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# Designing metal halide perovskite solar modules for thermomechanical reliability



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There is a significant deficiency in perovskite solar module (PSM) stability under thermomechanical stressors which is not well-understood. In this perspective, common issues seen with perovskite solar cell device fabrication related to thermomechanical reliability of PSM processing are discussed, with a focus on how the robustness of device layers and interlayer adhesion can be improved. Film stresses, adhesion of charge transport layers, and instability under light and heat are discussed with the purpose of providing insight on designing PSMs for durability. Processing conditions of encapsulation of PSMs and critical parameters to consider are also examined, and accelerated testing protocols for PSMs are discussed that probe mechanical degradation modes and ensure reliability of devices in the field.

Metal halide perovskites (MHPs) have attracted considerable research interest since their implementation in perovskite solar cells (PSCs) due to their record efficiency improvement since 2009 from 3.8% to 26.1%, low fabrication cost, and relative ease of manufacturing<sup>1-3</sup>. PSCs have the potential to transform the way in which solar energy is harnessed, and the most promising method of commercializing PSCs is through perovskite/silicon (PVSK/Si) tandem cells. The promise of PSCs for tandems lies in significantly higher efficiencies (PCE of 33.9%) with minimized additional overhead cost, enabled by the highly tunable bandgap of MHPs and compatibility with monolithic integration (directly processed on top of silicon PV wafers) in a 2-terminal configuration, or mechanically stacked (deposited on the cover glass above the encapsulant) in a 4-terminal configuration<sup>14</sup>. However, PSCs are challenged in their route to reach commercial deployment and large-scale manufacturing due to several drawbacks, primarily due to their thermomechanical instability, chemical instability, and rapid material degradation in the presence of oxygen, moisture, light, and heat<sup>5-8</sup>. This perspective highlights key parameters of device architecture and design that may aid in the thermomechanical stability of PSCs and perovskite solar modules (PSMs) with a focus on the fracture energy of material layers and interlayer adhesion, the effect of film stresses within the perovskite absorber, the impact of scribing in a PSM as the material is removed and new interfaces are introduced, and the importance of realistic accelerated degradation testing.

## Thermomechanical perspective of device challenges

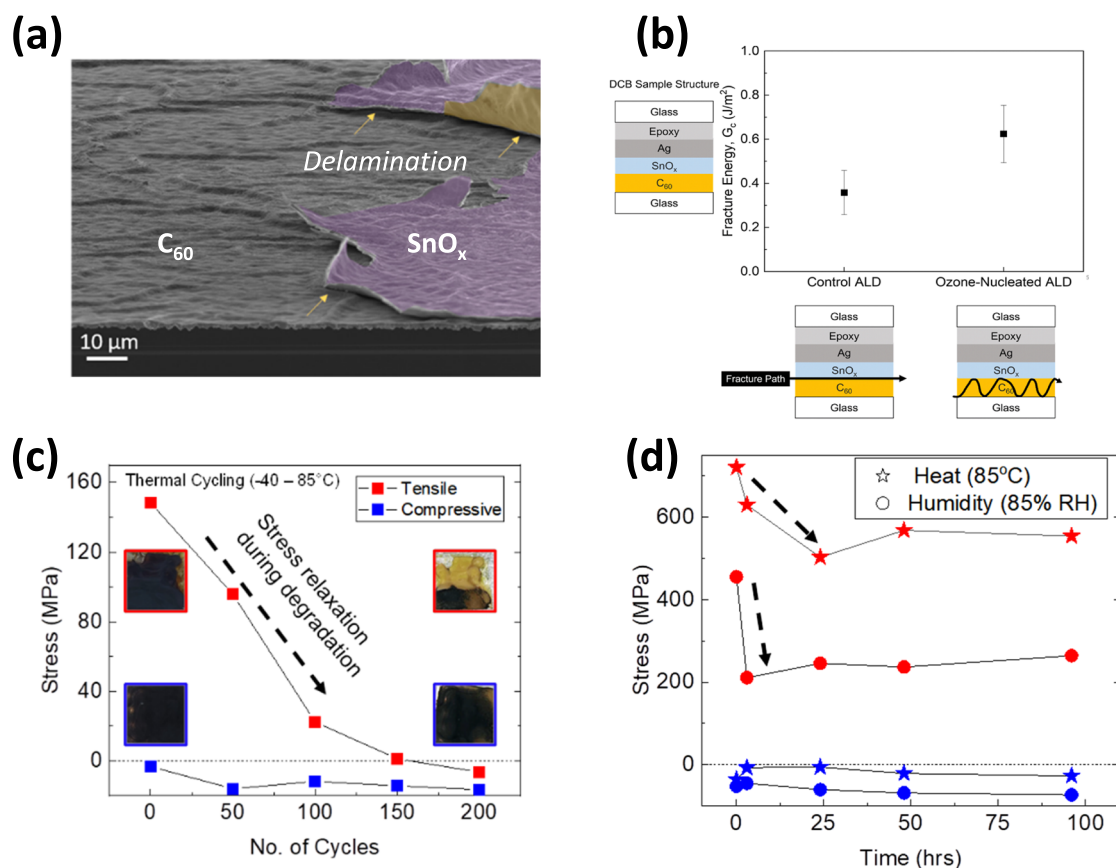
The thermomechanical instability of PSCs has been documented in two forms, the first of which is related to low fracture energy ( $G_c$ ).  $G_c$  is a material's resistance to the propagation of a crack, dependent upon both material/interface bonding energy and the ability of a material to deform

