Beneficial impact of materials with reduced dimensionality on the stability of perovskite-based photovoltaics

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Abstract
Organometallic lead-halide solar cells exhibited immense potential over the past years and reached the transition point from lab to industry-scale fabrication. However, bridging this gap and establishing perovskites as a viable competitor to conventional Si-based photovoltaics, hinges on the success of cost-effective upscaling process. The key factor impeding this transition is operational stability of solar cells under realistic photoconversion conditions. To this extent, reducing the dimensionality of cell constituents appears as a promising and very attractive approach to tackle this issue. The beneficial influence of such materials on device stability, which is explicitly tied to the engineered interface quality with underlying layers, comes as a result of complex interplay between energy alignment, strain-induced interactions and barrier-like properties of 2D components. The aim of this perspective is to briefly outline key challenges regarding the exploitation of 2D materials within the framework of perovskite photovoltaics, as well as to suggest further development directions.

1. Introduction
The development of organometallic lead-halide perovskite solar cells has matured to a point of industry-scale production, however their operational long-term stability still remains the main obstacle impeding direct commercialization of perovskite technology. Notably, numerous approaches were undertaken with various degree of success to assess this issue, ranging from encapsulation, control over photoconversion process and selection of appropriate materials for effective hole and electron extraction [1–5]. Among the plethora of possibilities, engineering materials with reduced dimensionality (RD: 0D, 1D, 2D or mixed hybrid structures) appears to be a very promising approach: their incorporation introduces minimal perturbation into the system and concurrently their intrinsic advantageous properties are positively reflected mainly on the device stability and incidentally, on power conversion efficiency (PCE) [6, 7]. In principle, 2D materials act simultaneously as barriers shielding the cell from moisture ingress/ion migration and energy alignment tool promoting efficient charge transfer. In this perspective, we reflect on the benefits and challenges these materials face from the standpoint of long-term stability pertaining to outdoors installation [8]. A number of literature report already demonstrated extremely good stability and durability characteristics with extrapolated lifetime of 10 years. Examples worth mentioning include: (a) passing relatively harsh lifetime testing conditions while maintaining acceptable performance loss upon illumination under 1 sun for 1.000 h [9, 10], (b) modules passing stability tests (at 85°C–100 °C) for 2160 h including outdoor stability tests for 720 h [11, 12] and (c) devices based on hybrid 2D/3D perovskites exhibited enhanced stability delivering high stabilized efficiency for >10,000 h at 55 °C with negligible performance losses [13]. To that extent, we report the progress regarding the application of 2D materials in respect to four key parameters: absorber composition, interface engineering, photocurrent hysteresis and device encapsulation. Within such framework, we first examine the role of 2D materials as a function of variables concerning quality/properties of perovskite crystals and the ways in which RD can provide benefits related to the structural integrity and light harvesting abilities of perovskite films. Next, we discuss ways to remediate the influence of internal and external factors impeding solar cell stability and the role of 2D
materials in this process, both as interlayers and dopants to conventional charge transporter layers. In addition, we evaluate the impact of low-dimensional constituents on the mechanism behind hysteresis phenomenon and how it affects the operational stability of perovskite systems. Finally, we estimate the potential nanolaminates to offer in encapsulation of perovskite solar cells and outline their major drawbacks. Within this perspective, we aim to provide readers with the main ideas regarding the research progress and future directions for RD materials and the place they have in the community gathered around perovskite photovoltaics.

