyrolactone (GBL), DMF, DMSO, and n-methyl pyrrolidone (NMP) is widely used to satisfy the ink requirements, whereas the environment-friendly solvent, ethyl acetate, is occasionally tried.\cite{zhang2018inkjet, zhang2020inkjet, zhang2021inkjet} Figure 6a shows the effect of solvent composition on the crystallization. The addition of DMSO to DMF effectively retarded the crystallization due to a higher boiling point of DMSO (189°C) compared with DMF (152-154°C).\cite{zhang2020inkjet, zhang2021inkjet, zhang2022inkjet}
which was further pronounced using NMP (boiling point \( \approx 204 ^\circ C \) and vapor pressure \( \approx 0.32 \) kPa).\(^{[39]}\) The use of a mixture of DMF and NMP (1:1 v/v) resulted in larger grain size around 500 nm with a low roughness of 6.5 nm.\(^{[39]}\) During the inkjet printing, the contact angle of ejected droplet on the substrate is carefully tuned by manipulating the surface tension of the ink and the surface free energy of the substrate. Because the contact angle between 5° and 10° is preferred to form a continuous wet film without local de-wetting or ink spread by overflow, a surface treatment, such as \( O_2 \) plasma, is preceded by inkjet printing.\(^{[38]}\) When the contact angle is maintained low, the distance between neighboring droplets (drop spacing), designated as printing resolution in dpi, governs the film thickness, as shown in Figure 6b.\(^{[38]}\) The perovskite film thickness was gradually increased from 400 nm for 600 dpi to 4 \( \mu \)m for 2000 dpi with a vertical growth of single grain extending over the entire film, being beneficial to the charge extraction. Notably, the crystallization with a preferred orientation was more evident, as the film thickness increased (Figure 6c),\(^{[38]}\) which differs from the crystal orientation of the conventional spin-coating process. Furthermore, it is remarkable that the inkjet printing successfully made a well crystalline perovskite film as thick as 4 \( \mu \)m with a comparably high roughness owing to a significantly slow vacuum-assisted drying process.

### 2.4. Thermal Vacuum Deposition

Thermal vacuum deposition is a fully mature technology for semiconductor industry, which is being widely used for a
commercial mass production of electronics including organic light-emitting diodes. Therefore, the vacuum deposition is generally regarded as the most feasible process for the scalable semiconductor productions. Furthermore, a solvent-free process of the vacuum deposition can completely avoid the environmental issue of toxic solvents and also allow the multistacking of materials without any concern for damaging underlying layers. Nevertheless, the progress of a vacuum deposition technique for PSCs was comparably retarded due to difficulties in controlling the sublimation of organic cation precursors as well as a high initial facility cost for the co-evaporation of multisources. Recently, the vacuum deposition technique has been rapidly grown and shown promising results for scalable PSMs. Figure 7a shows a schematic illustration for the co-deposition of perovskite precursors. The perovskite film is generally grown in a high-vacuum chamber (~10^-6 mbar), where crucibles containing each precursor are located. The stoichiometry of precursors and the overall film thickness are carefully controlled using quartz crystal microbalances (QCMs) located above the individual precursor in the crucible. The estimated values from QCMs (tooling factor, Z-factor, substrate rotation speed, etc.) are minutely calibrated by comparing with a real deposition thickness, which is mechanically measured by a profilometer. An extra QCM can be additionally located close...
to the substrate holder to monitor the overall deposition details for the perovskite layer.\textsuperscript{[48,49,52,53]} The deposition rate is varied by manipulating a thermal power for each precursor, which requires \(-250 \degree C\) for PbI\(_2\), \(70 \degree C\) for MAI, \(100 \degree C\) for formamidinium iodide (FAI), and \(410-430 \degree C\) for CsBr to initiate the sublimation.\textsuperscript{[46-48,51]} The relatively high sublimation temperature of PbI\(_2\) allows a fine control over the vapor deposition, whereas the low sublimation temperature of organic cation precursors, particularly for MAI, brings a complicated situation where the careful control of the organic precursor vapor is struggled by monitoring both QCM and a change in the partial pressure, induced by precursor vapor.\textsuperscript{[52,53]} In the beginning, dual sources of MAI and PbI\(_2\) were used,\textsuperscript{[51]} which paved a way for the current co-evaporation system. The co-evaporated MAPbI\(_3\)-based devices showed the high PCEs of 20.28\% for PSC (0.16 cm\(^2\) for active area) and 18.13\% for mini-module (21 cm\(^2\) for active area).\textsuperscript{[54]} More recently, triple (even quadruple) sources of two organic/cesium cation precursors (FAI, MAI, or CsBr) and PbI\(_2\) were explored to adopt the outstanding performance and phase stability of mixed-cation and -halide perovskite crystals where the FA\(^+\) and I\(^-\) composed the majorities,\textsuperscript{[47,48,55]} granting a benefit of using more controllable FA\(^+\) source than MAI.\textsuperscript{[48,56]} A high quality of FA\(_{0.9}\)Cs\(_{0.1}\)PbI\(_2\)Br\(_{0.3}\) film was grown by a co-deposition of three sources, FAI, PbI\(_2\), and CsBr.\textsuperscript{[47,55]} It is noted that the perovskite crystal grains increased from \(-50 \text{nm}\) as deposited at room temperature to larger than 1 \(\mu m\) after the post-annealing process (135–150 \(\degree C\)).\textsuperscript{[48,55]} though the thermal vacuum process frequently ensures the formation of perovskite crystals in the absence of additional annealing process,\textsuperscript{[57]} being applicable to plastic substrates.\textsuperscript{[58]} On the other hand, the annealing process for the FAI-rich perovskite film occasionally showed a detrimental effect by accompanying a reduction in absorbance and crystallinity.\textsuperscript{[48]} The crystallinity and morphology of the perovskite film are highly dependent on the substrates owing to the different surface chemistry between the substrate and adhesive precursors.\textsuperscript{[46,49,52,55]} A formation of perovskite film on poly(triarylamine) (PTAA)-coated ITO resulted in a larger grain size (Figure 7b) compared with the deposition on a glass substrate (Figure 7c). Among MA-free thermal evaporated perovskite films, a 500 nm-thick FA\(_{0.9}\)Cs\(_{0.1}\)PbI\(_2\)Br\(_{0.3}\) with 5\% excess PbI\(_2\) component at grain boundaries demonstrated the highest PCE of 18.2\% of stabilized PSC, as shown in Figure 7d, and maintained \(-90\%\) of initial PCE after the continuous operation over 35 h at maximum power point (mpp).\textsuperscript{[55]} Co-deposition of three sources of FAI, MAI, and PbI\(_2\) exhibited a promising PCE of 18.8\% with a superb stability retaining 90\% of initial PCE after a month of continuous operation at mpp.\textsuperscript{[49]} The phase stabilization of \(\alpha\)-FAPbI\(_3\) was facilitated using MA\(^+\) without any post-annealing step, showing a peculiar self-limiting process in FA\(_{1-x}\)MA\(_x\)PbI\(_3\), where the incorporation content of MA\(^+\) in the perovskite lattice was strictly limited regardless of MAI deposition rate.\textsuperscript{[49]} Moreover, the solvent-free thermal evaporation method enabled the adoption of scalable 3D/2D hybrid dimensional perovskite,\textsuperscript{[59]} which pursues the ensured stability of 2D without compromising on the superior performance of 3D.\textsuperscript{[60]} 