plastically. Due to the low bond strength and brittle nature of charge transport layers, PSCs suffer from extreme fragility and tend to fracture between commonly used fullerene-based electron transport layers (such as  $C_{60}$ ) and the MHP at a  $G_c$  of  $< 0.5 \text{ J/m}^2$ <sup>9</sup>. As such, there is a critical need to increase the  $G_c$  to enable more robust devices that can withstand processing and operating conditions without delamination of layers and failure<sup>9,10</sup>. As seen in Fig. 1a, De Bastiani et al. reported delamination of the tin-oxide ( $\text{SnO}_x$ ) layer from the  $C_{60}$  layer of a fully encapsulated device, indicating a weak interfacial adhesion that can lead to poor device performance<sup>11</sup>. This was confirmed by Li et al., who also suggested strategies to improve the interlayer bonding of  $\text{SnO}_x$  to  $C_{60}$  and increase the  $G_c$  of a device<sup>9</sup>. Figure 1b shows that treating the  $C_{60}$  layer with ozone nucleation prior to deposition of  $\text{SnO}_x$  increases the interlayer adhesion as it allows for improved contact area between the layers and changes the fracture path, thus indicating that the  $\text{SnO}_x/C_{60}$  interface is no longer the most fragile point in the PSC. Although this is the most fragile interface in a full device stack, the fracture energy of the MHP can also be comparably low ( $\sim 0.5 \text{ J/m}^2$ ) if the grain size is relatively small ( $< 100 \text{ nm}$ ) in comparison to the film thickness<sup>12</sup>. Designing the MHP for mechanical robustness is also of importance and can be achieved through improving morphology so the grain size is comparable to the film thickness ( $\sim 500 \text{ nm}$ ).

Another source of mechanical failure in PSCs originates from high tensile film stresses that are caused by the mismatch of the coefficient of thermal expansion between the MHP and the substrate after annealing<sup>8,13</sup>. Tensile stresses in PSCs have been shown to reduce device performance and stability as they increase defect density, reduce carrier mobility, and weaken interlayer strength with the MHP<sup>13,14</sup>. Previous work by Ahmad et al. proposes that the introduction of compressive stress into a MHP film can counter these effects and reduce degradation,