2. Perovskite composition and crystallinity

The quality (crystallinity, size, orientation, shape/morphology etc) and distribution (i.e. compactness) of grains within perovskite crystal defines the innate stability of the resulting films and limit the overall resilience of the solar cell towards external influences. Therefore, the process of nucleation and crystal growth has been a topic of intense study in an attempt to yield high-quality light harvesting films. The very process of nucleation is kick-started once the concentration of solution reaches saturation point and radius (r) of particles overshoots critical value, as defined by the relation between free energy related to the surface (ΔG_s) and bulk (ΔG_v) of the particle [14]:

$$\Delta G_\text{s} + \Delta G_\text{v} = 4 \cdot \pi \cdot r^2 \cdot \gamma + \frac{4}{3} \cdot \pi \cdot r^3 \cdot \Delta G_\text{v}.$$  (1)

The viability of nucleation pathway is conditioned by the interfacial energy (γ), which is a key factor determining whether homogeneous or heterogeneous direction will be favoured. During the film formation, large particles will maintain continuous growth rate, while smaller will be excluded from the system. This phenomenon is known as Ostwald ripening and is of a particular interest for the growth of perovskites films with large grains [15]. Obviously, reducing dimensionality of lead–halides from 3D to 2D structure is bound to have beneficial influence on the quality of resulting crystals and by extension on their stability. Indeed, substitution of organic cation at the A site of perovskite (APbX_3) with a bulkier cation (M) reduces dimensionality according to the following structural relation M_n(MA)_{1−n}PbX_{3n+1}, where for n = 1 perovskite assumes the actual 2D conformation. Reduced dimensionality of perovskites provides improved resilience towards thermal and photoinduced stress, as well as the moisture ingress. This occurrence is commonly assigned to the highly ordered 2D conformation and presence of large organic cation acting as a hydration barrier [16]. In particular, very strong van der Waals interactions between bulky organic cations ensure that critical formation energy is increased and the material is efficiently shielded from humidity. On the other hand, lead–halides characterized by 2D ordering exhibit inferior PCE in comparison to their 3D counterparts. One of the major influences causing their subpar performance stems from the fact that the capacity for light absorption is a direct function of structural integer n and further divergence from 3D conformation increases the bandgap of resulting 2D perovskite. Moreover, RD is accompanied by an increment of the exciton binding energy and subsequent charge localization, which further hinders effective charge separation and transport [17]. One way to tackle this obstacle, although technically challenging, is via directional growth of 2D perovskites, since perpendicular orientation of crystals ensures efficient transfer of charge carriers towards their respective electrodes. More promising approach to suppress the negative effects following RD, while maintaining the ability to utilize all benefits arising from such structures, is the incorporation of 2D/3D hybrid structures. These are usually achieved via direct incorporation of 2D units into the bulk of perovskite, deposition of 2D interlayer between absorber and electron transporter or by capping grains of 3D lead–halides with their 2D analogues (figures 1(a) and (c)) [18–21]. Such combinatorial flexibility provides remarkable resilience towards photoinduced decomposition, as large cation provided by 2D perovskite prevents unwanted charge accumulation during photoconversion process. Grätzel et al proved that such 2D/3D combination leads to substantial extension of solar cell lifetime at both room and elevated (150 °C) temperatures, while maintaining satisfactory operational stability [22]. An exemplary demonstration of advantages 2D modified perovskites provide was reported by Bai et al, who introduced in situ route towards formation of 2D/3D graded interface incorporated into the device with normal architecture [23]. Resulting highly efficient cells yielded PCE’s exceeding 19%, while demonstrating reduced recombination between perovskite and electron transporter, improved environmental stability and suppressed migration of ionic species caused by externally applied thermal stress. Moreover, inclusion of 2D perovskite layers into device structure brings the research community one step closer towards elimination of toxic footprint caused by the presence of lead. Namely, replacement of Pb by Sn in conventional perovskite absorbers results in a very unstable and inefficient devices, but 2D/3D approach offers a promising path towards alleviation of these problems. Notably, an important step in this direction was taken by Shao et al [24], who reported lead-free devices based on 2D modified FASnI_3 absorber. Solar cells fabricated in this way exhibited twofold improvement of PCE, while maintaining high reproducibility and reduced monomolecular
recombination. It is worth mentioning that discussion regarding 2D/3D perovskite stacks in the context of stability does not limit their application only to small area devices. Grancini et al. demonstrated satisfactory operational stability of such architecture surpassing one-year mark (see figure 1(b)) and confirming that multidimensional absorber concept is transferable to perovskite modules [13, 25]. Even though the benefits
provided by inclusion of 2D perovskites into cell design are undisputed, a number of issues needs to be addressed before reaching the stage where it’s possible to simultaneously maintain high PCE and operational stability of such devices. To this extent, additional efforts should be directed towards establishing standardized protocols to study, characterize and identify alternative functional groups suitable for the growth of 2D phase.