2.5. Hybrid Chemical Vapor Deposition

One of the difficulties of the thermal vacuum deposition is controlling the vaporization of organic cation precursors caused by their low sublimation temperature. From this perspective, hybrid chemical vapor deposition (HCVD) draws attention based on sequential depositions generally by a thermal vacuum evaporation for inorganic precursors (e.g., PbI\(_2\) and PbBr\(_2\)) and a CVD for organic precursors (e.g., MAI and FALI).\textsuperscript{[61-63]} While a single composition of perovskite film was prepared by HCVD for PSCs in the beginning,\textsuperscript{[62]} a mixed composition has been consistently applied to HCVD for scalable PSMSs.\textsuperscript{[61,63]} Figure 8a,b represents a schematic illustration of HCVD, where the inorganic precursors of PbI\(_2\) and CsBr are deposited by co-evaporation, which is followed by CVD of FAI to prepare the perovskite film with a mixed composition.\textsuperscript{[62]} Figure 8c,e represents the cross-sectional and surface morphologies of the PbI\(_2\)/CsBr film (yellow film as shown in the inset of Figure 8e), prepared by co-evaporation with a ratio of 0.1 mm s\(^{-1}\) for PbI\(_2\) and 0.01 mm s\(^{-1}\) for CsBr to determine the composition of perovskite film. The CVD of organohalides typically occurs in a tube furnace, which is divided into two zones, upstream and
downstream. The organic precursor is converted to gas phase in upstream zone by heating (190°C for FAI and 210°C for MAI) and then delivered by the aid of a carrier gas (N₂, Ar, air, mixed gas O₂/N₂, etc.) to reach the predeposited inorganic precursors substrate in the downstream zone with a milder temperature (~150°C). [61–63] Figure 8d,f shows the cross-sectional and surface film morphologies of the resulting Cs₀.1FA₀.9PbI₂.9Br₀.1 perovskite film (black film as shown in the inset of Figure 8f) after the reaction between FAI and PbI₂/CsBr substrate, demonstrating a large grain size with a reduced roughness. [61]

On the other hand, the HCVD method has also been implemented with a vacuum oven for CVD instead of the tube furnace. [64,66] (PEA)₂(PEAI)₉ Pb₀.₁₅N₀.₈₅ was prepared by the HCVD method where the PbI₂ was deposited using thermal evaporation with a deposition rate of 1 Å under 5 × 10⁻⁴ Pa, and a mixed vapor of MAI and C₆H₅CH₂CH₂NH₃⁺ (PEAI) (MAI:PEAI = 9:1) was generated in the oven by heating the precursors at 160°C under the decreased pressure of 10 kPa. [64] The incorporated PEAI⁺ by the mixed-vapor deposition effectively passivates defect states, leading to an increase in PV performance with improved humidity stability. [64] Similarly, (BA)₉(MA)₀.₈₅Pb₀.₁₅I₀.₇₅ (BA⁺ = CH₃(CH₂)₃NH₃⁺) was deposited with the vacuum oven for the vapor generation of organic precursors, but the vapor was sequentially exposed to the substrate from MAI at 180°C to BAI at 120°C. [66] Recently, a long processing time for HCVD was greatly reduced by adopting a rapid HCVD (RHCVD) based on a tube furnace equipped with an IR heating and cooling system. [63] While the conventional CVD methods require a long processing time of ≈2–3 hours for ramping and deposition, and cooling processes, which would be fatal for a mass production of PSMs, RHCVD takes less than 10 min owing to a rapid ramping and deposition process provided by a movable IR heating component along the tube furnace. [63] The decreased processing time not only proposed a way for high throughput and low cost of PSM production, but also led to high film quality and uniformity over relatively large area.

2.6. Technology-Dependent PV Performance

The perovskite PV technology seems to pass through a transition stage from a scalable unit cell to a large module, which is now intensively developed by perovskite PV companies, such as...
Frontier Energy Solutions, Solliance, and Saule Technologies. Remarkable performance recently started to be reported by scalable technologies. Table 1 shows the PV parameters of PSCs and PSMs manufactured by scalable technology. The blade coating enabled a uniform perovskite coating over 46 cm² [20] by controlling the crystal growth of MAPbI₃ in the presence of MA acetate, where a complete unit cell (area = 0.125 cm²) showed a 18.14% of stabilized PCE at mp.[14] When the unit cell was scaled up to compose a mini-module (area = 16 cm²) with four subcells, the PCE reached 13.85%.[14] A 25 cm²-large PSM was also prepared through the formation of HMPA adduct, demonstrating 17.01% of PCE.[23] Remarkably, a sequential blade coating of PbI₂ and mixed cation salts (FAI, MABr, and MACl) produced a highly crystalline perovskite film, which was ascribed to the inclusion of nonvolatile solvent (tert-butylpyridine) in PbI₂ precursor solution to control the nanoporous PbI₂ structure for efficient conversion to perovskite crystal.[22] The completed 100 cm²-sized module with 12 series-connected subcells demonstrated 13.32% of PCE with promising stability results.[22] Recently, the blade coating of perovskite precursor solutions using 0.1 wt% of a multifunctional zwitterionic surfactant, tetradecyldimethyl(3-sulfopropyl)ammonium hydroxide inner salt, demonstrated a remarkable stabilized PCE of 20.93% from an active area of 0.8 cm² based on a blade-coated substrate of 3 × 2 cm, opening a way of high-quality perovskite film with full coverage.[29]

On the other hand, a 168.75 cm²-large module with 25 subcells was successfully prepared by slot-die coating (Figure 9a) and showed 11.1% of PCE based on active area (Figure 9b), giving a negligible PCE difference compared with that from the conventional spin-coating process.[69]

In case of the inkjet printing, few results have been reported on large size though the technology itself is considered suitable for scalable dimension. A vacuum deposition process has recently released notable results despite a limited number of groups being involved in the previous studies.[34,72,73] A large PSM based on the thermally co-deposition of MAI and PbI₂ demonstrated 18.13% of PCE (area = 21 cm²), showing only 10% difference in PCE compared with the unit cell (20.28% of PCE with 0.16 cm²).[54] Furthermore, the vacuum process allows a flexible substrate to be easily adopted owing to the low temperature process. A sequential vacuum deposition of PbI₂ and MAI was used for a large PSM based on both flexible and rigid substrate, composed of eight subcells with 2 cm². The flexible PSM showed a 13.15% of PCE (area = 16 cm²), which was comparable to that of the rigid substrate, 15.06%.[72] The HCVD method enables the vapor of organic precursors to be easily controlled during CVD by separate optimization either in tube furnace or vacuum oven. A mixed composition of Cs0.1FA0.9PbI₂Br0.1 via HCVD demonstrated a comparably lower PCE of 13.3% from a 0.09 cm²-sized unit cell due to a negative impact of a vacuum annealing process on underlying SnO₂ layer during the HCVD.[61] which was more enhanced by adopting RHCVD coupled with a systematic optimization, leading to a PCE of 15.5%.