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**Fig. 1 | Delamination and fracture testing of brittle charge transport layers and aging of polymer-scaffold metal halide perovskite (MHP) films.** **a** Delamination of  $C_{60}$  from tin-oxide buffer layer ( $SnO_x$ ) after encapsulation. Reprinted (adapted) with permission from ACS Energy Lett. 2022, 7, 2, 827-833. Copyright 2022 American Chemical Society. **b** Double cantilever beam (DCB) results demonstrating how treatment of  $C_{60}$  prior to deposition of  $SnO_x$  increases interlayer adhesion and changes fracture path. Reprinted (adapted) with permission from Energy Adv.,

2024,3, 273-280. Copyright 2024 Energy Advances. Visible degradation and stress relaxation in a tensile MHP film (red symbols) compared with little degradation and minimal stress relaxation in a compressive film (blue symbols) under (c) thermal cycling and (d) high heat and humidity (stars and circles, respectively). Reprinted (adapted) with permission from ACS Appl. Mater. Interfaces 2023, 15, 44, 51117-51125. Copyright 2023 American Chemical Society.

as well as heal defects within the film<sup>15</sup>. Figure 1c shows an MHP film aged under thermal cycling ( $-40$  to  $85^\circ C$ ) and subsequent relaxation of the tensile stress within the film as bonds between the film and substrate are degraded. Visible degradation within the film is also seen as parts of the MHP turn yellow, where ion migration leads to the formation of lead (II)-iodide ( $PbI_2$ ). Ahmad et al. demonstrate through the use of polymer additives that the presence of an initial compressive stress had visibly less degradation of the MHP film with little  $PbI_2$  observed from X-ray diffraction structural characterization, verified by little to no change in film stress even as the sample was thermally cycled. These results indicate that engineering for compressive stress in a MHP film can increase its stability and lifetime. Figure 1d demonstrates how a similar trend can be seen with MHP films held at either  $85^\circ C$  or 85% relative humidity for 100 hours. Films with an initial tensile stress experienced significant stress relaxation as the MHP degraded, while films with an engineered initial compressive stress experienced only a slight increase in compressive stress after 100 hours. Note that these additives have also been shown to increase the fracture energy of the MHP, further improving prospects for long-term mechanical robustness<sup>16</sup>. Another technique to stabilize PSCs is through surface passivation and the use of 2-dimensional perovskites. Recent results from Azmi et al. demonstrate markedly improved stability of PSCs through the use of passivating 2-dimensional perovskites on either side of the MHP absorber, indicating that such layers can serve to increase both the stability of a device and the robustness of an interface<sup>17</sup>. These results strongly suggest the importance of engineering devices to target mechanically limiting factors of the

MHP, such as film stress and weak interlayer bonding, and indicate the relevance of exploring how a higher  $G_c$  enables mechanical robustness of PSCs at the device level.

### Module-specific considerations

Issues in mechanical robustness and device failure seen at small ( $\sim 1\text{ cm}^2$ ) PSCs are exacerbated in larger sized PSMs, with the added complexity of laser scribing and encapsulation for packaging<sup>3</sup>. Laser scribing is a method in which thin film solar cells are monolithically interconnected in series in order to make a solar module<sup>18</sup>. There are few reports examining the effects laser scribing has on the stress and stability of a device in the region immediately surrounding the scribe lines. The presence of scribes throughout the device layers introduces new interfaces—e.g., between the top electrode and the MHP absorber at the scribe edges—and the presence of these new interfaces can be origins for ion migration, resulting in accelerated chemical degradation in areas surrounding scribe lines, as well as sources for localized stress regions and potential points of delamination<sup>3,18</sup>. Mechanical delamination at scribe lines is detrimental to device performance and stability, and we expect PSMs to be more susceptible to delamination, particularly as they undergo packaging processing such as encapsulation. We highlight here the need for engineering robust devices with high interlayer adhesion/fracture energy.

