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Perspective

The path toward metal-halide perovskite industrialization

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SUMMARY

The advent of metal-halide perovskite solar cells has revolutionized the field of photovoltaics. The high power conversion efficiencies exceeding 26% at laboratory scale—mild temperature processing, possibility of fabrication on multiple substrates, and the easy composition-dependent band-gap tunability make perovskites suitable for both single-junction and tandem-multijunction solar cells. They are attractive in various applications, from utility-scale to building-integrated and indoor photovoltaics. To bring perovskite solar cells to maturity, the next big step is to funnel the efforts into developing reproducible and high-quality materials via high-throughput manufacturing, addressing the issues of the inherent material instability. Another key issue is the environmental sustainability associated with the presence of lead. In this perspective, a special focus is placed on real-world applications of perovskite photovoltaics, starting with material selection and deposition techniques and finally discussing the issue of stability and how to improve it toward wider industrialization.

INTRODUCTION

The development of metal-halide perovskite materials for various optoelectronic devices is currently one of the most exciting areas of research.[1–5] In terms of photovoltaic applications, the high power conversion efficiencies (PCEs) achieved in laboratory-scale devices have recently overcome 26% in single-junction devices and 33% in a tandem configuration with silicon solar cells.[2,6,7] This rapid and ground-breaking increase in performance combined with the mild processing conditions is attracting the interest of industrial research with the aim of promoting a rapid real-world application of perovskite solar cells (PSCs). Recently, Oxford PV announced the commercialization of the first 72-cell panels for utility-scale installation, using perovskite-on-silicon solar cells that can produce 20% more energy than conventional silicon panels.[8] However, in order to become be truly competitive with existing photovoltaic technologies, PSCs need to overcome some major issues, mainly related to the intrinsic material properties.[9] As very high efficiencies have already been demonstrated,[2,10–13] scientific and industrial research efforts should now focus on improving stability, developing a reliable and scalable low-cost deposition method, and mitigating the environmental impact related with the use of lead and toxic solvents.[14]

The main component of the most efficient perovskite materials for PSCs is lead, which is an extremely toxic metal. For this reason, lead has historically been banned for all applications, although it has been tolerated in some very specific cases where there is no viable alternative.[15] For this reason, several perovskite materials based on alternative metals to lead are currently being explored, such as group IVA metal

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based-perovskites and double perovskites including metals of group VA.[16] Among the different alternatives, tin (Sn) has shown to be the most efficient, and PSCs based on Sn have recently reached a certified power conversion efficiency of 15.7%[17] but suffer from a severe stability issue compared to lead. Concerning the other alternatives, although they have not yet achieved high performance, these materials are promising for portable technology and indoor applications.[18]

Besides materials toxicity, reliable and reproducible fabrication conditions should be found to produce PSCs at large-scale volume.[19,20] The intrinsically disordered self-assembly process of perovskite materials from precursors makes it very difficult to control the deposition of perovskite on any substrate for device integration in a simple, reliable, and straightforward manner.[21] The irreproducibility of PSC performance between different laboratories and under different conditions should be resolved, ideally by finding the most appropriate and highly standardized processing techniques that can also enable large-scale deposition. Disparate deposition methods are reported in the literature, from solution-based to vacuum-based approaches, all of them having pros and cons.[22–24] Some of these are scalable, but the reported record performances have been achieved using lab-scale techniques on millimeter-scale devices, and the highest certified PSC performance on small module-scale devices (1,027 cm^2) is 19.2%,[25] still well below this record.

Inherent material instability is perhaps the most pervasive issue affecting real-world applications of PSCs. Stability in excess of 20 years is required for truly commercial deployment as a rooftop or utility-scale technology.[26] The extreme intrinsic sensitivity of perovskite materials to high temperature and environmental conditions should be drastically improved. Several strategies are under investigation to find a sound solution to this problem, from different kinds of additives to interlayers[27–34] and different device architecture and electrodes.[35–40] These strategies should be combined with proper encapsulation, enabling the mitigation of both intrinsic material and device interface degradation, as well as external factors such as humidity.[41]

Here, we will focus on these three main issues related to the fate of this technology and then provide a materials science perspective aimed at tracing some guidelines for enabling large-scale production of PSCs. We will first discuss the lead content and explore the availability of potential alternatives to lead. Next, we will address the challenges related to large-scale manufacturing and the stability of metal-halide perovskite materials, which represent crucial prerequisites for achieving a solar cell technology that can truly compete in the market.

THE LEAD CONTENT ISSUE AND VIABLE ALTERNATIVES TO LEAD

The most efficient perovskite materials in solar cells are based on lead, which is extremely toxic.[42–45] Despite the low lead metal content in PSCs, $\sim$0.4 g of lead per square meter, compared to other lead-containing commercial products, such as batteries, the organic and inorganic salts composing lead-based perovskite and processing solvent are miscible with water, making them readily bioavailable. Potential lead leaching from PSCs can enter the food chain through soil and plants 10 times more effectively than other lead contaminants. Although strategies to capture the lead leaching from PSCs have been developed,[46–48] the substitution of lead with other less toxic alternatives is desirable to promote the commercialization of PSCs (Figure 1). However, this aspect has emerged as a challenging goal, as the use of alternative materials to lead-based perovskites has been shown to significantly compromise the power conversion efficiencies.[49]