3. 2D materials and engineering of interfaces

The idea of incorporating low-dimensionality materials as interlayers has been garnering attention of perovskite research community over the time (see figure 1(d)). The ability of 2D layers to transfer benefits of their unique optoelectronic properties onto perovskite solar cells without interfering with the mechanism of light harvesting, seems extremely attractive. The most prominent candidate for 2D interlayers is graphene, either in its innate sp2-hybridized monolayer form, or as one of its numerous functionalized derivatives. The very facile integration into lead-halide photovoltaics stems from the favourable optoelectronic properties of graphene, like transmittance, resilience towards in-plane deformations and remarkable drift mobility of charge carriers. Listed parameters are susceptible to further tuning via numerous functionalization protocols, resulting in doped hexagonal lattice characterized by altered chemical potential and work function. Such structural engineering flexibility allows strategic placement of graphene interlayers across various parts of solar cell with particular importance for the stability, as demonstrated by the work of Hu et al [26]. They employed graphene layer to block the leakage of gold ions from the top electrode into perovskite film and prevent oxygen/water ingress from the environment, which successfully extended operational stability of measured device, while maintaining >90% of initially observed PCE. Another way to improve operational stability of lead-halide photovoltaics includes oxo-functionalization of graphene (oxo-G1), which yields high work function sheets and provides the resulting device with larger built-in field and consequently, higher open circuit voltage. Hydrophobic properties of oxo-G1 ensure outstanding protection against humidity-induced degradation and contribute to the extension of long-term stability over 1000 h mark [27]. The incorporation of graphene into inverted planar devices is particularly appealing choice, since it allows replacement of fullerene-based materials, which are almost exclusively used as electron transporters in such architecture. To that extent, Castro et al reported the use of cove-edge graphene nanoribbons to enhance energy alignment, facilitate electron extraction, maintain operational stability and boost PCE of perovskite cells [28]. As the formation of highly-ordered graphene at larger areas becomes a challenging task, one way to overcome this obstacle is the employment of graphene flakes as a dopant to charge transporters, since beneficial properties of graphene are transferred on the host material. For example, doping of TiO2 layer with graphene flakes in perovskite solar cells with normal structure, preserves PCE during extended operational lifetime and remediates adverse photocatalytic behaviour of titanium dioxide induced by UV absorption. Moreover, further reduction of dimensionality to quantum dots (0D) introduces additional degree of freedom to alter the bandgap and fine-tune the energy transfer between perovskite and charge transporter. As a result, graphene quantum dots can be employed to improve conductivity of inorganic oxide layers and promote photocurrent enhancement within perovskite photovoltaics [29, 30]. Although the hydrophobic nature of graphene has beneficial impact on device stability, it also limits solution processability and complicates upscaling processes. To this extent, researchers usually employ graphene oxide (GO), which is more appropriate choice for cost-effective fabrication, due to the presence of hydrophilic groups attached to hexagonal core ensuring easier substrate coverage. In principle, GO is subjected to further chemical reduction to yield reduced oxide derivative (rGO), which is more suitable for light harvesting applications, as it demonstrates improved transparency and conductivity. Graphene oxides remain chemically inert towards the perovskite, while the presence of oxygen increases the work function at the cost of severely reduced conductivity. This trade-off can be minimized during GO reduction process, yielding quality 2D hole transporting layer suitable for implementation into inverted planar devices.

