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Challenges for commercializing perovskite solar cells

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Abstract

Perovskite solar cells (PSCs) have witnessed rapidly-rising power conversion efficiencies, together with advances in stability and upscaling. Despite these advances, their limited stability and their need to prove upscaling remain crucial hurdles on the path to commercialization. We summarize recent advances toward commercially-viable PSCs and discuss challenges that remain. We discuss the development of standardized protocols to distinguish intrinsic and extrinsic degradation factors in perovskites. We review accelerated aging tests in both cells and modules, an discuss the prediction of lifetimes based on degradation kinetics. Mature photovoltai solutions, which have demonstrated excellent long-term stability in field applications, offer the perovskite community valuable insights into clearing the hurdles to commercialization.

One Sentence Summary: Developments in perovskite solar cells are summarized and remaining challenges to commercialization portrayed.

Hybrid organic-inorganic perovskite solar cells (PSCs) have recently emerged as a promising photovoltaic (PV) candidate (*1, 2*). Perovskites have a general chemical formula of AMX_3, in which the M metallic cations and X anions form MX_6^{4-} octahedra with A cations occupying the 12-fold coordinated holes within the cavity. These materials exhibit desiring optical-electrical properties for PV devices, such as suitable and tunable bandgaps, strong optical absorption, long carrier diffusion length and high defect tolerance (*3*). The record certified power conversion efficiency (PCE) of PSCs has recently risen to 22.7% (*4-7*) - for a small-area device (~0.09 cm^2) whose active area is well below that of standard commercial crystalline silicon solar cells. Perovskite films are made using solution-based fabrication and of earth-abundant materials (*8*) (**Fig. 1A**), but the most efficient devices rely on costly gold and organic hole transport materials such as 2,2',7,7'-tetrakis(*N*,*N*-di-p-methoxy-phenylamine)-9,9'-spirobifluorene (spiro-OMeTAD).

Although the potential for low-cost PSCs offers a promising prospect for commercializing this PV technology, a complex interplay has unfolded between pursuing high efficiency and other commercially viable aspects – long-term stability and scale-up manufacturing. This review aims to provide an overview of recent progress toward the commercialization of PSCs.

Device configurations and performance

PSCs have layered device structure that includes a transparent conductive oxide (TCO)-coated glass substrate, an n-type semiconductor as the electron-transport-layer (ETL), a perovskite absorber layer, a p-type semiconductor as the hole-transport-layer (HTL), and a back contact (metal, or TCO, or carbon) (**Fig. 1B**). In mesoscopic PSCs developed following dye-sensitized solar cells (DSSC) investigations, the perovskite absorber sensitizes a mesoporous metal oxide layer (*e.g.* meso-TiO_2) used as a scaffold (*4, 9*). In planar PSCs, the perovskite layer is sandwiched between a planar ETL (*e.g.* compact TiO_2 (c-TiO_2), SnO_2, C_{60} and its derivatives) and a HTL (*e.g.* spiro-OMeTAD, PTAA and Poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) (PEDOT:PSS)) (*10-13*). Depending on the sequence of depositing the ETL and the HTL, the structure can be divided into formal (conventional) or inverted architectures (*14, 15*). We note that the design and fabrication of inverted devices is not a simple reversal from the formal device structure, and the two device structures typically select different ETLs and HTLs mainly due to the restrictions in the choice of processing solvents. In triple mesoscopic PSCs, the perovskite is deposited on a triple-layer scaffold, made of screen printed mesoporous TiO_2 layer, ZrO_2 spacer layer and carbon electrode (*16-18*); such devices use carbon electrodes to replace the noble metal back contacts and do not require a hole-conducting layer.

The prospects of PSCs as a top-cell for constructing tandem solar cells with other PV devices, such as silicon (*19*), copper indium gallium selenide (CIGS) (*20*), low-bandgap perovskites (*21*), have been reviewed elsewhere (*22*). Partnership with existing mature PV technologies not only boosts their efficiency beyond the Shockley-Quiesser (SQ) limit of single-junction devices (*23*), but also provides access to established market channels (*24*).

Progress in the efficiency of single-junction PSCs is summarized in **Fig. 1C** (*4-7, 12, 16, 25-37*). The perovskite layer and charge transporting layers are typically deposited by spin-coating for small-area (< 1.0 cm^2) lab cells. Enlarging the device area with current techniques to fabricate PSC modules leads to a decrease in PCE of all device structures (*9, 10, 38-47*). Screen printing (*16*), slot-die coating (*43*), spray coating (*42*), and soft-cover coating (*48*) have been developed to fabricate intermediate-sized PSC modules on the scale of tens of square centimeters. Alternative processing strategies must be explored to achieve large-area uniform films and efficient materials utilization.