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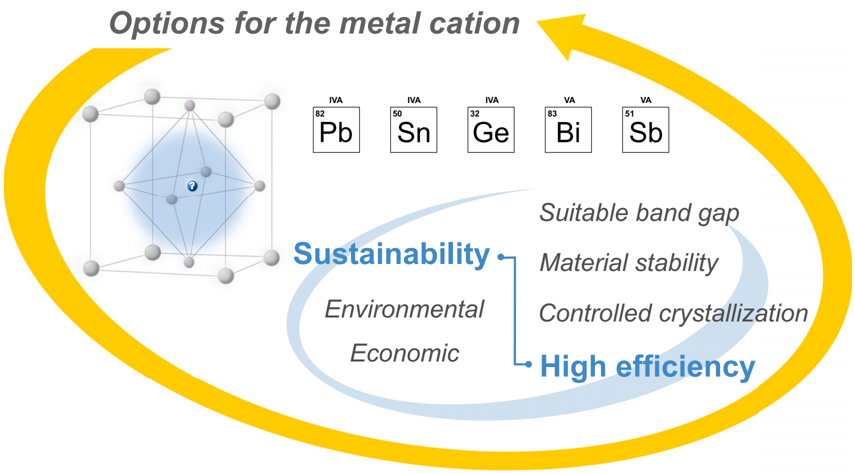

Figure 1. Metal alternatives to Pb for metal-halide perovskite materials
Shown are key metal alternatives to Pb and the primary factors that make them viable substitutes for Pb, fulfilling the requirements of high efficiency and sustainability.

The alternative materials to lead perovskites can be divided into two classes: group IVA metal based-perovskites with the formula $AM(II)X_3$ (A = organic or inorganic cations, M = metal cations, X = halogen anions) and double perovskites, including metals of group VA with the formula $A_2M(I)M'(III)X_6$. In the latter, a combination of monovalent and trivalent metals, such as Ag(I)/Bi(III) and In(I)/Bi(III), can replace lead in the perovskite structure. Although quite stable, double halide perovskites are characterized by an indirect band gap and high effective mass, which, combined with the formation of antisite defects that induce the generation of deep traps, hamper the achievement of high-efficiency solar cells.[50] Moreover, some of these trivalent metals, such as Bi and Ag, are critical raw materials, imposing a further obstacle of materials availability on the real applicability of double halide perovskites in photovoltaic (PV) panels on an industrial scale.

Group IVA metals, such as germanium (Ge) and Sn, are the preferred alternatives to lead (Pb) due to their similar valence and electronic configuration as well as their significantly reduced toxicity. These metals are being investigated as potential substitutes for Pb, aiming to preserve the excellent optoelectronic properties associated with the stable ns2 configuration typical for this group of metals.[51] Nevertheless, Ge-based perovskites exhibit a significant drawback; namely, a band gap exceeding 1.6 eV. This results in suboptimal alignment with the solar spectrum and reduced performance.[52]

Conversely, Sn-based perovskites possess a direct and narrow band gap, making them the most promising choice for replacing the Pb-based counterparts. Several computational studies have been conducted to screen thousands of $AM(II)X_3$ perovskites based on their electronic properties. These studies have agreed that only Pb- and Sn-based perovskites can provide PSCs with competitive efficiency.[53,54] Furthermore, among the various alternatives, only Sn can fulfill the material availability requirements and can show suitable optoelectronic properties.[55] Perovskites based on Sn can also offer new opportunities for PSCs free from the issue of ion migration, which severely affects Pb perovskites. Ion migration is responsible for phase segregation and hysteresis behavior, which ultimately lead to fast degradation of device performance, posing a serious challenge for the long-term stability of Pb-based PSCs. In the case of Sn perovskites, the stronger nature of the Sn-halide bonds suppresses the ion migration in both single- and mixed-halide perovskites, thus potentially improving the material stability.[56]

It is worth to underline that, in the case of Sn, the easy oxidation of Sn^{2+} to Sn^{4+} has hindered the achievement of record efficiencies.[57] Sn is indeed more susceptible to oxidation compared to Pb due to its lower ionization energy and lattice instability at passivated surfaces.[58,59] This instability leads to the generation of Sn^{4+} as a consequence of various oxidation pathways in a complex scenario. Exposure to oxygen during materials processing, exacerbated by the presence of water and/or impurities in the precursors, can accelerate the oxidation rate of Sn. Although this challenge can be effectively addressed by working in inert environments and using pure commercial chemicals, these simple measures are not always sufficient to prevent oxidation, as other pathways also play significant roles. For instance, interaction with solvents can significantly contribute to the oxidation process. The widely used dimethyl sulfoxide, highly useful for controlling the morphology of Pb-based perovskite thin films, is involved in redox reactions with Sn^{2+} and subsequent Sn oxidation, even at room temperature and further at higher temperatures.[60] Additionally, interaction with other layers composing the PV devices can also lead to oxidation of the perovskite layer after its formation. This phenomenon occurs in both direct and inverted solar cells. In direct configurations, oxide interlayers, such as TiO_2 or SnO_2 and Li^+ salt from 2,2′,7,7′-Tetrakis-(N,N-di-4-methoxyphenylamino)-9,9′-spirobifluorene (Spiro-OMeTAD), and in inverted devices, Poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS) are responsible for Sn^{2+} oxidation.[49] All of these pathways occur simultaneously and with varying degrees of contribution to the oxidation of Sn^{2+}, resulting in changes in crystal structures, the formation of Sn vacancies, and self-p-doping.[59,61] This process affects photocurrent and induces significant losses in open circuit voltage (Voc) with detrimental effects on the performance and stability of Sn-based PSCs.