When the unit cell was enlarged to compose a 22.4 cm²-sized module with seven cells in series, the PCE of 12.3% was achieved from the PSM based on RHCDV, whereas the conventional HCVD demonstrated a 91.8 cm²-sized PSM with a PCE of 9.34%.[61,63] In addition, soft cover deposition (SCD) was applied to produce a scalable perovskite film using a polyimide (PI) film.[74-76] The perovskite precursor solution was loaded on the substrate, which was covered by the PI film where the pressure is applied via compression plate. The peeling off the PI film with a speed of 50 mm s⁻¹ revealed the defectless crystallized perovskite film.[73] While the conventional SCD method showed a PCE of 15.7% from the 36.1 cm²-sized PSM, the low-temperature SCD method demonstrated 15.5% of PCE based on 5 cm² by controlling the thermal convection.[74] It is notable that the difference between PSC and PSM is substantial though the same deposition method was used. Strategies to reduce the performance decrease as increasing the area is summarized in Table 2 with expected advantage and disadvantage of a scalable deposition method.

3. Module Efficiency

3.1. Measurement Protocol for the Certification

The PV technology is evaluated by certified PCE based on stringent criteria to avoid uncertainties. Figure 10 shows a sequential protocol mostly taken from the technical report published by the National Renewable Energy Laboratory (NREL)[78] which satisfies the standard issued by International Organization for Standardization (ISO) 17025. 1) The area of a module is typically determined using a calibrated tape, whereas the area of a laboratory-scale cell is measured by a microscope with a 0.1 μm resolution. 2) The relative spectral responsivity is important to perform an appropriate calibration to minimize various errors originated from a photocurrent response of the cell, bandwidth, and waveform of the monochromatic light, and spatial uniformity of light source.[79] The spectral response is generally measured at short-circuit condition under a broadband DC bias light where the error sources are separated and refined.[80,81] 3) After the measurement of the relative spectral responsivity, a reference device is selected. It is recommended to choose a stable reference cell of the same material, because the calibration by the reference cell brings a removal of multiplicative errors during the spectral correction. Otherwise, a silicon cell is widely used as a reference cell for metastable devices on a research level. 4) The spectral mismatch parameter is determined using the measured spectral responsivities of a test cell and a reference cell, coupled with spectral irradiance of the light source.[81,82] 5) When the spectral mismatch parameter is obtained, the calibration is attempted with a reference cell by measuring its short-circuit photocurrent (Isc) at 25 ± 2 °C. The reference cell is placed at the location whose light level is equal to the average light intensity over the test module scale, which is calculated by a device placement software.[83] A different light source with various test bed is selected with considerations of the irradiance distribution and the module mountable bed size. 6) A solar simulator setup is adjusted to ensure the reference irradiance (1000 W m⁻²) where the measured Isc is within 2% of the spectrally corrected value from step 2). 7) The intensity fluctuation is measured by an intensity monitor depending on the location, particularly in module test beds. 8) The test cell or module is mounted on a designated place, related to the reference cell position in step 5), under the given reference condition (25 ± 2 °C and 1000 ± 20 W m⁻²).
Table 1. PV parameters of scalable PSCs* and PSMs* depending on the perovskite deposition method. PSM is composed of a series of cells, whereas PSC is based on a single cell. Area is distinguished by module size* and active area*. J_{SC} denotes the short-circuit photocurrent density.

| Method              | Perovskite composition    | Area [cm²] | J_{SC} [mA cm⁻²] | V_{OC} [V] | FF  | PCE [%] | Ref. |
|---------------------|---------------------------|------------|------------------|------------|-----|---------|------|
| Blade coating       | GA_{0.12}MA_{0.88}PbI_{3}  | 0.125⁷      | 21.03            | 1.19       | 0.78| 19.44   | [14]|
|                     | GA_{0.12}MA_{0.88}PbI_{3}  | 16⁵        | –                | –          | –   | 13.85   |      |
|                     | (FAPbI)_{3}(MAPbBr)₃   | 100⁴       | 1.80             | 11.8       | 0.62| 13.32   | [22]|
|                     |                           | 25⁺         | 3.47             | 6.71       | 0.71| 19.12   |      |
|                     | MAPbI₃                    | 8⁶ (16allen) | 5.82             | 3.98       | 0.68| 15.79   | [24]|
|                     | MAPbI₃                    | 0.09⁷       | 23.46            | 1.09       | 0.78| 20.08   | [17]|
|                     |                           | 1⁺          | 23.32            | 1.08       | 0.75| 19.12   |      |
|                     | MAPbI₃                    | 25⁺         | 3.52             | 5.41       | 0.68| 13.03   | [15]|
|                     | MAPbI₃                    | 0.09⁷       | 21.98            | 1.09       | 0.81| 19.41   | [25]|
|                     | MAPbI₃                    | 0.04⁷       | 23.68            | 1.21       | 0.78| 22.20   | [29]|
|                     | MAPbI₃                    | 0.08⁷       | 23.45            | 1.19       | 0.76| 21.02   |      |
|                     | (FAPbI)_{3.75}(CsPbBr)₃ | 18.66¹⁶ (25⁺)| 2.23            | 10.8       | 0.71| 17.01   | [21]|
|                     | CsPbI₃                    | 16          | 4.10             | 4.05       | 0.71| 11.87   | [67]|
|                     | CsPbI₃                    | 0.09⁷       | 20.67            | 1.12       | 0.82| 19.0    | [26]|
| Slot-die coating   | MAPbI₃                    | 0.06⁷       | 21.5             | 1.10       | 0.76| 18.0    | [28]|
|                     | MAPbI₃                    | 0.06⁷       | 2.85             | 5.73       | 0.63| 10.34   | [33]|
|                     | C_{0.15}FA_{0.85}PbI_{3}  | 0.01⁴       | 20.7             | 1.03       | 0.72| 15.2    | [37]|
|                     | MAPbI₃                    | 36²         | –                | –          | –   | 15.6    | [68]|
|                     | MAPbI₃₃Cl₃             | 149.5¹³     | –                | 20.8       | 0.71| 11.8    | [69]|
|                     | MAPbI₃₃Cl₃             | 168.8¹³     | –                | 21.2       | 0.68| 11.1    |      |
| Inkjet printing    | C_{0.05}MA_{0.45}FAPbI_{3} | 0.04⁴       | 23.48            | 1.11       | 0.76| 19.6    | [39]|
|                     | MAPbI₃                    | 0.04⁴       | 22.71            | 1.08       | 0.70| 17.04   | [41]|
|                     | MAPbI₃                    | 2⁻          | 22.45            | 1.04       | 0.66| 15.01   |      |
|                     | MAPbI₃                    | 2⁺          | 21.31            | 1.04       | 0.66| 14.67   |      |
|                     | MAPbI₃₃Br₃            | 4³          | 20.40            | 1.04       | 0.63| 13.27   |      |
|                     | MAPbI₃₃Br₃            | 2.02³       | 21.88            | 1.06       | 0.77| 17.74   |      |
|                     | MAPbI₃₃Br₃            | 0.08³       | 20.5             | 1.15       | 0.81| 19.0    | [70]|
| Vacuum deposition  | C_{0.1}FAPbI₃         | 0.04⁴       | 19.5             | 1.07       | 0.79| 16.6    | [48]|
|                     | FA₀.₅MA₀.₅PbI₃⁴         | –           | 22.6             | 1.09       | 0.76| 18.8    | [49]|
|                     | FAPbI₃₃Br₃(0.9Br₃)₃      | 0.15³       | 23.0             | 1.06       | 0.75| 18.1    | [55]|
|                     | MAPbI₃                    | 0.1⁴       | 22.61            | 1.05       | 0.72| 17.04   | [56]|
|                     | MAPbI₃                    | 0.01⁴       | 21.91            | 1.16       | 0.82| 20.83   | [51]|
|                     | MAPbI₃                    | 16¹         | 2.74             | 8.09       | 0.68| 15.06   | [72]|
|                     | MAPbI₃                    | 0.16¹       | 23.3             | 1.12       | 0.78| 20.28   | [54]|
|                     | MAPbI₃                    | 2¹          | 3.68             | 6.72       | 0.73| 18.13   |      |
| HCVD                | C_{0.05}FAPbI₃         | 91.8⁴       | 1.16             | 13.55      | 0.60| 9.34    | [61]|
|                     | MAPbI₃                    | 0.09⁴       | 21.7             | 1.06       | 0.68| 15.6    | [62]|
|                     | FAPbI₃                  | 8.8¹²       | 16.9             | 0.98       | 0.57| 9.5     |      |
|                     | C_{0.05}FAPbI₃         | 12¹²       | 17.8             | 0.94       | 0.54| 9.0     |      |
|                     | C_{0.05}FAPbI₃         | 0.1³       | 22.3             | 0.99       | 0.70| 15.5    | [63]|
|                     | C_{0.05}FAPbI₃         | 25¹       | 2.7              | 6.8        | 0.67| 12.3    |      |
|                     | (PEA)₂(MA)₁/₃PbI₃             | –           | 23.75            | 1.08       | 0.70| 18.08   | [64]|
|                     | MAPbI₃                  | 1¹         | 22.4             | 0.877      | 0.60| 11.7    | [73]|