Ohmic loss due to resistance from the front transparent electrode must also be reduced. The conductivity of FTO films can, in principle, be enhanced by increasing the thickness, but this will decrease its transparency. Depositing conductive grids and/or constructing interconnected unit cells can be used to reduce resistance in large-area modules at some sacrifice to the active area of the modules (*49*). Geometrical fill factor (GFF, the ratio between the active area and the total area of a module) can be further enhanced by enlarging the module area, since the ratio of the dead area associated with edges for encapsulation and frames can be reduced.

Stability of perovskite solar cells

One challenge in comparing stability among PSCs resides in the use of a range of non-standard testing protocols within the PSC community (*50*). For shelf-life stability tests in the dark, PSCs are encapsulated or unencapsulated and aspects of the ambient environment vary (e.g. atmosphere, humidity, and temperature) (*51*). Indoor light stability tests use different light sources (*52*) such as an AM 1.5 xenon lamp with or without an ultraviolet (UV) filter, a metal-halide lamp, a sulfur plasma lamp, a halogen lamp, or a UV-free light-emitting diode lamp. Outdoor tests with encapsulated devices (*18*), thermal stability tests (*53*), thermal cycling tests (*54*), and others (*55*) have also been reported. The lack of transparency in reporting stability tests needs to be improved in perovskite PV community. Most reports present normalized results without giving their initial photovoltaic parameters, hiding the fact that the devices being tested for stability are often much less efficient than the devices showing champion or even average performance as discussed in their reports. It is crucial and necessary for the community to present the original photovoltaic data and normalization parameters in the future. In summary, we urge the community to develop and then follow standardized protocols to enable accelerated tests and to predict lifetime of PSCs based on degradation kinetics specific to the cell components (*50*). The well-established standards (e.g. International Electrotechnical Commission (IEC) 61215 or 61646) in mature crystalline silicon and inorganic thin-film photovoltaics can be transferred to the PSC community as well as developing additional protocols to test the reliability of PSCs.

We firstly summarize dominant causes of instability in PSCs on device level. One source of instability is the chemically active charge-transporting material and interfacial degradation. The TiO_2 based ETL, which is widely used in high-performing cells, causes photocatalytic degradation under UV-illumination. Replacing TiO_2 with other UV-stable materials (e.g. $BaSnO_3$, **Fig. 2A**) (*56*) or inserting an interfacial modifier (e.g. CsBr, Sb_2S_3, Cl atoms, **Fig. 2B** and **C**) (*10, 57*) between the ETL and the perovskite absorber can improve stability. Commonly used organic HTMs such as Spiro-OMeTAD, poly(3-hexylthiophene) (P3HT), and poly(triarlyamine) (PTAA) encounter thermal or moisture instability in their doped forms. Substituting the organic HTM with inorganic materials (**Fig. 2D** and **E**), inserting buffer layers between perovskite and HTM, and deploying suitable encapsulation can improve stability (*51, 58*). Metal electrodes such as Au, Ag, and Al have stability issues as well. Ag and Al electrodes suffer from corrosion by ion migration in perovskite, resulting in an apparent color change and PCE decay (*59, 60*). Even relatively inert Au diffuses across the organic HTM layer at temperatures exceeding 70°C, leading to an irreversible performance loss (*61*). Interfacial layers such as Cr, MoO_x and reduced graphene oxide (rGO) have been introduced to prevent metal diffusion and thus improve long-term stability (*51, 57*). Replacing the metal electrodes with indium tin oxide (ITO) can also improve stability both by providing an unreactive electrode, and by creating a barrier layer that keeps moisture out and retains volatile components of perovskite (*19, 54*).

Triple mesoscopic PSCs based on TiO_2/ZrO_2/Carbon/mixed-cation perovskite have passed relatively harsh lifetime testing conditions. With initial efficiencies of 10 to 12%, the PSCs and modules exhibited negligible performance loss after illumination under an unfiltered Xe lamp for 1,000 hours (**Fig. 2, F and G**). The devices also exhibited acceptable stability in thermal stability tests at 85° to 100°C for 2,160 hours and in outdoor stability tests in Jeddah (Saudi Arabia) for 168 hours and Wuhan (China) for 720 hours (*18, 62, 63*). Improved stability in triple mesoscopic PSCs was enabled by removal of unstable components (such as organic HTM and metal electrodes) (*64*) and incorporation of bulky organic ligand (5-ammoniumvaleric acid, 5-AVA) to form low dimensional perovskites ($(5\text{-AVA})_x(MA)_{1-x}PbI_3$, where MA is methylammonium (*17*).