In recent years, considerable efforts have been made to address the challenge of Sn oxidation in perovskite materials.[62] However, finding a definitive solution has proven to be complex and not univocal. For example, Abate et al. conducted a study screening various solvents for formamidinium Sn iodide ($FASnI_3$) perovskites in order to identify an optimal solution for controlling perovskite formation and preventing Sn oxidation. They found that a combination of N,N-diethylformamide and 1,3-dimethyl-tetrahydropyrimidin-2(1H)-one[63] or the use of terbutyl pyridine[45] is effective for this purpose. Other studies have successfully used acidic environments to stabilize Sn^{2+} by incorporating formic or acetic acid.[64,65] Additionally, the use of additives, particularly reducing agents, such as Sn fluoride (SnF_2),[66] or hydrazine-based compounds,[67,68] has emerged as a promising strategy.

These examples, while not exhaustive of all the approaches and additives explored for Sn-based perovskites, highlight the significant interest in tackling this issue and the promising potential of Sn-based perovskites. However, despite the promise of these approaches, further advancements are needed to improve the efficiency of Sn-based PSCs, which currently achieve a certified 15.7% efficiency, still considerably lower than that of Pb-based perovskites.[17,69,70]

Another critical issue with Sn-based PSCs is the poor energy alignment between the Sn-based perovskites and the charge transport layers typically used for Pb-based perovskites. This misalignment results in low Voc and poor device efficiency. Several strategies have been employed to improve this energy alignment, such as using alternative oxides in the direct device configuration, like Nb_2O_5,[71] or doping the commonly used phenyl-C61-butyric acid methyl ester (PCBM) layer.[72,73] Notably, modifications to the PCBM layer have successfully increased the Voc to up to 0.94 V. However, significant efforts are still required to develop transport layers specifically designed for Sn-based perovskites.

In addition to the search for an alternative to Pb, preventing Pb leakage and recycling of Pb from exhaust panels should be pursued in order to guarantee sustainable production and use of perovskite materials in PVs. Apart from all of the production, installation and decommissioning phases, which entail the real-world application of perovskite solar panels, the potential for Pb leakage from installed solar panels in the event of accidental breakage represents the most unpredictable and uncontrollable risk associated with this technology. The release of Pb from the panels can readily contaminate the soil and plants, subsequently entering the food chain.[74] This has prompted active research to identify Pb-absorbing materials that can be used in conjunction with the encapsulation, with the objective of efficiently capturing Pb and avoiding the Pb release from PSCs. Different resins and polymers, and also TiO_2 sponges, have been applied in different parts of the PSC layout to the scope,[47,75–77] achieving Pb capture up to 99.8% from broken devices.

Regarding the recycling of Pb-based perovskites, a cation exchange resin has been employed to separate and release Pb from PSC mini-modules, resulting in a recycling efficiency of 99.2%.[78] More recently, a hot aqueous extraction of Pb and recrystallization of PbI_2 upon cooling has been also reported as a straightforward and non-hazardous way to separate Pb from decommissioned solar panels.[79]

It is important to note that the overall amount of Pb present in the devices is relatively low.[44] Consequently, the majority of the potential cost savings associated with the recirculation of exhausted PSCs can be related to the glass and transparent oxide electrode substrates. Nevertheless, achieving near-100% Pb recycling remains a desirable goal, given the environmental sustainability of the technology.

LARGE SCALE AND HIGH-THROUGHPUT MANUFACTURING

Record power conversion efficiencies above 26% are reached on small-area single-junction devices, which are fabricated at lab scale by hardly scalable deposition methods.[2] The first step in bringing a new PV technology to market is to demonstrate the technology's scalability, which means increasing the size of the device without compromising its PCE. Considering that the formation of perovskite films occurs directly on arbitrary substrates via an intrinsically disordered self-assembly process,[21,80] this raises several issues in terms of reproducibility between different laboratories and atmospheres (i.e., glove-box, ambient air, relative humidity, etc.), among different operators, and also for different device architectures,[81,82] amplified by moving from lab-scale to large-area devices. In fact, the last certified PCE of 22.7% achieved for mini-modules (14.61 cm^2)[83] and of 19.2% and 18.2% for small (1,027 cm^2)[19,25] and standard (7,200 cm^2)[26] modules, respectively (as shown in Table 1), show a relevant reduction ($\approx$30%) if compared to more mature technologies ($\approx$10%) such as copper indium gallium (di) selenide (CIGS) or CdTe.[19,84–86]

Control of the perovskite crystallization and assembly onto target substrate during the film deposition is critical to ensure high-throughput production of high-PCE, large-area perovskite devices. The most diffuse "solution-based" and "vapor-based" deposition approaches show different pros and cons and require different equipment, as shown in Figure 2.