Such rGO interlayers provide numerous benefits ranging from the promotion of perovskite crystal growth and charge collection, all the way to the prolongation of operational lifetime, compared to conventional hole transporters [33]. Another interesting family of 2D materials suitable for charge transport applications are transition metal dichalcogenides assembled by a strong covalent bonding between metal and chalcogen (Se, Te or S) atoms. Notable examples of such materials are Mo and W disulfides [31, 34], well-known for their high charge mobility and adjustable direct bandgap. Although the inorganic nature of 2D dichalcogenides has a positive impact on the stability of perovskite solar cells, the main hindrance preventing mass application of these materials is a lack of standardized, low-cost fabrication protocol able to produce high-quality layers without unwanted vacancies, dislocations and out-of-plane bonds. It should be kept in mind that the low dimensionality of metal dichalcogenides renders them very sensitive to the way in which the interfacial contact with underlaying film is established, which may have negative impact on resulting sheet resistance. Nonetheless, MoS2 is a good example of versatility these 2D chalcogenides offer, since it can govern both electron and hole transport, either as
an interlayer or a dopant. In both cases, significant improvement of device stability and energy alignment with adjacent layers were reported for both inverted and normal perovskite solar cells [35, 36]. Kakavelakis \textit{et al} demonstrated potential of MoS$_2$ to improve operational stability of inverted perovskite solar cells by suppressing light-induced decline of photocurrent [37]. Moreover, the presence of MoS$_2$ interlayer between the absorber and hole transporter improved charge extraction and retarded carrier recombination process. Besides, MoS$_2$ acts as a quality foundation suitable for epitaxial growth of large grain perovskite films and facilitates efficient hole collection, resulting in stable solar cells with remarkable PCE (>20%) [38]. Another very promising addition to the group of prospective 2D materials for perovskite photo voltaics are boron monolayers, also known as borophene. Namely, borophene is the lightest 2D material with metallic properties and it possesses several key advantages over graphene, such as lateral strength, stiffens and superior resistance to mechanical stress due to the phase shift preventing direct fracturing of the monolayer. Unfortunately, borophene displays certain level of sensitivity towards the oxygen, upon exposure to higher concentrations [39]. Even though the oxidation process appears to be limited to its edges, it can certainly interfere with the charge distribution over the layer. Nonetheless, borophene being a highly conductive medium opens possibilities for application in conjunction with boron nitride or other previously mentioned 2D materials, which are able to act as a shield from the negative ambient influences [40]. Additional research efforts should be directed towards design of scalable and standardized solution processing fabrication methods for 2D materials applicable to perovskite photovoltaics, without sacrificing photoconversion efficiency and overcomplicating device design.