In low dimensional perovskites, large organic ligands, due to their hydrophobic nature, act as a protective window from moisture (*65*) and enhance the moisture stability **(Fig. 3A** and **B)**. (*66*) (*67*) (*68*). However, the large ligands impede charge transfer transport and sacrifice efficiency (**Fig. 3C**) (*37, 69, 70*). Alternatives such as shorter chain cations (*71*) and conjugated cations (*37*) can potentially increase efficiency without sacrifice of the stability.

Protecting interfaces and grain boundaries - sites where degradation may begin - can slow decomposition. Small molecule additives such as thiophene, pyridine (*72*), urea (*73*), phenyl-C_{61}-butyric acid methyl ester (PCBM) (*74*) and quaternary ammonium halide (*75*) have demonstrated improved stability. The hydrophilic side interacts strongly with perovskites and the hydrophobic side faces outward to protect the surface of perovskites from moisture. In addition, formamidinium (FA^+) or Cs^+ were used to replace MA^+ cation (*76, 77*) and Cl^-, Br^- or SCN^- to replace the I^- anion (*78*), yielding various perovskites with a series of tolerance factors (*79*): this tunes the Pb-I bond and the perovskite crystal systems influencing the formation energy and changes the inherent stability of perovskites.

Upscaling and future applications

Recently, several PV companies have set out to manufacture large-area PSC modules. Microquanta Semiconductor (China) obtained a PCE of 17.4% for a rigid perovskite mini-module with an active area of 17.8 cm^2 (**Fig. 4A**). Solliance (Netherlands) developed 169-cm^2-sized perovskite modules with a GFF of 90 % using laser ablation to define the cells that exhibited a total efficiency of 10% and an active area efficiency of 11.1% (*43*). The processing steps were carried out at temperatures below 120°C and are compatible with flexible substrates and roll-to-roll process (**Fig. 4B**). Saule Technologies (Poland) used inkjet-printing to fabricate perovskite solar modules on flexible substrates, which allows the design of free-form patterns (**Fig. 4C**). Greatcell Solar (formerly Dyesol, Australia) is providing chemicals for PSCs fabrication, and it has achieved 12% conversion efficiency for a 10 × 10 cm^2 module and is committed to full-scale production in 2019 (*80*). *p*-OLED (China), and Solaronix (Switzerland) are also engaging in the upscaling of PSCs by providing chemicals and equipment around the world.

Oxford PV and Tandem PV (formerly Iris PV) are attempting to use perovskites to upgrade the performance of silicon solar cells, and fabricate high-efficiency perovskite-Si tandem cells. Recently, Oxford PV has announced a record certified PCE of 27.3%, for 1 cm^2 perovskite-Si tandem solar cells (*81*). HUST in China initiated the upscaling of printable triple mesoscopic PSCs and has achieved efficiency > 10% with an area of 100 cm^2 (**Fig. 4D**). A 7.0 m^2 solar panel was assembled with these cells in early 2015 (*18*). Recently, a 110 m^2 perovskite PV system with screen-printed triple mesoscopic PSC modules (3600 cm^2 for each) was launched by WonderSolar Co. Ltd (**Fig. 4E**) (*82*).

Various device architectures and manufacturing processes are under research and development. It is yet unclear which architecture or fabrication method will become a dominant. At large scales, it is still challenging to control morphology and obtain uniform perovskite thin films using either solution- or vacuum-based deposition techniques (*83-85*). The crystal growth of the perovskite absorber in solvents varies as a result of different evaporation rates at different locations. When the concentration of the precursor solution deposited on a large-area substrate varies, ion migration may occur and may lead to inhomogeneous crystallization. For PSCs in typical mesoscopic or planar structures, the thickness of the perovskite layer is on the order of several hundred nanometers. Pinholes in perovskite films lead to direct contact between the ETL and HTL and cause current leakage and voltage loss for the modules. Although slot die coating has proved to be an effective method to deposit perovskite thin films on large scales, further optimizations on the compositions of the precursors and crystallization process are required.

The toxicity of the solvents used to prepare perovskite films and other charge transport layers merits further study and adaptation: solvent-free deposition methods (*39, 84*) and solution-based deposition approaches that use reagents with lower toxicity (*86*) warrant further development. sah

In addition to the concerns regarding stability and mass production, a thorough life-cycle assessment including materials, production processes and environmental impact should be performed. The lead contained in PSCs has given rise to toxicity concerns. The amount of lead contained in PSCs is below that produced annually by the coal industry when it generates an equivalent amount of electricity (*87*) and is roughly equivalent to that already contained in a 1-cm-thick layer of natural soil (~0.3-1.2 g m^{-2}) having the same area (*88-90*). Nevertheless, lead in PSCs is more soluble in water, and thus can move more readily through the biogeochemical cycle. Therefore, replacing Pb with other non-toxic elements to develop Pb-free PSCs will generate interest if comparable performance can be achieved. It may also be optional to develop additives that immediately convert water-soluble lead compounds into insoluble complexes when destruction of a large perovskite solar power plant occurs.