Table 1. Record performance reached on large-area deposition for the most scalable techniques. (da: designated area)

Size	Deposition technique	PCE (%)	Area (cm^2)	Company	Year
Mini-module ($10–200\ cm^2$)	–	16	16.29	Microquanta	2017
	–	17.25	19.277	Microquanta	2018
	–	18	19.276	Microquanta	2019
	–	21.4	19.32	Microquanta	2021
	–	21.8	19.35	Microquanta	2022
	–	22.4	26.02 (da)	EPFLSion/NCEPU	2022
	blade coating	15.3	205 (da)	EMSSU & OIST	2022
	–	20.6	215.5 (da)	KRICT and UniTest	2024
	slot-die coating	22.73	14.61	Zhou et al.[83]	2024
	thermal co-evaporation	18.13	21	Li et al.[87]	2020
	thermal evaporation	16.6	51.1	Ritzer et al.[88]	2021
	thermal co-evaporation	19.87	14.4	Li et al.[89]	2022
Submodule ($200–800\ cm^2$)	based on meniscus coating	11.7	703 (da)	Toshiba	2018
	based on meniscus coating	11.6	802 (da)	Toshiba	2018
	based on meniscus coating	15.1	703 (da)	Toshiba	2021
	–	17.25	277 (da)	Microquanta	2018
	–	16.1	802 (da)	Panasonic	2019
	inkjet printing	17.9	802 (da)	Panasonic	2020
	–	19.5	810.1 (da)	Utmolight	2023
Small module ($800–6{,}500\ cm^2$)	–	19.2	1027.1 (da)	SolaEon	2023
Standard module ($6{,}500–14{,}000\ cm^2$)	–	18.2	7200	Utmolight	2023

The solution-based approaches consist of depositing a perovskite precursors' ink on a target substrate, and then film formation occurs, upon solvent evaporation, by self-assembly of the precursors. The thickness, morphology, and optoelectronic quality of the resulting films are strictly related to the ink properties (i.e., precursor concentration, solvent or mixture of solvents, viscosity, etc.). Therefore, in solution-based deposition, the ability to customize the ink composition, even through the use of additives,[30,90,95,96] is a crucial aspect to develop high-quality perovskite materials. The precursor inks can be tailored according to the technique used and adjusted to meet the needs of large-area coating or printing methods. The processing of perovskite materials using a solution-based approach can integrate conventional graphic art techniques with modern printing technologies, making it an appealing choice for cost-effective, high-throughput manufacturing. Indeed, some techniques, such as gravure printing, screen printing, slot-die, etc., can be upscaled in roll-to-roll and sheet-to-sheet lines, which permit the deposition of high volumes of materials at a wide range of speeds, up to about 900 m/min, according to the techniques employed.[19,97] This results in a notable reduction in production costs and allows for the deposition on flexible substrates.[98] Furthermore, certain techniques, such as flexography and screen printing, enable the creation of detailed patterns on the printed layers. In the case of inkjet printing, the pattern to be printed is digitally defined, which is a significant advantage for the customized deposition of materials and reduces solvent waste.[19] Hence, the development of high-quality solution-deposited perovskite materials for real-word applications will depend not only on the optimization of the inks' composition but also on the reduction of solvent waste and sensitivity to environmental conditions such as moisture, oxygen, and the risk of particle contamination. It has been estimated that, if perovskite material can be

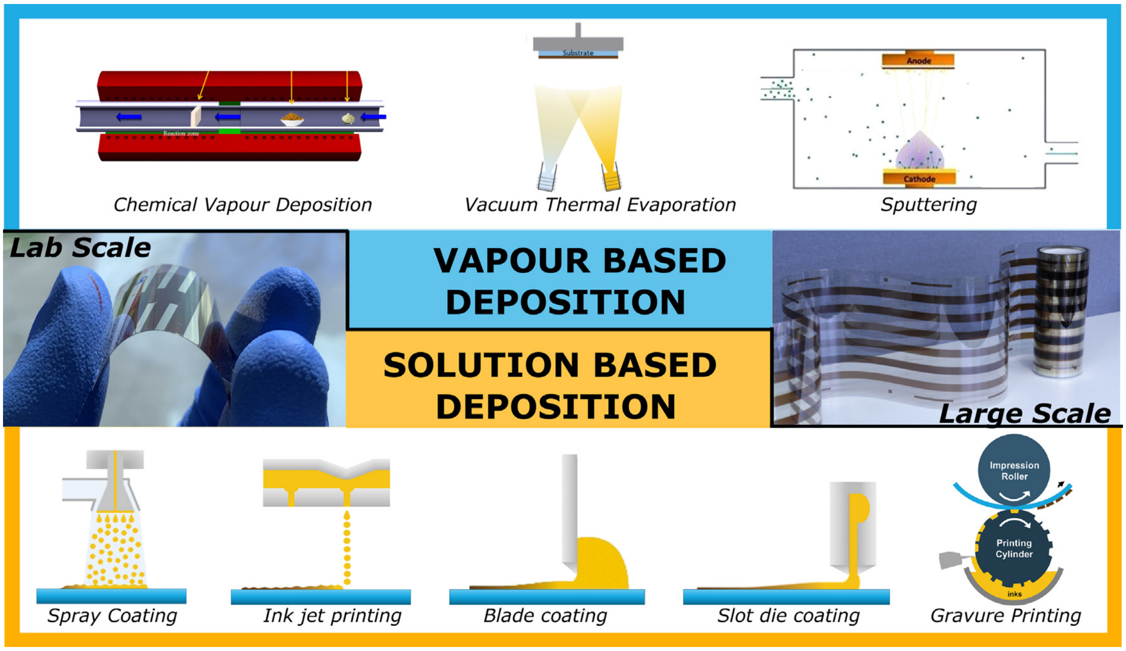

Figure 2. Approaches for scaling up PSC deposition

Shown are various methods for advancing from lab-scale to large-scale production, including VBD and solution-based deposition techniques. VBD methods include CVD, VTE, and sputtering. Solution-based methods include spray coating, inkjet printing, blade coating, slot-die coating, and gravure printing. Figures are adapted with permission from Elsevier,[90] Wiley,[91] and Nature.[92–94]

efficiently produced via these printing techniques, then perovskite modules with a cost of \$0.25–\$0.30 per W might be produced in the near future.[99]