⁷GA: guanidinium; FA: formamidinium.
Figure 9. a) Photograph and b) $I$–$V$ curves (black) and estimated power curve (red) of 6 in $\times$ 6 in-sized PSM based on 25 interconnected subcells. The module dimension is 168.75 cm.$^{[3]}$ Reproduced with permission.$^{[69]}$ Copyright 2020, Elsevier.

Table 2. Advantage and disadvantage of the deposition method with effective strategies resulting in the least PCE loss as increasing the size of devices.

| Method          | Advantage                                      | Disadvantage                                             | Strategy to suppress the performance reduction with increasing the area |
|-----------------|------------------------------------------------|----------------------------------------------------------|------------------------------------------------------------------------|
| Blade coating   | Relatively simple process$^{[70]}$              | Difficult to supply the continuous ink$^{[68]}$           | Use of additive, tBP (20% PCE drop by increasing the area from 25 to 100 cm$^2$)$^{[20]}$ |
| Slot-die coating| Preset of a wet film thickness$^{[27]}$         | Additional treatment to control the crystal growth (e.g., gas quenching)$^{[20,23]}$ | Employment of high-pressure nitrogen extraction to assist crystallization (13% PCE drop by increasing the area from 0.09 to 7.92 cm$^2$)$^{[70]}$ |
| Inkjet printing | Noncontact process (no stress during deposition)$^{[42]}$ | Strict requirements for ink composition$^{[41]}$          | Addition of DMSO as an additive in the mixed solvent of DMF/NMP (26% PCE drop by increasing the area from 0.1 to 4.04 cm$^2$)$^{[39]}$ |
| Vacuum evaporation | Solvent-free process$^{[45]}$                  | Difficult to control the vapor of organic precursors (e.g., MAI)$^{[48]}$ | Change of ETL from TiO$_2$ to SnO$_2$ Surface treatment based on MAI and potassium acetate (11% PCE drop by increasing the area from 0.16 to 21 cm$^2$)$^{[34]}$ |
|                 | Preset of a wet film thickness$^{[27]}$         | Stringent parameters optimization of each precursor for reproducible evaporation$^{[35]}$ |                                                                  |
|                 | High compatibility with industry standard (e.g., roll-to-roll process)$^{[21]}$ |                                            |                                                                  |
|                 | Low-temperature process$^{[35,34]}$             |                                            |                                                                  |
|                 | Mature technology in industrial fields$^{[46]}$ |                                            |                                                                  |
| HCVD            | Independently optimized condition for inorganic and organic precursors$^{[77]}$ | Long deposition time (2–3 h)$^{[61,62]}$                  | Employment of RHCVD (21% PCE drop by increasing the area from 0.1 to 22.4 cm$^2$)$^{[63]}$ |

Figure 10. Schematic procedures of efficiency measurement suggested by NREL technical report (NREL/TP-5J00-66873).
The performance of the test cell or module is indicated by current–voltage (I–V) characteristics. The voltage sweep is made to record the corresponding photocurrent at a given voltage from short-circuit to open-circuit condition. The open-circuit voltage ($V_{OC}$) is indeed measured before and after the I–V scan to evaluate the metastability. The measured $I_{SC}$ is corrected by considering the intensity fluctuation measured in step 7). An uncertainty analysis software package can calculate the sample-specific uncertainty and, thus, help the correction, determining the calibrated $I_{SC}$ and the maximum power ($P_{max}$). The I–V data points are recorded with a variety of information, such as the calibrated $V_{OC}$, $I_{SC}$, $P_{max}$, temperature, time, cell type, manufacturer, reference cell and its calibration, and the change in $V_{OC}$ before and after the I–V measurement.\[78\]