Life cycle assessment of perovskite/tandem solar cells has also raised concerns regarding the environmental impact of gold and Spiro-OMeTAD (because of use of solvents) (*91*). Developing HTL-free and noble-metal-free PSC architectures that do not compromise PCE is therefore a topic of growing interest.

When lead is used in PSCs, the recycling of perovskite modules is required. Experience and lessons from the CdTe PV industry offer hints on the treatment and recycling of lead and cadmium in absorbers. Perovskite solar panels also include frames, glasses, synthetic materials (e.g. HTMs), encapsulation materials (e.g. EVA) and others, that require further development of recycling strategies. One proposed procedure is to remove the layers of PSCs by using adhesive tape and via immersion of the substrate in different organic solvents. PbI_2 can then be recycled from the perovskite solar panel following recrystallization (*92*). For specific devices such as the printable triple mesoscopic structure, the device scaffold can be reused by washing away the perovskites and reloading them (*93*). In-situ recycling of PSCs through a methylamine gas post-treatment has also been reported (*38*). For silicon solar cells, recycling efficiency can reach impressive values of 85%, which is important given that up to 78 million tons of waste will be generated by silicon solar panels by 2050 (*94*). More than 95% of the glass and all-external aluminum frames can be reused.

The remaining materials are fired to evaporate the encapsulation plastic and the silicon can be melted again for manufacturing new silicon modules.

Reliability concerns for perovskite solar modules

For commercial PV technologies, international standards of “Crystalline silicon terrestrial photovoltaic (PV) modules – design qualification and type approval” and “Thin-film terrestrial photovoltaic (PV) modules – design qualification and type approval” have been established and released by IEC. Specific endurance tests need to be passed before the modules are put into field applications. These tests can determine the electrical and thermal characteristics of the modules and show that the modules are capable of withstanding prolonged exposure in climates described in the scope. The actual lifetime expectancy of the modules so qualified still depends on the design, installation, and environment conditions under which they operate.

At present, silicon and inorganic thin-film PV modules have demonstrated exceptionally high level of reliability in field applications and are able to provide a warranty of 25 years, for which the solar panels can maintain a power output ≥ 80% of their initials after 25 years. Notably, the lifetimes of such PV modules are not only predicated by accelerated aging tests but also have been verified by statistical results obtained with modules under actual outdoor operating conditions.

For PSCs, the intrinsic instability associated with the perovskite absorber itself and device interfaces is one of critical hurdles for practical applications. The light soaking test for PSC mini-modules (active area ~50 cm^2) has prolonged to 10,000 hours with UV-filtered lamps, for which the total irradiation is equivalent to the integrated solar irradiation for ~10 years in most of Europe. With rigorous device design and proper encapsulation, PSCs (active area ~1cm^2) have passed the dark damp heat test for 1,000 hours described in the IEC design qualification testing protocol 61215 for “Crystalline Silicon Terrestrial Photovoltaic (PV) Modules”. These results are mainly obtained with small-sized lab devices which are far from the scale of standard commercial solar cells. Up to now, there is no report of PSCs or perovskite modules that can successfully pass the IEC design qualification testing protocols for crystalline silicon or thin-film terrestrial PV modules. Thus, in the first step, it is crucial to develop PSCs and modules (with proper encapsulation) which can pass the IEC standards for the reliability tests before we can commercialize this emerging PV technology.

In addition to heat and irradiation, mechanical shock may also influence the stability of PSCs. Perovskites have a low fracture energy and their thermal expansion coefficient is more than 10 times higher than that of glass and the TCOs, and this may lead to delamination during temperature cycling. Cells have passed the industry standard test involving 200 cycles between -40°C and 85°C when encapsulants with a moderate Young's modulus, such as ethylene vinyl acetate (EVA), were used to help relieve strain in the solar cells. It remains important to ensure that full-sized panels, in which the perovskite layers have been scribed, remain durable during temperature cycling (*54*).