Another option for scaling up device fabrication is represented by vapor-based deposition (VBD) approaches, which can be classified into chemical vapor deposition (CVD) and physical vapor deposition (PVD). The first process, which has undergone various modifications over time (such as hybrid CVD and the evolution to atomic layer deposition [ALD]), involves depositing solids from gases through a chemical reaction on a suitable substrate.[19,100] This method can achieve high-quality films with precise stoichiometry, but this requires high temperatures and may produce hazardous by-products. Instead, PVD involves the deposition of a uniform and high-quality film by condensing materials in the vapor phase onto a substrate in a high vacuum chamber. The most diffuse PVD-based techniques are vacuum thermal evaporation (VTE) and sputtering. Specifically, in VTE, the materials, heated under high-vacuum conditions ($p = <10^{-6}$ Pa)) sublimate and condense on a suitable substrate, which can also be flexible, as the sample holder does not reach high temperatures. Instead, in the sputtering process, atoms or molecules are ejected from a target material by high-energy particle bombardment and condensed as a thin film on a substrate. PVD techniques offer advantages over solution-based processes as they can potentially solve issues such as irreproducibility, solution aging, poor film uniformity, substrate limitations, toxicity, and safety concerns.[21,101–103] Additionally, they allow for good control of layer thickness, the possibility to deposit a pattern using a proper mask, and multilayer perovskite films. However, they have higher manufacturing costs and slower throughput due to the high-vacuum and high-temperature deposition conditions required, together with the slow speed of processing, resulting in high energy and time consumption.[91]

These techniques are particularly suitable to build high-efficiency (~16.6%) small solar modules with an area of ~50 cm^2.[88]

Recently, PCEs of 26.21% (certified) and 24.88% (certified)[7] have been achieved on small-area devices of 0.1 cm^2 and 1 cm^2, respectively, fabricated by sequential vacuum deposition, showing good stability and approaching the record of solution-based PSCs. Moreover, the recorded PCE of 19.87%[89] for a mini-module (14.4 cm^2, aperture area) and 18.13%[104] for a 21 cm^2 PSC demonstrate the potential of this technique toward process scalability.

METAL HALIDE PEROVSKITE MATERIAL STABILITY

In typical PV applications, it is necessary to ensure stable module operation in an outdoor environment for more than 20 years[26] In this context, the biggest challenge in commercializing PSC technology is the inherent material instability of the perovskite active layer. Metal-halide perovskites feature, indeed, a soft and dynamic crystal lattice, which is characterized by mixed ionic and covalent bonding.

Intrinsic material hygroscopicity, ion migration, and thermal instability, combined with extrinsic environmental factors such as moisture, oxygen, illumination, high temperature, and continuous bias,[105–111] limit the long-term stability of perovskite films. As a result, rapid degradation occurs, particularly in the presence of those factors. Perhaps the most detrimental extrinsic environmental factor is exposure to water molecules. The formation of hydrated intermediates, which, following the organic cation-water H-bonding interaction, can weaken the bond between the organic cation and the PbI_6 inorganic framework, promotes degradation.[112,113] This indeed induces a rapid desorption of organic cations and haloid acid, leaving photoinactive metal-halide species or oxides, which adversely affect the device performance. Illumination and heating of the materials, as occurs under standard device operating conditions, further speed up this degradation process.[114,115]

The oxygen-induced degradation occurs via the diffusion of oxygen into perovskite films, which is facilitated by the formation of iodide vacancies under illumination. This process results in the generation of highly reactive photo-induced superoxide species that deprotonate the organic cations, leading to decomposition products, such as methylamine, Pb iodide, and iodine.[112,116,117] In this regard, exploiting antioxidant additives can mitigate the perovskite photooxidation.[118] The presence of water can accelerate the photooxidation process, and the water-accelerated photooxidation mechanism has been found to dominate the degradation kinetics of $MAPbI_3$ in ambient air with >1% of relative humidity at 25°C[119] Importantly, the light-induced degradation can also take place in the absence of moisture and oxygen and can be attributed to the migration of ions.[117] The interstitial halogen defects are the dominating defects in perovskite, which can trap electrons and holes following the photoexcitation. The reaction of two filled traps (I_i^0) can lead to the formation of I_2 molecules. This latter bimolecular reaction is favored by a high trap density and I_i^0 migration occurring upon irradiation of the material.[120]

Thermal degradation is mainly due to volatilization of the organic cation (i.e., methylammonium or formamidinium) in hybrid perovskite compositions. The released gaseous species has been identified as CH_3NH_2/HI and CH_3I/NH_3 for $MAPbI_3$.[121] In addition, due to the polymorphic nature of perovskite, temperature variation can induce phase transitions to different phases, which can also be photo inactive.[122] Concerning this, it is worth noting that, more recently, the effect of water and oxygen has been also shown to induce unwanted phase transformation to the photo-inactive phase in Cs and formamidinium (FA)-based perovskite.[123,124] In this respect, the use of Cs as an A-site cation to develop fully inorganic perovskite materials is considered a method of stabilizing perovskite materials at high temperatures. However, due to the small radius of the Cs^+ cations, the tolerance factor for the $CsPbI_3$ perovskite is 0.81, which is at the lower limit of the range for perovskite formation.[125] The instability of the perovskite phase at room temperature is caused by the rattling of the Cs^+ and octahedron distortion, which has hindered its widespread application. Nevertheless, research on $CsPbI_3$ perovskites is currently ongoing and has yielded promising results in terms of solar cell performance.[126–128]