Another aspect of full-module packages to consider is the presence and processing of the encapsulant itself, a polymer whose functions are to protect the PSM from environmental factors, such as moisture ingress, and adhere to a top barrier (usually cover glass). Encapsulation is generally done

under vacuum hot-pressing, which requires temperatures ranging from 100 to 150 °C and applied pressures ranging from 40 to 70 kPa for up to 15 min<sup>19,20</sup>. Encapsulation temperature, pressure, and time must be carefully tuned in order to properly ensure complete encapsulation without damaging the PSM as the thermal cycling and force from the process can introduce new stresses (specifically shear stresses) into the PSM and lead to delamination and/or degradation. We expect encapsulation will accelerate both delamination and degradation around scribe lines as new interfaces introduced by laser scribing and removal of material layers can weaken interlayer bonding and compromise the mechanical integrity of a PSM. Commonly used encapsulant materials for PSMs are ethylene vinyl acetate (EVA) and thermoplastic polyolefin (TPO), although there are reports utilizing alternative polymers such as thermoplastic polyurethane (TPU)<sup>21</sup>. Effective encapsulant materials for PSMs have low stiffnesses, with reports of effective laminations achieved by encapsulants with elastic moduli ( $E$ ) ranging from 4.5 to 22 MPa<sup>21,22</sup>. The effectiveness of a low  $E$  encapsulant can be attributed to lower shear stress and pressure imparted on the device when the encapsulant is cooled down, thus reducing the potential for delamination. While vacuum lamination is generally the most effective encapsulation technique for long lasting, protected devices, Toniolo et al. found issues with delamination when using vacuum encapsulation for PVSK/Si tandem mini-modules, as seen in Fig. 2a, b<sup>21</sup>.

Toniolo et al. found that while TPU-encapsulated modules had little to no resulting delamination, the mini-modules encapsulated in TPO had occasional delamination issues. This can be attributed to the higher  $E$  of TPO (22 MPa vs 4.5 MPa). However, it is crucial to investigate how other mechanical and material properties of an encapsulant, such as glass transition temperature ( $T_g$ ) and yield strength, affect the extent to which delamination is observed in PSMs. For example, at temperatures below the  $T_g$  of the encapsulant material, a polymer will no longer deform plastically and thus will not be as accommodating of any strain changes from the device layers that accompany temperature fluctuations, particularly at low temperatures during thermal cycling. Toniolo et al. found the  $T_g$  of TPO and TPU to be  $-45$  °C and  $-31$  °C, respectively, through differential scanning calorimetry. While the TPU-encapsulated modules exhibited less delamination due to their lower  $E$ , one can expect from the higher  $T_g$  that TPU will exhibit more stress and potential delamination at low temperatures.

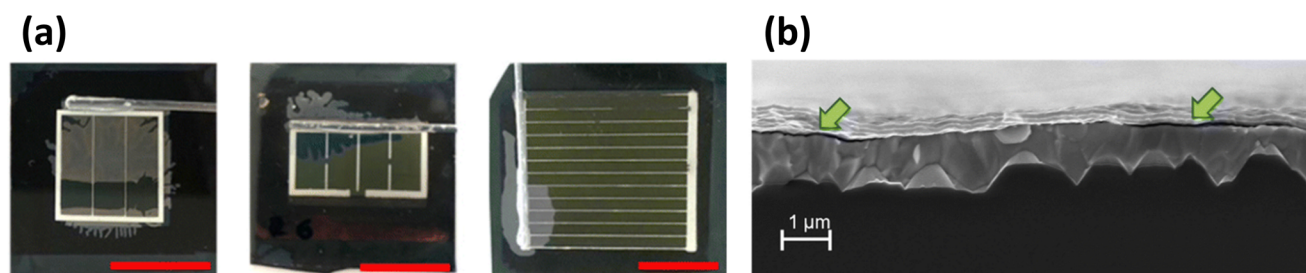
There are still several other module-specific considerations that require discussion and examination. For example, the design properties of the encapsulated module itself need to be considered, such as encapsulant and cover glass thickness. While reports have already demonstrated that low encapsulant stiffness is a critical property for effective encapsulation, we also expect the thickness of the encapsulant and cover glass to have an impact on the stress experienced within the device, and therefore affect the potential for delamination. The edge seal is another factor that should be considered; a polymer with a very low water vapor transmission rate (generally butyl rubber or polyisobutylene) is placed on the sides of an encapsulated module to prevent lateral moisture and oxygen ingress. The presence of an edge seal introduces an additional mechanical constraint in the module and can affect stress distribution, and few reports examine how its presence affects