In practical applications, cells are exposed to high voltage stress compared to ground since they are connected both in series and in parallel. Therefore, PSCs should be assessed by potential-induced degradation (PID) testing protocols and this is not as trivial as other commercial PV technologies given that inorganic thin-film PV technologies are more prone to PID than c-Si (*95*). For silicon solar cells, PID has been regarded as a reliability concern with varied failure mechanisms, including irreversible ones such as electrochemical corrosion and sodium ion damage and reversible ones due to the accumulation of static charge on the surface of cells. For PSCs, ion migration and charge accumulation are present in cells and lead to the anomalous photocurrent hysteresis effect. The presence of ion migration may lead to severe PID and is harmful for the long-term reliability of modules. It is thus crucial to develop method to suppress the ion migration in PSCs. In the future studies, we shall seriously consider PID testing for PSCs to understand the PID mechanisms to ensure long-term module reliability.

Partial shade stress tests remain another topic requiring in-depth studies for perovskite modules: in PV systems, shaded cells will be under reverse bias due to operation of other cells. IEC 61215 describes partial shade stress test to quantify permanent performance loss (*96, 97*). Noticeably, PSCs has much lower breakdown reverse-bias than crystalline silicon and inorganic thin-film solar cells. A reverse-bias breakdown occurs in PSCs between -1 and -4 V (*98*). In addition, local heating resulting from partial shade may lead to damage or even deposition of perovskites since organic-inorganic hybrid perovskites are usually not temperature-tolerant. It is important for the perovskite community to carry out further study of partial shade stress and increase understanding of behavior under reverse bias, and to find means to improve durability under these conditions, such as interconnecting bypass diodes within the monolithic modules.

Perspective

PSCs can be produced from low-cost materials using a simple fabrication processes. They have achieved major advances in efficiency, obtaining high efficiencies in lab cells, and will continue to advance in maturity as stability is enhanced. In general, PSCs feature both low materials cost and short energy payback time (< 2 months) (*95, 99*).

At present, the major challenge for PSCs is to achieve long-term stability on module level and to ensure durable outdoor operation. It is necessary for the perovskite PV community to develop a consensus on the stability measurement protocols and release standardized testing protocols within the community. New protocols should be designed to accelerate the aging process under operation conditions and effectively predict the lifetime of PSCs. Although the degradation of the perovskite absorber caused by moisture or oxygen in ambient air can be inhibited by appropriate encapsulations, the performance decay induced by illumination and heat should be addressed through the construction of stable materials and interfaces. Compositional engineering should aim at maximizing formation energies, and will likely be benefited from emerging machine learning algorithms and robotized synthesis.

A loss in efficiency is seen when PSC area increases (*83*). With continued efforts from the research and industrial communities on scaling up of PSCs, we expect that the efficiency gap between lab cells and industrial modules will be minimized and achieve a level comparable to other PV technologies. Cost per kilowatt-hour is dominated by the efficiency and lifetime of solar panels. The abundant low-cost raw materials and simple processing for PSCs indicate pathways to lower production cost than conventional PV technologies. In the long run, the lower production cost can provide a more sustainable reduction in total module cost, contributing to making PSCs competitive.

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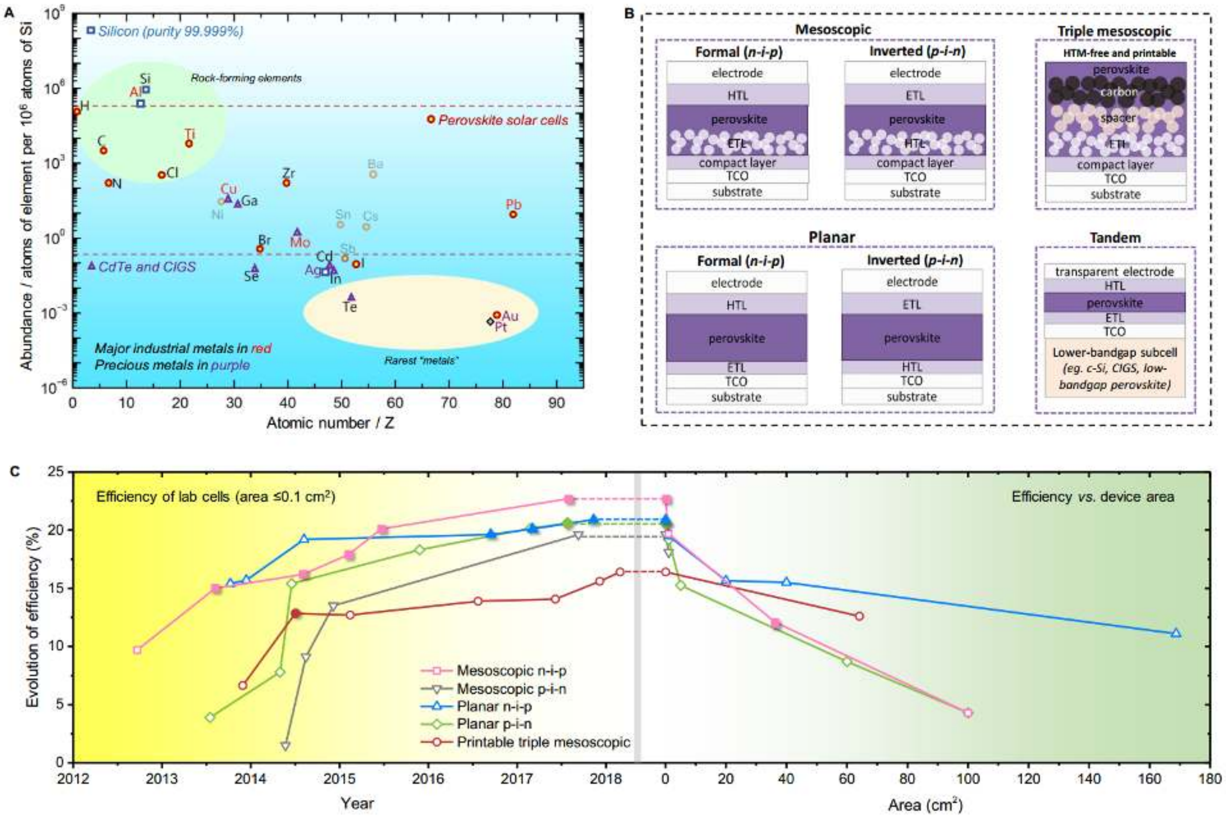