### 3.2. Efficiency Status of PSMs

**Figure 11** and **Table 3** represent the certified champion module efficiencies depending on the PV technology, published by NREL.\[85\] Panasonic recorded 16.1% of PCE on the PSM category with an area of 802 cm$^2$ in 2019, accredited by Advanced Industrial Science and Technology (AIST) in Japan, which was further enhanced to 17.9% under mpp tracking in 2020 by the same company. Though the stabilized $I_{SC}$ and $V_{OC}$ were not reported together with the efficiency, the progress in PSM is highly impressive when considering that a first module certification was merely made in 2018 by Toshiba with a submodule type (44 cells, 703 cm$^2$), demonstrating 11.7% with remarks that the efficiency was roughly measured from the not-stabilized initial performance. On the other hand, one of the most commercialized silicon modules showed the best efficiencies of 24.4% for monocrystalline Si module (13 177 cm$^2$ hetero/rear-junction with 108 cells, prepared by Kaneka in 2016) and 20.4% for multicrystalline Si module (14 818 cm$^2$, 60 serial cells, prepared by Hanwha Q-cells in 2019). In case of an amorphous Si type, a single junction-based module with 9.8% was certified in 1990 and has suspended, whereas the two junction-based modules have been occasionally reported, showing the best efficiency of 12.3% in 2014 by TEL Solar (14 322 cm$^2$, stabilized at manufacturer to the 2% level according to the IEC criteria). Among III–V compound semiconductor, single-crystalline GaAs showed a remarkable certified efficiency of 25.1% by Alta Devices in 2017 based on a small module size of 866.45 cm$^2$. When the four junctions were built by a GaInP/GaAs/ GaInAsP/GaInAs bonded structure, the efficiency reached 38.9% under lens with a concentration of 333 (812.3 cm$^2$, outdoor measurement). For chalcogenide PV technology, a CdTe thin film demonstrated 19.0% efficiency with large module scale (23573 cm$^2$) in 2019.

**Table 3. Certified champion module efficiencies depending on PV technology.**\[84\] Modules based on a single junction without concentrator.

| PV technology | Companies/institutions | Module size [cm$^2$] | PCE [%] |
|---------------|------------------------|----------------------|---------|
| Perovskite    | Panasonic              | 802                  | 17.9    |
| Si monocrystalline | Kaneka             | 13 177               | 24.4    |
| Si multicrystalline | Hanwha Q-cells        | 14 818               | 20.4    |
| Amorphous Si  | Solaris                | 933                  | 9.8     |
| GaAs          | Alta Devices           | 866.45               | 25.1    |
| CdTe          | First Solar            | 23 573               | 19.0    |
| CIGS          | Solar Frontier         | 841                  | 19.2    |

**Figure 11.** Certified champion module efficiencies depending on PV technologies. The chart “Champion PV Module Efficiency Chart” is reproduced with permission.\[85\] Copyright 2020, NREL.
and copper indium gallium selenide (CIGS) showed 19.2% efficiency with small module scale (841 cm²) in 2017, particularly with Cd-free composition though the accurate composition was not defined.

It is apparent that the efficiency of PSM is still required to be much enhanced to compete with other existing PVs from the industrial point of view. The technology on PSM has been consistently evolved as an emerging PV technology, which recently shows a rapid increase in module efficiency, whereas the conventional PV technologies show a long plateau in the module efficiency chart.\(^{(83)}\) The efficiency difference of monocrystalline Si between the certified cell (PCE = 26.1%) and module (PCE = 24.4%) is only 1.7%, whereas the perovskite technology still exhibits a significant difference of 7.6% between the certified cell (PCE = 25.5%) and module (PCE = 17.9%), indicating still immature technology of modules as well as much room for development.\(^{(3,85)}\) PSCs have a peculiarity in the perovskite material as well as in the device structure. The composition of perovskite has been consistently evolved and now converged into the mixed-cation and -halide composition for efficient and stable device performance.\(^{(86)}\) While a key material for light harvesting in other PV technologies has a relatively definite composition (e.g., Si, CIGS, and CdTe), the perovskite crystal exhibits a wide window for compositional engineering as long as the tolerance factor is satisfied,\(^{(86)}\) which would be beneficial to bandgap engineering. The wide range of composition with various precursors, however, brings a difficulty in controlling a film quality over large area,\(^{(86)}\) closely related to the crystal growth. Therefore, reproducing the film quality by other scalable deposition methods unavoidably left plenty of parameters to be considered to control the growth of perovskite crystals for large-scale PSMs.\(^{(86,87)}\) Furthermore, the perovskite material as an intrinsic layer requires two selective contacts (electron transporting material [ETM] and hole transporting material [HTM]) for respective charge extraction, whereas most of light absorption layers in other PV technologies already act as either p-type or n-type layer as well as light harvesting layer at the same time. The perovskite with two neighboring layers further brings more components to be optimized when being scaled up to PSM. Unlike CIGS and CdTe, an absence of chemical bond with adjacent selective contacts in PSC is likely to make the interface physical junction significantly dependent on the process.\(^{(86,88)}\) Nevertheless, the parameters causing the huge difference between PSC and PSM have been resolved in each scalable deposition method, gradually narrowing the gap.\(^{(22,39,54,63,69)}\)

In addition, outdoor performance test of perovskite mini-module (17 and 50 cm²) in varied atmospheric conditions, 18–42 °C for temperature and 0–1200 W m⁻² for irradiance intensity, supports the potential of PSMs, which results in comparable PV parameters with commercial Si modules.\(^{(89)}\) The observed positive dependence of $V_{OC}$ on the temperature under high irradiance particularly makes the PSM more competitive with conventional PV modules.\(^{(89)}\) Furthermore, PSMs are highly promising in terms of lightweight,\(^{(90)}\) exhibiting 29 W g⁻¹ power-per-weight compared with that of 8.31 W g⁻¹ for Si-solar module.\(^{(90)}\) The ultrathin film technology opens up a way for versatility of PSMs by granting flexible and stretchable structure.\(^{(89,90)}\)

### 4. Module Stability

#### 4.1. Measurement Protocol for the Certification

While the efficiency of PSCs has reached over 25% in a short period,\(^{(3)}\) the device stability has been thoroughly studied to estimate a market potential of PSMs as commercial products. The early study was mainly carried out on laboratory scale with a few limited conditions, which were roughly taken either from the international summit on OPV stability (ISOS) or the IEC standards.\(^{(91,92)}\) As various stability tests with the primitive conditions started to produce promising results,\(^{(91,92)}\) the IEC standard for terrestrial PV modules is now actively encouraged to be applied to PSMs.\(^{(93)}\) The IEC 61215, composed of systematic and methodological test procedures for specific stresses, was particularly designed to anticipate a long-term performance of solar modules by intentionally providing harsh conditions to accelerate the module aging. The IEC 61215 is considered as a minimum requirement to enter the PV market. Figure 12 shows a flowchart of module test based on IEC 61215, which consists of standard test condition (Sequence A), outdoor exposure and hot spot endurance test (Sequence B), humidity freeze (HF) test (Sequence C), thermal cycling (TC) test (Sequence D), and damp heat (DH) test (Sequence E). The standard test condition is given as the temperature of 25 ± 2 °C under an irradiance intensity of 1000 ± 100 W m⁻² according to IEC 60904-1. The wet leakage current test refers to the insulation test by measuring insulation resistance of the module being immersed in a solution (<3500 Ω cm⁻¹) and applied with a high voltage (>500 V). The outdoor exposure test allows to assume the actual degradation of modules at local area with a normal latitude of ±5° by applying bias or load equal to mpp under the irradiance of at least 60 kWh m⁻².\(^{(95)}\) On the other hand, the hot spot endurance test in Sequence B examines the stability under reverse bias, which can be fostered by partial or whole shadowing. The reverse bias tends to exhaust power, possibly leading to a rapid overheating or breaking down the junction. The module is applied with maximum power at 50 ± 10 °C under 1 kW m⁻² for at least 1 h (<5 h) to derive the worst case by reverse bias effect.\(^{(94)}\)