In parallel to the development of fully inorganic perovskites, different strategies have been proposed to enhance the intrinsic stability of organically based metal-halide perovskite materials. These strategies include surface modification using long-chain cations,[27–29] polymers, or molecules as well as the intrinsic reinforcement of perovskites through the inclusion of different molecules or macromolecules.[30–32,34,129] The presence of hydrophobic species at the interface can protect the whole perovskite film from moisture attack and passivate the uncoordinated surface halide vacancies. The use of molecules that can act as an intrinsic reinforcement can strengthen the grain boundaries, limiting further moisture ingress and passivating the defects.

The use of disparate functional additives is also pivotal to controlling the crystallization of perovskite materials via solution processing. By taking advantage of specific additives that can interact with perovskite precursors already in solution, the crystallization process can be modulated, resulting in larger and/or more cohesive perovskite grains and compact film morphologies that can impart improved stability.[30–32,129–131] A piperidine additive has been exploited recently to assist perovskite growth to achieve a higher surface fraction of the (111) facet in $FAPbI_3$, which is less vulnerable to moisture compared to the (100) facet. The stability test on non-encapsulated PSCs in ambient conditions showed that this approach resulted in 95% of the initial PCE being retained after $\sim$2,000 h.[132] The identification of the most efficient additive/class of additives to universally and drastically improve the stability of metal-halide perovskite materials will be a major milestone toward PSCs evolution.

Besides the inherent stability of the perovskite material, the formation of decomposition byproducts can cause degradation of the hole and electron transport layers and electrodes. For instance, HI and I_2 can interact at the interface with metal oxides or can diffuse through the transporting layers and then interact with the most common metals (Al, Ag, and Au).[133] In particular, Ag and Al can be easily corroded by iodide ions, whereas the inert Au can diffuse into the transporting layers upon device operation, forming metal-halide complexes.[134] Recently, the use of anticorrosive agents has been reported to prevent this issue.[135,136] Considering the variety of hole and electron transporting materials used for the fabrication of PSCs, different reactions can occur at these interfaces.[137] A comprehensive review of possible interactions at those interfaces, leading to deterioration of device performance, can be found elsewhere.[137–139] Another strategy being developed to solve the corrosion of the metal rear contacts concerns the use of carbon-based electrodes. Efficiencies above 19% have been already demonstrated, combined with a stability of 2,500 h at 65°C and under 1 sun, obtained with printable planar carbon-based electrodes.[35]

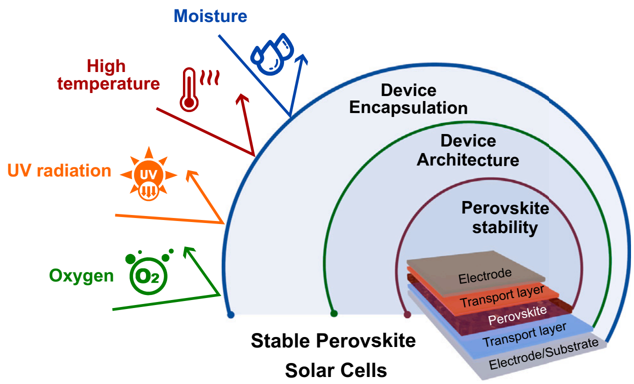

Figure 3. Multilevel approach to enhance PSC stability

Shown is a schematic overview of combined strategies aimed at improving PSC stability, addressing aspects from the intrinsic stability of the perovskite material to device architecture and encapsulation techniques.

Besides the degradation of the metal electrodes, the different thermal expansion characteristics of the perovskite active layers and the substrate/electrode can induce mechanical film stresses, which can result in delamination of the device stack caused by temperature variations.[134,140–142] To alleviate the residual stress, in addition, the processing conditions (i.e., annealing, deposition method, etc.) and the use of functional additives, balancing the nucleation and growth and changing the morphological/structural properties of the perovskite films, can also help.[130]

In terms of device architecture, not only the electrodes but also the choice of transport layers can be a key factor in making PSCs more robust. Inorganic transport layers, such as oxides (NiOx, SnO_2, etc.),[143,144] graphene,[145] transition metal dichalcogenides (MoS_2, WS_2, etc.),[36] Mexenes,[37] and CuO, CuSCN, and CuI,[38–40] have been demonstrated to be a good choice to improve PSC moisture and thermal stability.[146]

With regard to the preferred device configuration, inverted p-i-n PSCs, in which the hole-selective contact is underneath the perovskite active layer, have demonstrated enhanced stability relative to their n-i-p counterparts. This enhanced stability, combined with mild temperature processing and compatibility with integration into tandem devices, makes the p-i-n PSC configuration a highly promising avenue for future real-world applications of this technology.[147–152]