4. The effect of reduced dimensionality on the photocurrent hysteresis

Photocurrent hysteresis observed in lead-halide based photovoltaics is one of the most pressing concerns within the perovskite community, since it inevitably interferes with the monitoring of device performance and serves as a clear indicator of unwanted underlying processes occurring during photoconversion and idle cycles. The exact origin of this phenomenon remains a subject open for interpretation to a certain degree and clear consensus on this matter is yet to be reached. Nonetheless, it has been proven that hysteresis is heavily influenced by the migration of mobile species, distortion of balance during the collection of charge carriers, arrangement of ferroelectric domains and monomolecular recombination process, which further exemplifies why exact identification of hysteresis roots is so convoluted [41–44]. The key step leading to elucidation of its nature is reliable mapping of charge trapping population ion migration and, which are the main catalysts for generation of recombination centres. It is of the utmost importance to regulate the ratio between mobileionic species and traps, since device poling during operation introduces additional degree of freedom for ion migration, which in turn promotes charge recombination at the existing trapping sites [45]. Furthermore, device ageing introduces additional variables into hysteretic system, causing fluctuation of activation energy related to the movement of ions and altering distribution of halide ions within the inorganic cage of perovskite crystals. Successive polarization and depolarization of organic cations as a result of interplay with ferroelectric domains, leads to the carrier accumulation at the surface, emulating interactions between two codependent capacitors [46]. In such cases, reducing dimensionality of materials either to independent 2D layers or 2D/3D perovskite stack conformation, imposes itself as a promising approach towards tackling the aforementioned issues. Besides successfully suppressing redistribution of ionic species and reducing defect population, 2D structures also serve to improve device resilience towards applied thermal stress [47]. Since the inception point of perovskite decomposition induced by elevated temperature is located at the grain boundaries where trap population is high, dimensionality engineering serves as viable tool to circumvent such hindrances and further promote operational stability of perovskite solar cells [16]. Additionally, structural factors like morphology and crystallinity of the light harvester need to be equated into the assessment of hysteresis impact on the stability and performance of lead-halide photovoltaics. In principle, pinhole-free perovskites exhibiting highly uniform substrate coverage indirectly alleviate the negative influence of hysteresis by reducing the number of grain boundaries and overall non-radiative recombination. Since absorption process is intertwined with the charge carrier collection at respective electrodes, various approaches outside of perovskite modification can be undertaken to further confine hysteresis within the acceptable limits. Viable strategies commonly include doping of hole/electron transporters, fine tuning of the interfaces to facilitate easier charge injection and passivation of trapping defects [48–51]. These are exemplary cases when RD provides beneficial effect either through insertion of 2D interlayers or modification of the absorber itself, as described earlier in the manuscript. The most commonly utilized approach towards passivation is via inclusion of fullerene transporting layer, which has a twofold effect on the reduction of trap states. Firstly, fullerenes directly reduce the trap density via surface coverage and accumulation at grain boundaries and secondly, they suppress unwanted ion leakage via dominant migration pathways [52]. However, such passivation mechanism fails to prevent appearance of hysteresis at low temperatures due to transition of non-passivated traps from shallow to deep levels, as a consequence of thermal activation energy
requirements [53]. Moreover, fullerenes are UV sensitive materials and lack of proper shielding causes photoinduced polymerization that may interfere with electron extraction, which is why incorporation of fullerenes in perovskite cells with normal architecture limits their usage to dopants for inorganic electron transporters. On the other hand, 2D materials introduce different mechanism of hysteresis reduction including prevention of defect formation through increment of perovskite crystal quality, rather than mitigation of unfavourable defect distribution on already formed crystals [54]. Optionally, 2D interlayers like functionalized GO are proven to serve as passivation mediators via interactions with uncoordinated clusters of lead ions and additionally improve charge extraction, while conveniently boosting open circuit voltage of such devices [55]. Described advantages coupled with a photostable nature, allow 2D materials to circumvent abovementioned limitations and impose themselves as a promising choice in design of perovskite photovoltaics. Finally, we would like to reflect on the lack of proper figure of merit used to quantify the effect of photocurrent hysteresis. The most widespread approach regarding this issue is estimation of ‘hysteresis index’ in an attempt to measure divergence of PCE recorded during forward and reversed current–voltage scans:

\[ HI = \frac{PCE(\text{reverse scan}) - PCE(\text{forward scan})}{PCE(\text{reverse scan})}. \]  

Unfortunately, such approach is based on discretionary selection of transient values and it leads to unreliable conclusions, as correctly pointed out by Habisreutinger et al [56]. Until a suitable figure of merit is proposed, researchers should refrain from using aforementioned index to describe the effect of photocurrent hysteresis on the system, as underestimation of mobile species and trap distribution impact inevitably leads towards erroneous conclusions regarding the stability of perovskite solar cells.

5. The role of 2D materials in encapsulation process

Any discussion regarding long term stability of perovskite photovoltaics would be incomplete without evaluation of device encapsulating options, which provides fundamental protection from the negative environmental effects. Eventual barrier failure would leave solar cell exposed not only to combination of oxygen and moisture, but also to UV stress, which has multiple adverse effects on the structural and operational parameters of devices. Performance efficiency of encapsulating materials is usually quantified via water vapour transmission rate (WVTR), which estimates penetration rate of moisture in g m\(^{-2}\) day\(^{-1}\), as defined by Fick’s law of diffusion [57]:

\[ WVTR = \frac{D \cdot S}{d} \left[ 1 + 2 \sum_{n=1}^{\infty} (-1)^n \cdot \frac{\left( -D \cdot n^2 \cdot \pi \cdot t \right)}{d^2} \right], \]  