In Sequence C, the UV precondition test is performed by exposing modules at 60 ± 5 °C to UV light between 280 and 400 nm whose irradiance is lower than 250 W m⁻².\(^{(93,94)}\) The TC test in Sequences C and D is facilitated by a climatic chamber equipped with a temperature control, where the condensation is suppressed. The temperature is varied between -40 and 85 °C with a minimum dwell time of 10 min at both extreme temperatures, whereas the heating and cooling rate is set between 45 and 100 °C h⁻¹. It is also noted that a certain level of current ($-I_{mpp}$) should be applied during the heating process. The number of cycles is then determined by the test sequence either C or D. The HF test in Sequence C is also monitored in the climatic chamber whose temperature is varied again between -40 and 85 °C but with different dwell time and heating and cooling rate, because the dwell time at 85 °C should be kept at least for 20 h, whereas -40 °C is maintained at least for 30 min.\(^{(91,94)}\) The temperature heating and cooling rate is kept between ±75 and ±100 °C h⁻¹ above 0 °C, whereas the rate can be increased between ±75 and ±200 °C h⁻¹ below 0 °C. The DH test in Sequence E is conceived to study the effect of humidity exposure
for a long period of time. The temperature is maintained at 85 ± 2 °C with a relative humidity (RH) of 85 ± 5% for 1000-1048 h. The hail and static mechanical load tests in Sequence E are carried out by hitting ice balls (7.53 g) with a fast speed of 82.8 km h⁻¹ and mounting a static load (>2400 Pa) for 1 h on the module, respectively.
The current commercial PV technologies (Si, GaAs, etc.) not only completely passed the full IEC 61215 criteria but also are challenged by more intense conditions to guarantee 25-year-long PV lifetime. Figure 13 represents the product qualification program issued in 2020 by PV Evolution Labs (PVEL), where the back sheet durability, light and elevated temperature-induced degradation (LeTID), and mechanical stress are newly included to enable objective evaluation of industrial suppliers. Furthermore, the efforts to establish more proper standards for solar modules have been continuously made by IEC, PV Quality Assurance Task Force, and PVEL.\(^6\)

4.2. Stability Status of PSMs

Though PSMs were encouraged to be tested according to the full systematic standards, such as IEC 61215, in laboratories, extremely simplified conditions are selectively applied for the stability test of PSCs or PSMs. The most representative test for perovskite PV was either the temperature test at 85°C or the humidity test at 85% RH.\(^{91,95}\) As the stability of perovskite has been gradually established, the operational stability at mpp under the full sun irradiance (1 kW m\(^{-2}\)) is frequently reported for PSCs.\(^{12,13}\) One of the parameters hindering the perovskite PVs from obtaining the long-term stability is the widely used HTM of 2,2',7,7'-Tetrakis[N,N-di(4-methoxyphenyl)amino]-9,9'-spirobifluorene (spiro-MeOTAD) in the presence of bis(trifluoromethane)sulfonimide lithium salt (LiTFSI) dopant.\(^{100}\) A simple replacement of the dopant of spiro-MeOTAD had a positive effect both on efficiency and stability, showing an even 2% increase over 600 h at mpp tracking under the full sun intensity at room temperature.\(^{13}\) Otherwise, a replacement of spiro-MeOTAD has been generally tried to ensure the long-term stability of devices. PTAA was introduced

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**Figure 13.** Product qualification program being widely adopted by PV manufacturers. TC, DH, UV, HF, MSV: maximum system voltage, LeTID, PAN: “.PAN” extension file with Pvsyst software to specify the module performance, and IAM: incidence angle. Reproduced with permission.\(^95\) Copyright 2020, DNVGL.
as HTM, leading to an enhanced operational stability with an initial PCE drop by 10%, from 20% to 18%, over 250 h.\textsuperscript{12} Furthermore, an incorporation of CuSCN as HTM, coupled with reduced graphene oxide at the interface, resulted in a remarkable operational long-term stability at 60 °C, showing a 95% of initial PCE over 1000 h.\textsuperscript{[12,67,96]} More recently, a long-term stability of PSMs has been reported.\textsuperscript{[12,67,96]} It is well known that 3D perovskites are vulnerable to moisture, making an encapsulation indispensable. The 2D perovskites, on the other hand, exhibit significantly enhanced moisture and thermal stabilities regardless of their inferior PV performance.\textsuperscript{[97]}

To this end, the 3D/2D bilayered structure has been developed to take the advantages of both 3D layer in performance and 2D layer in stability.\textsuperscript{[60]} The formation of 3D/2D bilayered PSCs based on a 3D bulk film of MAPbI\textsubscript{3} with an ultrathin 2D capping layer of (HOOCC(CH\textsubscript{2})\textsubscript{4}(NH\textsubscript{3}))\textsubscript{2}PbI\textsubscript{4} demonstrated a PCE of 14.6% for a typical architecture with a gold contact and 12.9% for a monolithic structure, in the absence of HTM, with a carbon contact.\textsuperscript{[96]} When the HTM-free monolithic architecture was scaled up to a 100 cm\textsuperscript{2}-sized module, producing 11.2% of PCE active area of 47.6 cm\textsuperscript{2}, a superior long-term stability over 10000 h was ensured under full sun intensity at 55 °C without any encapsulation in ambient air.\textsuperscript{[96]}

The employment of poly(3-hexylthiophene) as HTM with a thin 2D capping layer grown from n-hexyl trimethyl ammonium bromide on 3D perovskite indeed led to a great operational stability, showing 95% of an initial PCE over 1370 h under full sun at mpp.\textsuperscript{[96]} The moisture stability was also enhanced, showing a PCE drop by 20% over 1008 h at 85% RH without any encapsulation. A 23% of stabilized PCE of PSC was reduced to 16% when the unit cells composed a 25 cm\textsuperscript{2}-large module, whereas the module stability was unfortunately not reported.\textsuperscript{[98]}

Similar to the bilayered structure, graphitic carbon nitride was applied as a 2D diffusion barrier (DBL) on top of 3D surface.\textsuperscript{[68]} Figure 14a shows a photograph of 36.1 cm\textsuperscript{2}-large PSM based on DBL, demonstrating 15.6% of PCE from the I–V reverse scan (Figure 14b).\textsuperscript{[68]} The encapsulated PSM showed an outstanding thermal, moisture, and operational stabilities, where 95% of initial PCE was retained after 1000 h under 85% RH and at 85 °C (Figure 14c), whereas 91% of initial PCE was maintained over 1000 h under full sun intensity at mpp condition (Figure 14d).\textsuperscript{[68]}

Moreover, α-CsPbI\textsubscript{3} was adopted along with a surface treatment by 18-crown-6 ether, aiming at the long-term stable PSMs.\textsuperscript{[68]} The interface engineering of α-CsPbI\textsubscript{3} was responsible for the enhanced thermal and moisture stabilities of PSCs. The encapsulated PSM, prepared in the same manner, retained 95.2% of initial PCE (~11.8%) over 500 h at operational condition.\textsuperscript{[64]} A holistic interface stabilization strategy was finally made to respective components in the PSM, including ETM, perovskite, HTM, and encapsulation, leading to the superb operational and thermal stabilities.\textsuperscript{[99]} The encapsulated 22.4 cm\textsuperscript{2}-large PSM maintained 86% of initial efficiency over 2000 h at operational condition and more than 80% of initial PCE after 1000 h-long thermal stress at 60 °C. Table 4 summarizes the stability results of PSCs and PSMs with respective aging condition.