The manipulation of the properties of the active perovskite materials, the implementation of the most stable device architecture, and the use of encapsulation technology to protect PSCs from the ambient atmosphere can be employed in conjunction with one another to enhance stability (Figure 3). Effective encapsulation strategies are indeed essential to guarantee the stability of PSCs in real-world conditions in the presence of moisture and oxygen.[153] Glass-glass encapsulation,[154] polymer encapsulation,[155] and thin-film encapsulation by means of thin oxide interlayers[156] are all strategies pursued to provide protection against moisture-induced degradation of PSCs. A combination of the different encapsulation strategies has been reported as a means to further improve the stability of PSCs.[157] A multi-barrier comprising Al_2O_3 deposited via ALD and parylene thin-film encapsulation completed by a cover glass, along with the use of an inert Bi metal electrode, proved to be a robust approach to improve the light-heat operational stability of PSCs, which retained 90% and 93% of their initial PCE after continuous operation at 45°C for 5,200 h and at 75°C for 1,000 h under 1 sun, respectively.

As a general consideration, measurement conditions are also fundamental to determine the actual PSC stability and to compare it among different laboratories. Standard tests used for other solar cell technologies cannot be easily applied to PSCs, as the latter can be subject to ionic migration under applied voltage and reversible degradation, making a correct determination of stability really difficult. ISOS (International Summit on Organic and Hybrid Solar Cell Stability) testing protocols were established with the aim of providing a more reliable evaluation of PSC stability.[82] In addition, it has been reported recently that light-cycling tests, which mimic the diurnal/night cycle of light that occurs in the real world, are more consistent with outdoor tests than constant-light tests because they allow metastability processes to reset. Nonetheless, these tests, carried out indoors, do not really allow the stability of the PSCs to be assessed; consequently, long-term outdoor stability tests should be coupled to indoor accelerated aging protocols to have more reliable results that can fulfill the PV industry requirements.[158]

Concerning this, accelerated tests using ISOS-L-3 protocols were exploited to extrapolate the stability of PSCs after 5 years of constant illumination at 35°C.[159] For this purpose, fully inorganic $CsPbI_3$ perovskite with a two-dimensional $Cs_2PbI_2Cl_2$ capping layer between the perovskite and the hole transporting layer (HTL) was selected to boost the thermal stability. Extrapolation of the aging time was performed by means of accelerated aging tests taken at different temperatures up to 110°C at a relative humidity of ~65%, that the as-developed solar cells should still perform at 80% of its efficiency after 5 years. An Arrhenius temperature dependence of the efficiency degradation was found over the entire temperature range tested, suggesting that the same degradation mechanism occurred in this range, which the authors attributed to ion migration. The developed $Cs_2PbI_2Cl_2$ capping layer suppressed this detrimental ion migration by passivating iodine vacancies at the surface of the perovskite active layer. More recently, a screening of stability tests based on the ISOS protocols, performed on state-of-the-art p-i-n solar cells based on mixed halide/mixed cation perovskites with PCE above 25%, showed that a better correlation between outdoor and indoor tests can be found when indoor tests are performed under illumination and at high temperature (i.e., 85°C), as they accurately predict the 6-month outdoor stability.[160] Also, in this case, it was found that blocking the ion migration, in particular at the self-assembly monolayer indium tin oxide (ITO) interface, can improve the stability of the p-i-n solar cells.

Comprehensive reviews of the various approaches developed in recent years to increase the stability of PSCs are available elsewhere.[26,161,162] Some of these are listed in the Table 2.

OUTLOOK

This perspective offers a materials science point of view to make perovskite materials for solar cells a robust alternative to more established PV technologies. As PSCs have already achieved impressive record performance in comparison to conventional PV technologies, and the commercialization of the first perovskite-silicon solar panels has been announced recently, at this stage, it is crucial that the community addresses sustainability, scalability, and stability concerns.

Table 2. Some of the most recent multilevel approaches developed to increase the stability of PSCs

Encapsulation strategy	Developed approach	Stability	Aging test protocol	Year
Intrinsic	tailoring the dimensionality (n) of the 2D perovskite at the electron-selective interface of inverted PSCs to enable efficient top-contact passivation through 2D perovskite passivation layers	retained more than 95% of initial PCE after 1,000 h	damp-heat testing (85°C and 85% relative humidity [RH])	2022[27]
	interface optimization through a thermally robust hole-selective layer NiOx/MeO-4PADBC	retained 90% and 74% of initial PCEs (≈25.6%) after 1,200 h at 65°C and 85°C	1-sun illumination at 65°C and 85°C ISOS-L-2I	2023[163]
	use of 1,3-bis(diphenylphosphino)propane Lewis base that passivates, binds, and bridges interfaces and grain boundaries stabilizing inverted PSCs	PCE stable after 3,500 h	Maximum Power Point Tracking (MPPT) under continuous 1-sun illumination in an N_2 environment at 40°C	2023[164]
		retained 90% of initial PCE after 1,500 h	ISOS-D-2	
		PCE stable for 1,500 h	85°C and humidity of ~65%. Open circuit (OC) condition and continuous ~0.9-sun illumination (measured every 208 s)	
	use of a multifunctional fluorinated additive 3-fluoro-4-methoxy-4′,4″-bis((4-vinyl benzyl ether) methyl)) triphenylamine (FTPA) to control and stabilize FA-based perovskite	retained >95% of the initial efficiency (24.1%) for 1,000 h	ISOS-LC-1	2023[165]
	–	retained >95% of the initial efficiency (24.1%) for 2,000 h	ISOS-D-1	
	use of β-poly(1,1-difluoroethylene) to control the triple-cation halide perovskite crystallization and improve the surface hydrophobicity	retained 96% of the initial PCE for 1,000 h	ISOS-L1 continuous MPPT	2023[1]
		retained 93.9% at 80°C and 88.7% at −60°C of its initial value after 120 thermal cycles	unencapsulated devices aged under rapid thermal cycling between −60°C and +80°C, swept at a rate of 20°C per minute	
	use of piperidine as additive to assist $FAPbI_3$ perovskite growth	retained 95% of the initial PCE after ~2,000 h	non-encapsulated PSCs in ambient conditions (40% RH)	2023[132]
	use of cationic cellulose derivative to manage the crystallization engineering of perovskite	retained 91.3% of initial efficiency (24.71%) after 3,000 h	continuous operation in a conventional air atmosphere	2024[166]
Extrinsic	Use of printable planar carbon electrodes as replacement for thermally evaporated metals	PCE stable for 2,500 h	1 sun, 65°C aging test (ISOS-L-2I)	2023[35]
	Encapsulation of FA cesium-based PSCs with bismuth electrode and Al_2O_3/parylene thin-film multiple barrier	Retained 90% of initial efficiency at 45°C for 5,200 h and 93% of initial efficiency at 75°C for 1,000 h	continuous MPPT under 1 sun equivalent white-light light emitting diode (LED) illumination at 45°C and 75°C	2023[157]