Fig. 1. Chemical elements used in different solar cells, device configurations and efficiencies of PSCs. **(A)** Atom fraction of elements used for different types of solar cells. The major elements for silicon solar cells are labeled in blue squares, perovskite solar cells (PSCs) in red circles, and CdTe and CIGS thin-film solar cells in purple triangles. Ni, Sn, Cs, Ba and Sb elements are labeled in semitransparent color, since they have been used in PSCs but not in the mainstream architectures. For the signs of the elements, major industrial metals are in red and precious metal are in purple, according to Goldschmidt classification (*8*). **(B)** Four device configurations of PSCs: mesoscopic structure; planar structure; triple mesoscopic structure; tandem structure with lower-bandgap subcell. **(C)** Evolution of the best reported lab-cell (≤ 0.1 cm²) efficiencies and the large-area (≥ 1.0 cm²) device efficiencies.

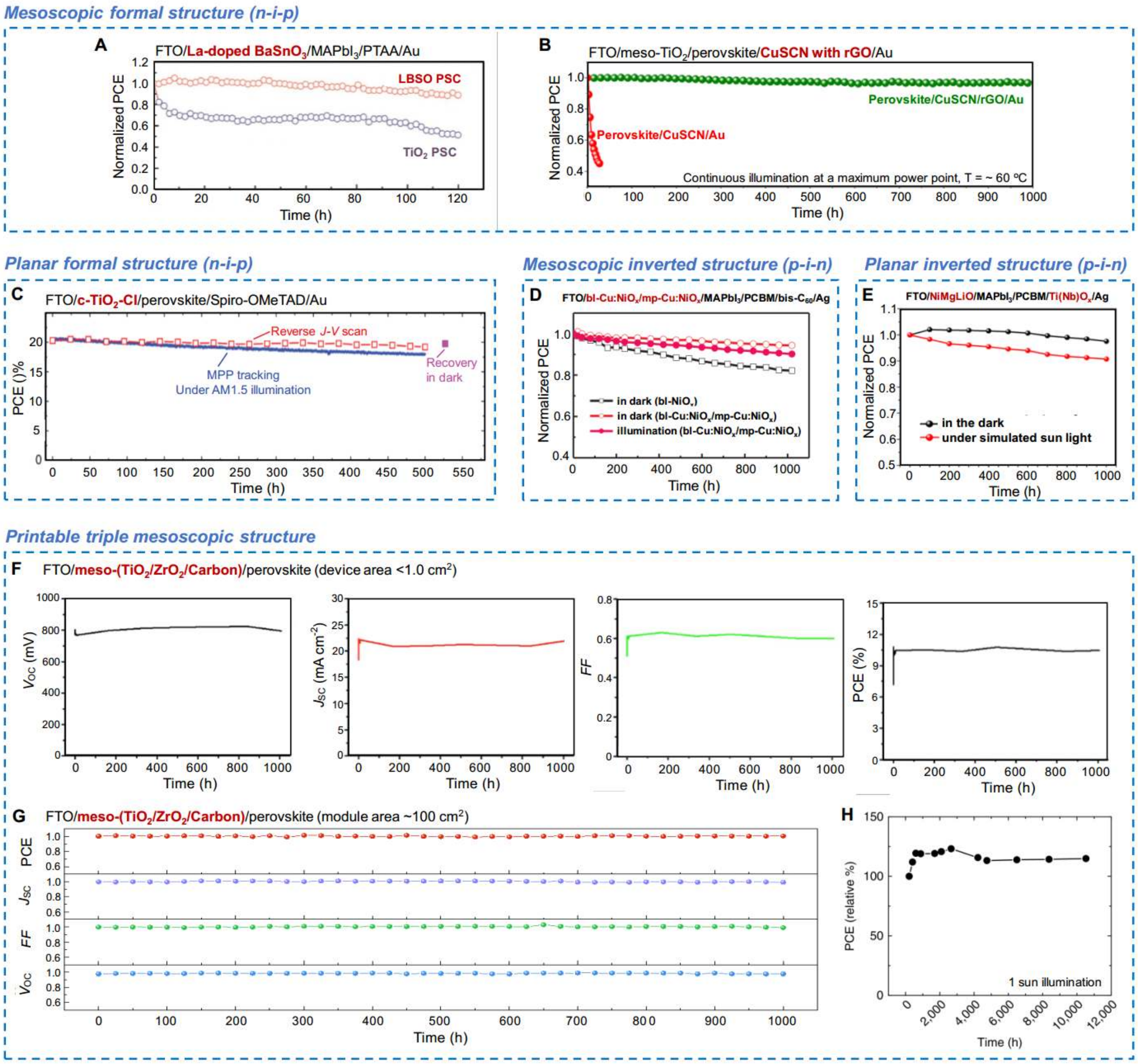

Fig. 2. Summary of operational stability for PSCs with various device architectures. Mesoscopic formal structures (n-i-p) with organic **(A)** and inorganic **(B)** HTLs [Reproduced from (*56*) and (*57*) with permission]; **(C)** planar formal structure (n-i-p) [Reproduced from (*10*) with permission]; **(D)** mesoscopic inverted structure (p-i-n) [Reproduced from (*9*) with permission]; **(E)** planar inverted structure (p-i-n) [Reproduced from (*32*) with permission]; **(F-H)** printable triple mesoscopic structure. Relatively stable devices have been reported for all structures by using appropriate charge-transporting materials, perovskite and electrodes [Reproduced from (*17, 18*) with permission].