delamination, stress, and overall stability of a full-package module based on the thermomechanical properties of the edge seal. Furthermore, volatilization of the MHP can release degradation species, such as iodine gas ( $I_2$ ) and iodomethane, that remain trapped within an encapsulated device and further accelerate degradation<sup>23,24</sup>. Trapped gas within an encapsulated module could also increase the pressure exerted upon the PSM and could induce more stress than the advance potential delamination, leading to accelerated degradation as individual layers are compromised.

It is also critical to recognize that failures of devices and device lifetimes within a laboratory setting differ from failures in the field and field lifetimes when testing for commercial viability. For example, the Perovskite PV Accelerator for Commercializing Technologies (PACT) highlights the need to test the stability of PSCs and PSMs at elevated temperatures and light simultaneously as these conditions lead to rapid degradation of MHPs<sup>25</sup>. The accelerated degradation of MHPs under both light and heat has been well documented, however few demonstrate device stability under both stressors (and none at the module level), which is crucial to demonstrate MHP stability in the field. Additionally, reports from PACT of outdoor field testing of single junction PSMs show significantly lower stability lifetimes than PSCs tested in a laboratory, indicating the presence of expected additional degradation mechanisms in a PSM<sup>26</sup>. To effectively test accelerated degradation of devices in a situation more similar to the operation in the field, we propose the introduction of measuring thermal cycling under constant illumination in addition to constant heat under illumination. There is a significant difference in degradation mechanisms between applying constant heat to a device and thermally cycling a device, as the effects from thermal cycling will become more prevalent at locations with higher stresses and larger mismatch of coefficient of thermal expansion, such as near the scribe lines in a PSM. The inferior stability of PSMs can be derived from a number of causes; however, we hypothesize that the decreased thermomechanical robustness of PSMs due to scribe lines likely plays a key role in decreasing the long-term stability of PSMs tested in the field. We propose that the degradation mechanisms of PSMs are centered around the new interfaces and localized stress regions introduced by scribing, and that additional PSM processing (e.g., encapsulation) has unexplored and not-yet understood effects on device degradation.

### Proposed solutions

There are several reported and proposed methods in mitigating the thermomechanical fragility of MHPs. The most direct methods address increasing the  $G_c$  of individual layers within a PSC and lowering the film stress within the MHP and the overall module. Increasing the toughness of layers can be done in a number of ways, such as through use of a top carbon electrode—a hydrophobic layer that prevents moisture ingress—and/or the implementation of a scaffold, which provides mechanical support<sup>27–29</sup>. Note that carbon also serves as an inert electrode, while silver (Ag) has been shown to degrade MHPs through ion migration<sup>30</sup>. Reports in the literature have demonstrated a PSC with scaffolding exhibits longer lasting stability, with various material classes used for scaffolding, from mesoporous metal oxides to natural clays<sup>28,31</sup>. Long-term stability from scaffolding has been



**Fig. 2 | Delamination seen in perovskite/silicon (PVSK/Si) tandem modules.** **a** Visible delamination of the encapsulant layer off the PVSK/Si tandem on a macroscopic scale. **b** Microscopic delamination between the  $SnO_x$  buffer layer and the

$C_{60}$  layer due to encapsulant lamination. Reprinted with permission from Nanoscale, 2023, 15, 16984–16991. Copyright 2023 Nanoscale under CC BY 3.0: <https://creativecommons.org/licenses/by/3.0/>.

attributed to surface passivation, facile crystallization of the MHP, and decreased defect concentration; however, one can expect the mechanical robustness of a scaffold-MHP to be greater than that of a thin film MHP, further protecting a device against degradation mechanisms, particularly as a device undergoes additional processing steps with PSM fabrication. Another method of increasing the stability of MHPs is through stress engineering. As mentioned previously, Ahmad et al. utilized low-cost polymer additives in MHP thin films in order to induce compressive stress. Low stress in an MHP film can increase its robustness and stability, even under thermal cycling and high humidity, when compared to a typical MHP film in tension. These additives also serve as scaffolds that improve mechanical properties relevant to the stability of PSCs, with  $G_c$  values greater than  $5 \text{ J/m}^2$ .