5. Cost Efficiency

PSMs have a strong advantage in manufacturing cost over the conventional PV technologies. A key material of the PSM is lead, which is far from rare-earth elements and costs only $2 m\textsuperscript{2}.

![Figure 14. a) Photograph and b) I–V curve of 36.1 cm\textsuperscript{2} large PSM based on the 2D BDL of graphitic carbon nitride. Forward scan (black) and reverse scan (red) were measured from \textit{I}_{SC} to \textit{V}_{OC} and from \textit{V}_{OC} to \textit{I}_{SC}, respectively. Long-term performance of encapsulated PSMs with c) heating at 85 °C and 85% RH for 1000 h and with d) operational condition at 60 °C and mpp under the full sun intensity as a function of the presence of DBL. Reproduced with permission.\textsuperscript{[68]} Copyright 2019, Cell Press.](image-url)
Table 4. Reported stability results of PSCs\(^a\) and PSMs\(^b\). Area is distinguished by module size\(^a\) and active area.\(^a\)

| Test condition       | PCE [%] | PCE\(_{0}/\text{PCE}_{1} \times 100\) | Time [h] | Area [cm\(^2\)] | Ref. |
|----------------------|---------|--------------------------------------|----------|-----------------|------|
| Temp.                | RH      | Light                               | Voltage  |                 |      |
| –                    | inert   | full sun                             | mpp      | 22 (stabilized)  | 102% | 600 | 0.16\(^{±1}\) | [13] |
| 25 °C                | inert   | full sun                             | mpp      | 21.1 (stabilized)| 90%  | 250 | –\(^{*}\) | [12] |
| 60 °C                | encap   | full sun                             | mpp      | 20.5 (stabilized)| >95% | 1000 | 0.16\(^{±1}\) | [11] |
| 65 °C                | encap   | dark                                 | mpp      | 18.8             | 90%  | >250 | –\(^{*}\) | [49] |
| 55 °C                | ambient air | full sun                             | short-circuit | 11.2 | ≈100% | >10000 | 47.6\(^{±0}\) (100\(^{±0}\)) | [96] |
| 25 °C                | 85%     | dark                                 | mpp      | 23.0 (stabilized)| ≈80% | 1008 | –\(^{*}\) | [98] |
| –                    | 30% (encap) | full sun                             | mpp      | 95%             | 1370 | 25\(^{±*}\) |      |
| 85 °C                | 85% (encap) | dark                                 | –        | 15.6            | 95%  | 1000 | 36.1\(^{±0}\) | [68] |
| 60 °C                | ambient air (encap) | full sun                             | mpp      | 91%             | 1000 |      |      |
| 85 °C                | N\(_2\) | dark                                 | –        | 16.91           | 91%  | 1000 | 0.1\(^{±0}\) | [67] |
| 25 °C                | 20%     | dark                                 | –        | 90%             | 2000 |      |      |
| 25 °C                | 60% (encap) | full sun                             | mpp      | 11.8            | 95.2%| 500  | 8\(^{±1}\) (16\(^{±1}\)) | [99] |
| –                    | <5% (encap) | full sun                             | mpp      | 16.6            | 86%  | 2000 | 22.4\(^{±1}\) |      |
| 60 °C                | –       | –                                    | >80%     | 1000            |      |      |      |
| –                    | inert   | full sun                             | mpp      | 12.3            | 90%  | 800  | 25\(^{±*}\) | [63] |
| 25 °C                | full sun | mpp                                  | 9.34     | 80%             | 500  | 100\(^{±0}\) | [61] |

\(^a\)PCE\(_{0}\): initial PCE; PCE\(_{1}\): terminated PCE; full sun: 1 kW cm\(^{-2}\); encap: encapsulation with inert atmosphere.

Other materials including ETM and HTM are estimated to cost around $20 cm\(^{-2}\), whereas a front glass with sputtered transparent conductive oxide (TCO) such as FTO and ITO is considered as the most expensive material, costing $10 cm\(^{-2}\).[101] Therefore, a total production cost of $40 cm\(^{-2}\) is calculated for manufacturing of PSMs,[101] which can be further reduced by altering the manufacturing process or adopting low-cost materials for ETM and HTM. Song et al. demonstrated a strong economic potential for PSCs by cost analysis based on a bottom-up cost model, where ITO/NiO/MAPbI\(_3\)/ZnO/Al was chosen for the feasible device architecture for manufacturing process of PSMs.[102] The material cost still accounted for 76% of the total manufacturing cost, including materials, utilities, labor, maintenance, and depreciation costs. From the cost analysis, $31.7 ± 5.5 m\(^{-2}\) of the manufacturing cost with $0.41/W\(_{p}\) of the minimum sustainable price was calculated for the PSM, providing a levelized cost of energy (LCOE) of 4.93-7.90 US cent kWh\(^{-1}\) under the assumption of 16% of PCE for a 30 year-long unsubsidized condition in the United States.[102] When the PSM is assumed to demonstrate 20% efficiency for 15 year-long lifetime, the LCOE is further decreased to 3.5 US cent kWh\(^{-1}\).[103] On the other hand, the manufacturing cost and LCOE for conventional multicrystalline Si modules are currently estimated as $80 m\(^{-2}\) and 5.5 US cent kWh\(^{-1}\), respectively.[105,109] Regarding crystalline Si modules, the estimated LCOE is in the range of 7.04-11.90 US cent kWh\(^{-1}\) based on an efficiency of 33.6% with 15 year-long lifetime.[102,104] In case of CIGS and CdTe PV technologies, the only process cost already reaches $27-29 m\(^{-2}\) compared with $6.8 ± 1.2 m\(^{-2}\) for the PSM.[102] While the PSM apparently takes the lead in the manufacturing cost, the LCOE value is comparable to that of Si module. When considering that the LCOE, defined as a ratio of the total cost of a PV to the total energy generated by the PV during its whole life cycle, is significantly dependent on the efficiency and lifetime of device, the establishment of PSMs as efficient as PSCs with ensured stability will definitely reduce the LCOE and surpass the Si PVs.