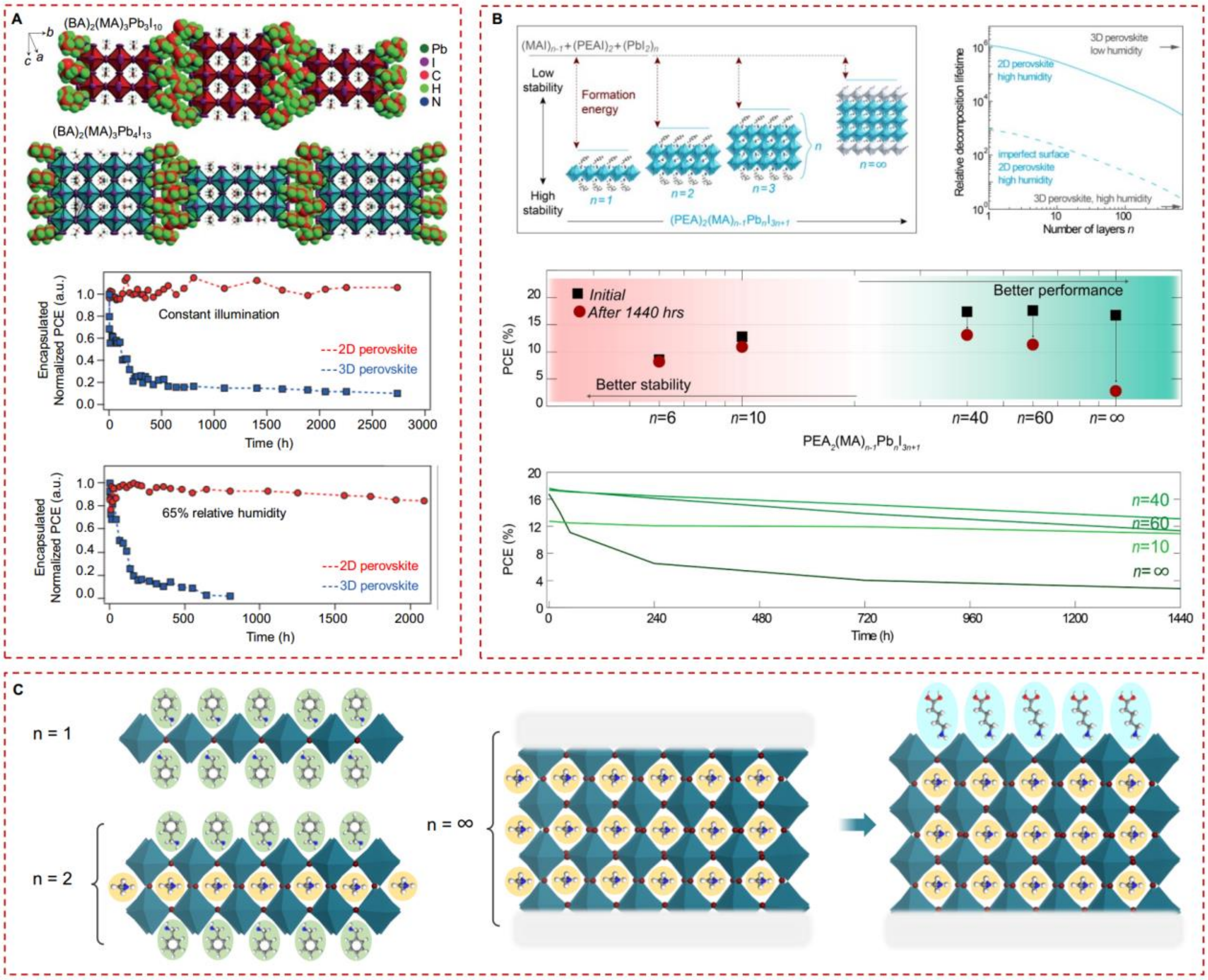

Fig. 3. Strategies to improve the stability of perovskite absorbers. **(A)** Encapsulated PSCs based on two-dimensional Ruddlesden-Popper perovskite exhibit much reduced degradation under constant AM 1.5G illumination and high relative humidity [Reproduced from (*68*) with permission]; **(B)** Improved stability of PSCs by tuning the dimensionality of perovskite absorber [Reproduced from (*69*) with permission]; **(C)** Taking advantage of the hydrophobicity of large cations, the stability of perovskite can be effectively improved by tuning dimensionality. Protecting the interfaces/grain boundaries of perovskites is another way to slow down the degradation.