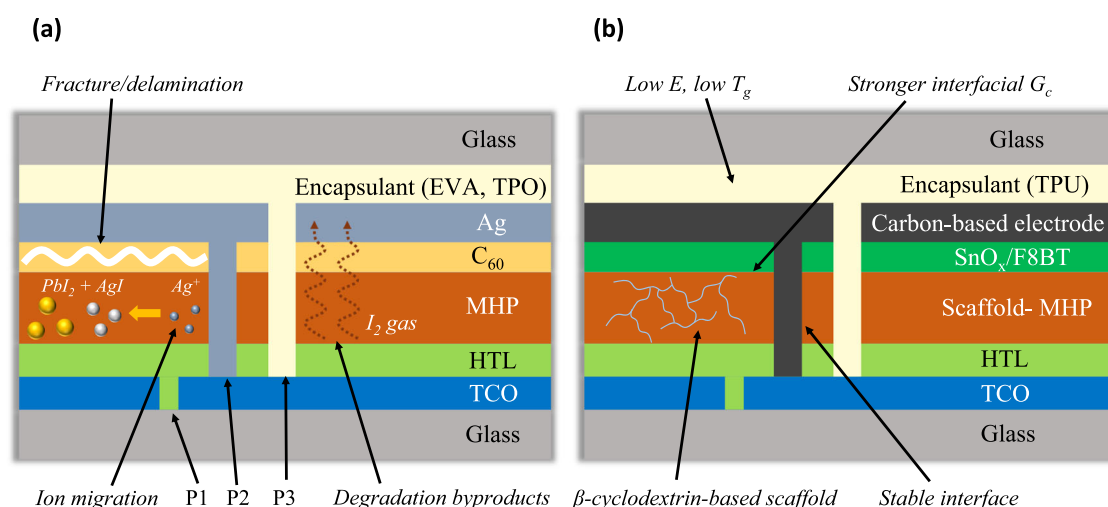
Another pathway to increase the thermomechanical reliability of MHPs is through careful engineering of encapsulant and module design. This includes understanding how the presence of scribes in a module can impact the stability of a device. The P2 scribe, which allows for interconnection of one cell to another, significantly weakens a device as it removes most layers in the stack to allow for the deposition of the top electrode. This could prove to be a source of induced stress and delamination, as well as a new interface for ion migration and chemical degradation, particularly if the top electrode is Ag<sup>33,32</sup>. The P3 scribe, which electrically isolates cells from the top electrode, could prove to be a reinforcement mechanism as the encapsulant will fill the P3 and could provide a type of “anchor point” that increases the fracture energy of a device. Additionally, the encapsulant stiffness and thickness must be tuned to lower the probability of delamination and not induce additional stress into the module. As mentioned previously, Toniolo et al. have demonstrated effective encapsulation of devices by utilizing a low  $E$  encapsulant such as TPU while few reports mention tuning encapsulant thickness and observing effects on delamination and stress. A PSM engineered for robustness and stability must target specific degradation mechanisms, including ion migration, volatile byproducts ( $\text{I}_2$  gas) that further accelerate degradation, and brittle layers ( $\text{C}_{60}$ ). Figure 3a shows a typical PSM and common degradation pathways, while Fig. 3b demonstrates how engineering compressive stress and tuning layer properties can improve the thermomechanical reliability of a device.