where \(D\) stands for diffusivity, \(S\) for surface saturation over time \((t)\) and \(d\) represents the barrier thickness. Commonly used encapsulation materials like UV curable epoxies, ethylene derivatives or glass/adhesive combinations are selected as a conservative and efficient choice to prevent formation of pinholes, which are proven to exhibit the most detrimental effect on the shielding ability of encapsulant. To that extent, thick multilayer barriers offer a way to separately address chief encapsulation concerns (processing temperature, pinhole reduction, UV/IR cut-off, etc) within the unified laminated solution. However, such structures inevitably introduce additional transient factors into consideration, besides previously described steady-state permeation rate. Moisture ingress in these complex systems is best described using thermal activation energy for each of degradation parameters: moisture permeability \((E_{\text{perm}})\), adhesive interconnections \((E_{\text{adh}})\), solubility of barrier materials \((E_{\text{solute}})\) and their correlation with temperature \((T)\), relative humidity \((RH)\) and overall degradation rate \((R_d)\). Mutual relationship between these parameters allows estimation of the ratio between device lifetime under normal \((L_{\text{norm}})\) and increased stress \((L_{\text{acc}})\) conditions within the framework of accelerated stability testing, according to the diffusion-controlled mechanism [58, 59]:

\[ \frac{L_{\text{norm}}}{L_{\text{acc}}} = \exp\left( \frac{E_{\text{adh}} + E_{\text{solute}} + E_{\text{perm}}}{2R_d \cdot \frac{1}{T_{\text{norm}}} - \frac{1}{T_{\text{acc}}}} \right) \left( \frac{RH_{\text{acc}}}{RH_{\text{norm}}} \right). \]  

considering all listed variables, it comes as no surprise that conventional encapsulation solutions introduce a significant trade-off between desirable substrate coverage and the degree of control over \(O_2/\text{H}_2\text{O}\) permeation. On the other hand, RD of protective barriers opens possibilities to increase regulation over structural properties of the material and further simplifies encapsulation process. Namely, very thin metal oxide layers fabricated via atomic layer deposition (ALD) have already been demonstrated as a high-quality barriers compatible with perovskite solar cell architecture [60]. Benefits of ALD grown thin films of metal oxides directly onto lead-halide films reach beyond simple barrier provision and offer additional functionalities, such as charge transporter engineering and simplified integration into tandem solar cells. The most promising results of these
nanolaminates were reported for a combination of aluminium oxide with group-IV transition metal oxides (ZrO$_2$, HfO$_2$ and TiO$_2$) and for oxide/organics hybrids [61–63]. Notably, Brinkman et al demonstrated an ALD growth, electron extraction interlayer consisting of aluminium-doped zinc oxide and tin oxide revealing the formation of remarkably dense gas permeation barrier able to effectively suppress moisture penetration towards the perovskite and leakage of decomposition products from the lead-halide film [64]. Nonetheless, we should keep in mind that ALD reactors operate in the presence of oxygen plasma, elevated temperature and water vapour pulses, which may have detrimental effect on the stability of underlying perovskite films. Thus, the careful optimization of film deposition on case-by-case basis may be necessary in order to ensure that operational stability of encapsulated solar cells is left undisturbed. As an alternative to ALD, Christians et al used a low-temperature, solution-based processing to design a SnO$_2$-based electron transport layer with the insertion of ultrathin MoO$_3$ layer, resulting in very stable devices (without the need for additional encapsulation) able to retain 94% of their peak PCE after 1000 h of operation [65]. On the other hand, solution processed graphene imposes itself as a viable choice for effective and affordable encapsulation relying on its resilient 2D structure. Li et al [32]. demonstrated the use of single-layer graphene as a molecular shield capable of isolating underlying crystals from degradation caused by elevated temperature and solvent penetration as illustrated in figure 1(e). Furthermore, physical deposition of graphene provides a reliable route towards fabrication of coatings with high optical transparency (>90%), making them suitable for sealing of light-emitting diodes. Graphene nanolaminates grown in this way present remarkable barrier against moisture ingress and demonstrate substantial level of flexibility, as they can be used either alone or in combination with semiconductor oxides [66]. Since intrinsic metallic properties of graphene govern the extent of interaction with charged species present within the light harvesting system, the effects like electron screening and exciton quenching often accompany devices where graphene forms interfaces with charge transporting materials. Optionally, other 2D materials with large bandgaps may be employed to overcome any potential hurdles arising from undesirable electron interactions transfer between the barrier and underlying films. Good example of such materials comes from the family of anisotropic boron polymorphs in combination with insulating hexagonal boron nitride, allowing fine-tuning of interfaces until desirable conductivity properties are reached [40]. Another cost-effective lamination approach relies on the rGO films in combination with polyethylene naphthalate to yield highly transparent, flexible encapsulation coatings [67]. Properties of such rGO barriers are conditioned by the stacking quality of 2D sheets and suppression of resulting moisture ingress via nanochannels formed by the unfavourable spatial alignment of rGO building blocks. Literature reports covering the topic of barriers based on 2D materials deliver promising results and showcase advantages of such films over bulkier, more complex conventional solutions. Notwithstanding, further research is still necessary to adequately upscale and standardize fabrication of nanolaminates, as well as to establish the balance between deposition/material costs and benefits provided by such encapsulants.