In terms of sustainability, the most efficient perovskite materials for PV are based on Pb, which is extremely toxic for human health. However, no viable alternative to Pb-based perovskite appears to be available at present, other than Sn, which suffers from severe oxidation and reproducibility problems. It is also worth underlining that Pb is an abundant material, with a global production on the order of tens of thousands of tons per year, of which almost 60% comes from recycling. Overall, Pb features one of the modest global warming potentials of all metals (1.3 kg CO^2 equiv per kg).[44,167] Recently, several studies have reported a life cycle assessment of Pb -based perovskite solar panels. The environmental impact and energy balance at every stage of the panel life, from raw material, synthesis of precursors, panel fabrication, use, and decommissioning are evaluated and compared with the existing PV technologies.[146,168,169] The minimum sustainable price of solution-processed PSCs was recently estimated to be in the range \$53–70 per m^2 and the levelized cost of energy between 3.6 and 5.9 €/kWh depending on the country.[170] Moreover, PSC panels can be fully recycled, and the active perovskite material can be reused to produce PbI_2 with a recycling efficiency of 99.2%.[78] Thanks to the low proportion of perovskite materials, the most expensive components of PSCs are transparent conductive oxide/glass substrates and encapsulation glasses, and so cost savings related to Pb recycling are negligible.[171] The possibility to reuse the PSC substrates can make this technology appealing from a circular economy perspective.[172–174] This means that Pb-based perovskite can be an affordable material for PVs, both economically and in terms of CO_2 emissions, if specific regulations are put in place for the production, use, and disposal/recycling of perovskite-based solar panels. In view of this, it will be mandatory to control and stem Pb leakage throughout the life cycle of PSCs, from production to installed panels (in the event of accidents) and during panel recycling.[74,175]

Concerning the scalable deposition techniques, thermal evaporation PSCs have recently reached an efficiency of 26% on lab-scale devices, which is very close to the record obtained via solution processing. The possibility of building multilayer perovskite device stacks and solvent/annealing-free processing are advantages related to the thermal evaporation method in view of a possible upscaling of PSC technology. In contrast, solution-based techniques have been the subject of the most extensive research due to their inexpensive integration in laboratory equipment and the possibility of engineering perovskite properties; for instance, by introducing functional additives that can be exploited to render the perovskite material more robust. However, the use of toxic solvents and their disposal, which represent a significant environmental hazard and an economic cost, may hinder the potential use of solution-based approaches in the industrial production of PSCs.[176] Reuse and recycling of toxic solvents have been demonstrated to be an option to avoid the production of solvent wastes.[177] The disadvantages associated with the use of solvents can be partially offset by the compatibility of solution processing with various industrially scalable film coating processes already available (slot die, gravure, etc.) and the ability to print at high throughput (up to thousands of meters per minute). In contrast, vacuum evaporation techniques are characterized by higher capital expenditure (CAPEX) and slower throughput, which will affect the overall manufacturing cost.[178] Both approaches are currently being pursued, and no consensus has yet been reached on the best alternative.

To make this technology truly competitive with silicon and to overcome the 20-year operational limit, the stability of perovskite materials and the lifetime of the devices must be improved dramatically. Therefore, major advances and real breakthroughs should be made to render the intrinsically labile perovskite material more robust.

In this regard, the manipulation of the crystallization process, the utilization of a functional additive or a combination of additives, and the incorporation of passivation layers can be combined to enhance material stability. Nevertheless, efforts should be concentrated not only on the stability of the perovskite active layer but also on the stability of the device architecture and on the selection of the most effective encapsulation technology. Finally, it is necessary to define more specific protocols for the assessment of PSC and module stability. These should include both outdoor and indoor accelerated stability test protocols.

Overall, a comprehensive approach integrating complementary material science, synthetic chemistry, materials characterization, and device engineering is essential to achieve improvements in stability and to address the long-term stability issue for widespread use of metal-halide PSCs.

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DECLARATION OF INTERESTS

The authors declare no competing interests.

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