An inert electrode such as carbon (as either graphene or carbon nanotubes) should avoid degradation pathways associated with Ag corrosion and ion migration, provide a hydrophobic top layer, and also enable use

in tandem configurations due to optimal optoelectronic properties<sup>33</sup>. Although carbon is generally used as a top contact in n-i-p and architectures without a hole-transport layer due to its work function aligning with hole-transport mechanisms, there have been reports of its utilization in p-i-n architectures, specifically as carbon nanotubes. Note that the use of carbon in a p-i-n architecture generally requires a buffer layer for optimal electrical contact, such as chromium, or a modifier such as polyethyleneimine to adjust the work function of the interface<sup>34–36</sup>. The presence of a scaffold within the MHP can increase the stability of the device by inducing compression into the MHP and can also address other degradation mechanisms, such as volatile byproducts, if an  $\text{I}_2$  absorbing compound is utilized. For example, Li et al. demonstrate improved long-term stability of PSCs through the use of an iodine-trapper,  $\beta$ -cyclodextrin, and they report increased stability of PSCs at elevated temperatures as the iodine-trapper increases the temperature at which iodine volatilizes<sup>37</sup>. A non-fullerene electron transport layer, such as  $\text{SnO}_x$ , can serve as a more robust alternative to increase the  $G_c$  and interlayer adhesion of a device. It is important to note that there are challenges with degradation of the MHP during  $\text{SnO}_x$  deposition, however there is work being done on depositing buffer layers to address this issue<sup>38</sup>. There has also been previous work demonstrating the use of a polymeric electron transport layer, Poly (9,9-dioctylfluorene-alt-benzothiadiazole) (F8BT), in conjunction with  $\text{C}_{60}$ , indicating the potential use of F8BT with  $\text{SnO}_x$  to serve as both a barrier layer for the MHP and charge transport layer if deposited prior to  $\text{SnO}_x$ <sup>39</sup>. The interconnection of delamination throughout a device stack, corrosion of the Ag, and degradation of the MHP must be recognized and addressed, as all three pathways can lead to device failure.

## Outlook

The future of stable and efficient PSMs lies in understanding the interconnection between various degradation modes (mechanical, thermal, and chemical) under multiple stressors (light, heat, and humidity). The presence of scribe lines in a PSM fundamentally changes the device architecture, and thermomechanical robustness must be achieved to ensure devices can withstand processing steps (encapsulation) to mitigate degradation. An empirical estimate is to achieve modules with a minimum  $G_c$  of  $1 \text{ J/m}^2$  to ensure the ability of the MHP module to undergo processing and packaging steps without mechanical failure, as well as reduce the potential for delamination and accelerated degradation. Realistic testing of devices with



**Fig. 3 | Schematic comparing a typical perovskite solar module (PSM) and a PSM engineered for robustness that utilize the same transparent conducting oxide (TCO) and hole-transport layer (HTL).** **a** PSM utilizing a non-reinforced metal halide perovskite (MHP), a brittle electron-transport layer (ETL) composed of fullerene ( $\text{C}_{60}$ ), and a reactive silver (Ag) electrode. Degradation pathways due to ion migration and release of iodine gas ( $\text{I}_2$ ) is depicted, and commonly used

encapsulants, such as ethylene-vinyl acetate (EVA) and thermoplastic polyolefin (TPO) are shown. **b** Robust PSM utilizing an inert, hydrophobic carbon-based electrode, an  $\text{I}_2$ -absorbing scaffold MHP (utilizing  $\beta$ -cyclodextrin), a non-fullerene ETL utilizing tin-oxide ( $\text{SnO}_x$ ) and F8BT, and a low stiffness ( $E$ ), low glass transition temperature ( $T_g$ ) encapsulant, such as thermoplastic polyurethane (TPU).

multiple simultaneous stressors to simulate operation in the field is crucial to bring them closer to commercial maturity. Progress is being made on device stability under laboratory testing and device efficiencies are steadily rising, however, the stability gap of PSMs and PSCs must be studied more thoroughly to address and overcome failure mechanisms in PSMs and enable MHP commercial viability.

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### Author contributions

M.C. wrote the paper. M.C. and N.R. contributed to the discussion and revision of the paper.

### Competing interests

The authors declare no competing interests.

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