6. Outlook

Long-term stability of solar cells under realistic operational conditions becomes the chief concern for researchers, as perovskite technology closes the gap between realized progress and prerequisites set by the industry. Materials with low dimensionality display remarkable potential to aid in overcoming obstacles related to device stability, release the strain-induced phenomena and simplify fabrication routes [68, 69]. First, it is vital to develop complete and predictive comprehension of the complex interplay mechanism between 2D materials and perovskite absorber apart from improved stability, as inclusion of additional constituents into light harvesting system exhibits adverse effect on PCE. It is fairly obvious that the diversity of 2D materials complicates the establishment of a unified characterization approach able to promptly reveal ways to prevent the environmental degradation and suggest options to improve charge transport and/or perovskite growth. Accordingly, we need to extend our fundamental understanding of the way RD interacts with charge carrier injection, photoconversion process, oxygen/water ingress, fabrication processing conditions and material doping. Utilizing the full potential of 2D materials in perovskite photovoltaics requires standardized and cost-effective fabrication protocols, ensuring reproducibility and acceptable material quality outputs. To this extent, it is of paramount importance to maintain the innate quality of 2D materials upon transition from physical deposition to a cheaper, large-scale fabrication methods and to address the inevitable trade-off arising from such compromises. Since solution exfoliation route yields high-quality, albeit small 2D flakes retaining optoelectronic properties on par with their physically deposited counterparts, it is prudent to utilize doping capacity of 2D materials in engineering of conventional charge transporting materials. It is highly probable we will witness further expansion of this trend, as it allows researcher to reap the benefits of 2D materials without necessity to obtain uniform coverage of large areas. Alternatively, phase engineering appears to be a very promising way to tackle the increasing rate of disorder resulting from scale-up processing, since formation of interfaces between 2D and underlaying materials has enormous impact on behaviour and opto-mechanical
properties of resulting contact. With the appropriate focus and selection of industry compatible fabrication process, there will be little to no barrier for devices including 2D materials to meet the expectations for commercialization. Moreover, perovskite technology offers both low materials cost and short energy payback time (EPBT) (<0.22 years), which is considerably less than the conventional Si photovoltaics (>10 years). Much lower cost base than incumbent photovoltaic technologies, makes perovskite solar cells a viable business option with less than 20 years of lifetime (Si standards). Therefore, demonstrated device lifetime over 10 years under accelerated ageing pushes perovskites close to deployment benchmark. Finally, designing a framework to understand and anticipate functional response of 2D materials relative to various roles they have within the solar cell structure, should be viewed as a major benchmark conditioned by both theoretical and experimental advancements in the field.

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