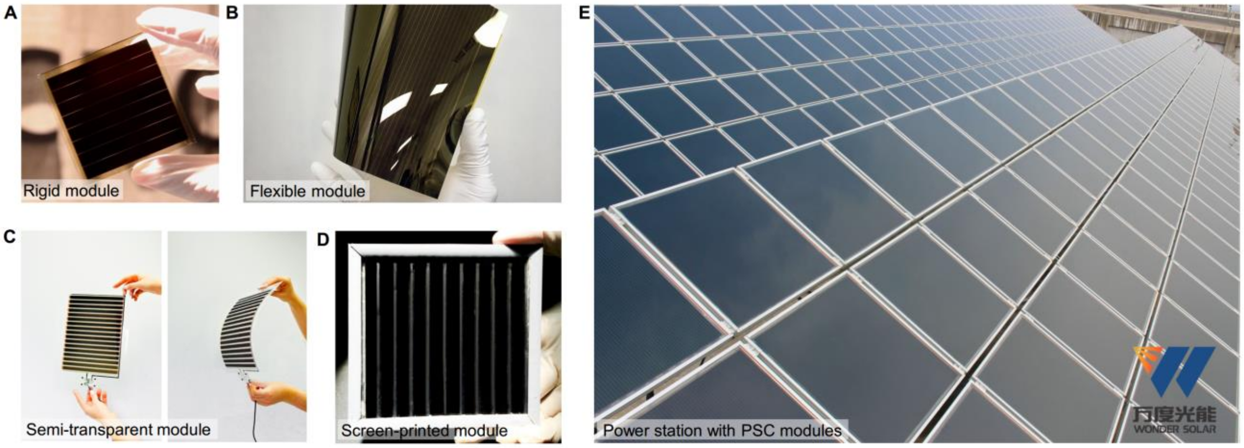

Fig. 4. **Perovskite solar modules.** (**A**) Rigid perovskite mini-module, courtesy of Microquanta Semiconductor; **(B)** R2R processed flexible module, courtesy of Solliance Solar Research; **(C)** Semi-transparent module fabricated with inkjet printing technique, courtesy of Saule Technologies; **(D)** Screen-printed module developed by WNLO at HUST. Figure reproduced from (*18*) with permission; **(E)** Power system with Printable triple mesoscopic PSC modules, courtesy of Wonder Solar.