6. Safety Issue

6.1. Environmental Impact of Pb Species

The toxicity of Pb has been a consistent concern from the perspective of commercialization of PSM technology.[105] Lead leakage from PSMs was, therefore, assessed by assuming accidents to study the health and environmental hazards associated with Pb.[105] The accident scenarios, allowing the lead release, can be roughly categorized into flooding safety and fire safety. The former flooding safety covers situations where water invades into damaged panels, leading to a decomposition of perovskite to PbI\(_2\).[106] The PbI\(_2\) would substantially flow in the groundwater though its solubility constant is extremely low on the order of 10\(^{-8}\).[107] According to the conducted experiments,[106] 1 g of Pb leaking from 1 m\(^2\)-sized panel showed a high Pb concentration of 70 ppm mostly near the surface (first a few cm below from the ground). The scenario results indicate the higher Pb level than the uncontaminated environment (<10-30 ppm); however, it is still lower than the low level of Pb contamination ranging between 150 and 400 ppm.[106] Nevertheless, the Pb leakage through the water should be taken seriously because of its peculiarity.[105] Pb is not only prone to be strongly adhesive to the soil, which prevents Pb from being easily dispersed,[106] but also more bioavailable compared with the other sources of the lead contamination, threatening a food cycle.[107,108] For
the latter case of the fire safety, the degraded component of PbI₂ at high temperature generated PbO and PbO₂ by reacting with oxygen. The Pb-containing combustion was readily evacuated into environment unless the encapsulation system captured them under a low flame (<760 °C) which stresses out the importance of encapsulation technology for PV panels.

6.2. Strategy for Handling Pb Hazard

The efforts to reduce the Pb hazard have been intensively made with various strategies ranging from a reduction of Pb contents, to a sequestration of Pb, and a development of recycling technology. Pb-less or Pb-free perovskite materials have been widely studied as a direct way to reduce the Pb content in the material. The most promising component so far is tin, Sn₂⁺, whose tendency to be oxidized to Sn⁴⁺ easily results in unstable phase stability of Sn-based perovskite. The phase stability of Sn-based perovskite has been, therefore, reinforced by many approaches including a component engineering. The cutting edge technology of Pb-free perovskite demonstrated PCE over 10%, reaching 13.24% of PCE in 2020. However, the apparent PCE gap between Sn- and Pb-based PVSs is likely to derive a skeptical response. An alternative way to handle the Pb safety is passively prepared by devising a Pb sequestration architecture to cope with an accident and establishing a recycle system to control Pb uptake. The Pb sequestration, so-called “safe-by-design,” is devised to prevent the Pb release from an uncontrolled situation. A porous metal–organic framework (MOF) polymer composite was utilized for the safe-by-design. The designed MOF polymer composite effectively acted as a Pb scavenger in the presence of water, converting the water to reach the drinkable water limit. The MOF polymer composite was successfully scaled up for a mini-module with a size of 45.5cm². Besides MOF, ethylene vinyl acetate (EVA), sandwiched between a back glass and a PSM, is found to effectively reduce the Pb leakage rate from a damaged PSM based on the self-healing property by cross linking of the polymers. The encapsulation with EVA not only suppressed the Pb leakage, in case of glass damage by mechanical impacts, but also improved operational stability, maintaining 90% of initial PCE after 200 temperature cycles. Furthermore, the extraction of Pb species during the recycling process can be another attempt, where Pb is forced to be separated from the Pb-contained waste by the aid of electromagnetic coils in the presence of hydroxyapatite/Fe (HAP/Fe) as a scavenger, leading to a negligible Pb content in output (<15 ppb). It is noted that the Pb-adsorbed HAP/Fe can be recycled by 99.97% to reproduce PbI₂, indicative of a feasible management process.

6.3. Green Solvent

A scalable transition from PSC to PSM suffers from toxic solvents including anti-solvent and precursor solvents used to fabricate unit cells showing high efficiency. There is no doubt that the toxic solvents should be replaced with other green solvents for a successful module implementation of the perovskite PV technology. The anti-solvent-assisted fast crystallization is one of the most widely used methods to produce a high quality of the perovskite film. Though the use of anti-solvent based on the spin-coating is restricted, the anti-solvent method still has a chance to be incorporated to other scalable deposition methods, such as bathing and inkjet printing. Chlorobenzene and toluene are generally used as the anti-solvent but hardly recommended for scalable process due to their toxicity and low volatility. From this perspective, environmental friendly solvents with relative polarity ranging between 2 and 4.5 have been tried to reduce the safety issue. The ethyl acetate drew interest by forming the hydrogen bond with DMSO in the perovskite precursor solution, which notably retards the crystallization to enhance the film quality, being responsible for PCE of 19.43% compared with that of 18.67% for chlorobenzene as anti-solvent. The addition of hexane to ethyl acetate was found to passivate the surface defects and, thus, produce a pinhole-free perovskite film. Propyl acetate and isooctane were found to affect the PbI₂ nucleation and induce the rapid crystal growth of perovskite. Furthermore, alcohols (e.g., isobutyl alcohol, n-butanol, n-propanol, and ethanol) were also tried as anti-solvent in spite of concern about decomposition induced by alcohols. The improved performance was monitored when the mixture of ethanol and chlorobenzene (1:3) was used as the anti-solvent. On the other hand, the perovskite precursor-coated substrate bathing in n-butyl alcohol resulted in a notable PCE of 17.67% and 13.85% from 0.16 cm²-sized PSC and 100 cm²-sized PSM, respectively, owing to the reduced defects. Isodipropyl ether along with ether was used to induce intermediate phase and remarkably exhibited a high PCE of 21.26%. Regarding the green solvent for perovskite precursors, only few results have been reported to replace the conventional solvents, such as DMF, DMSO, and NMP. The mixture of polyethylene glycol and GBL was used as a solvent for PbBr₂, followed by deposition of CsBr in water using a sequential deposition method. Finding a promising candidate of green solvent for perovskite precursors is significantly challenging due to the solubility problem. Therefore, the scalable deposition method based on precursor vapors tends to receive attention from the environmental perspective. Nevertheless, efforts have been consistently made to take various advantages of a solution-based process.

7. Conclusion and Perspective

Perovskite PVSs have a significant potential for commercialization based on a certified high PCE as well as a remarkable stability of PSCs. Scalable coating technologies, such as blading coating, slot-die coating, inkjet printing, and vacuum deposition, have been accordingly adapted to PSMs, presenting promising results though each technology is not fully matured yet. A PCE over 17% was demonstrated from a 19 cm²-large PSM by blade coating, whereas the slot-die coating showed over 11% of PCE of a mini-module based on an area of 168.6 cm². Notably, thermal vacuum deposition showed 18.13% of PCE based on an area of 21 cm². It is likely for PSMs to have a chance soon to get compared with conventional PVSs. To further compete with other PV technologies, it is highly required to satisfy the IEC standard to provide objective and reliable performance data. A specific measurement or aging condition, which has been hardly studied with
PSMs, for example, reverse bias test, HF test, and mechanical stress test, is more encouraged to be investigated. In addition, Pb-safety issue should be resolved for commercialization, approached either by development of Pb-free/-less perovskite or by establishment of Pb control system including the Pb sequestration under accident and Pb extraction during recycling process. Finally, toxic solvents in the current manufacturing process are strongly recommended to be replaced with green solvents for the commercialization of PSMs.

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Conflict of Interest
The authors declare no conflict of interest.

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Research data are not shared.

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