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RESEARCH ARTICLE



Review of technology-specific degradation in c-Si, CdTe, CIGS, dye sensitised, organic and perovskite solar cells in photovoltaic modules: Understanding how reliability improvements in mature technologies can enhance emerging technologies

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Abstract

A comprehensive understanding of failure modes of solar photovoltaic (PV) modules is key to extending their operational lifetime in the field. In this review, first, specific failure modes associated with mature PV technologies, such as crystalline silicon (c-Si), copper indium gallium selenide (CIGS) and cadmium telluride (CdTe), are framed

Abbreviations: ALT, Accelerated life testing; a-Si, Amorphous silicon; BOS, Balance of system; CdS, Cadmium sulphide; CdTe, Cadmium telluride; ClGS, Copper indium gallium selenide; c-Si, Crystalline silicon; EL, Electroluminescence; EVA, Ethylene vinyl acetate; FA, Formamidinium; FF, Fill factor; HTM, hole-transporting materials; IEC, International electrotechnical commission; IR, Infrared; ITO, Indium tin oxide; J_{sc}, Short-circuit current density; LETID, Light and elevated temperature-induced degradation; LID, Light-induced degradation; MPP, Maximum power point; NFA, Nonfullerene acceptors; OPV, Organic photovoltaic; PCE, Power conversion efficiency; pc-Si, Polycrystalline silicon; PID, Potential-induced degradation; PSC, Perovskite solar cell; PV, Photovoltaic; RH, Relative humidity; sc-Si, Single-crystalline silicon; T₈₀, Time taken to fall to 80% of original value; TCO, Transparent conducting oxide; UV, Ultraviolet; V_{oc}. Open circuit voltage.

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by sources of specific failure modes, their development from the early-developmental stages onwards and their impact upon long-term performance of PV modules. These failure modes are sorted by both PV technology and location of occurrence in PV modules, such as substrate, encapsulant, front and rear electrode, absorber and Interlayers. The second part of the review is focused on emerging PV technologies, such as perovskites solar cells, dye sensitised and organic PVs, where due to their low to medium technology readiness levels, specific long-term degradation mechanisms have not fully emerged and most mechanisms are only partially understood. How-ever, an in-depth summary of the known stability challenges associated with each emerging PV technologies are reviewed and considerations are given in to how these might be applied to the further development of emerging technologies. Namely, any emerging PV technology must eventually pass industry-standard qualification tests, while warranties for the life time of modern c-Si-based modules might be extended beyond the existing limit of 20 to 25 years.

KEYWORDS

climate, degradation, energy payback time, photovoltaics, reliability, solar cells, solar photovoltaic modules, stress, wearout

1 | INTRODUCTION

The degradation of photovoltaic (PV) modules is one of the key factors that influences the cost of the electricity produced over their warranted life time of 20 to 25 years,^{1,2} while several PV manufacturers are now estimating a useful life of more than 40 years.³ To reduce the degradation, it is hence imperative to know the degradation and failure phenomena. During their operational lifetime, PV modules are subjected to numerous environmental stresses such as light, heat, moisture and mechanical stress, which are largely responsible for these phenomena.⁴ To optimise reliability and predictability and to enhance the module lifetime, it is crucial that degradation and failure mechanisms are known and can be easily recognised and contained.

This review article provides a comprehensive review of degradation and failure phenomena in existing mature and new emerging PV technologies. For this review, literature originating from the past 30 years has been explored by a team of 16 PV experts from various organisations who are members of Working Group 2 on Reliability and Durability of PV of COST Action PEARL PV in the period from September 2018 until July 2021. As failure modes can be different between differing technologies, this review has been partioned into three sections. Part 1 focuses on the technological specific degradation modes of mature PV technologies and introduces degradation modes found in silicon PV (Section 2.1), cadmium telluride (CdTe) (Section 2.2) and copper indium gallium selenide (CIGS) (Section 2.3). In part 2, a review of known failure modes and areas of future research for emerging technologies such as dye sensitised solar cells (DSC) (Section 3.2), organic PV (OPV) (Section 3.3) and perovskite solar cells (PSCs) (Sections 3.4 and 3.5) is enclosed. In the final part, it is discussed how known failure mechanisms and developmental issues that were discovered in mature and commercially available PV technologies might impact emerging PV technologies. The long-term stability of third generation PV technologies, such as perovskite PV, remains a challenge and needs to be addressed for rapid commercialisation. Learning lessons from mature technologies might speed up development of these emerging technologies.

2 | DEGRADATION AND FAILURE OF MASS-PRODUCED TECHNOLOGIES

Even though different types of environmental, electrical and mechanical stress factors are common to all PV technologies, responses of each technology to the stress loads can vary largely. Degradation rates, mechanisms and failure modes depend on the materials chemistry, structuring of the cell stack and the packaging of each system. This section examines the technology-specific phenomena for solar cells and PV modules based on c-Si, CdTe and CIGS as well as OPV, DSC and PSC. For each technology, failure modes and degradation mechanisms are described in detail along with typical mitigation strategies. As such, Table 1 has been prepared to highlight the key <u>specific</u> <u>degradation and failure mechanisms</u> for each PV technology in order to provide an overview for the reader. In the following sections, these mechanisms are discussed in greater detail.

TABLE 1 Summary of the specific degradation and failure mechanisms of the PV technologies discussed in this article

	, ,	J	5	
	Substrate	Encapsulant	Front (f) and rear (r) electrode	Absorber (A) and interlayers (I)
c-Si	Backsheet cracking and delamination	Burn marks	 Snail Trails (f) Solder bond and ribbon failures (f, r) 	Cell cracks (A)Hot Spots (A)PID, LID, LETID (A)
CdTe		Edge sealant failure	 PID (f) Cu diffusion to front Junction (r) Molybdenum back contact oxidation (r) 	Recombination centres due to sodium (glass migration) (A)
CIGS		Edge sealant failure	 Increase of ZnO:Al resistivity (f) PID (f) Molybdenum back contact oxidation (r) 	 PID (A) Alkali accumulation at the ZnO:Al and pn junction (I) Wormlike defect formation (A)
DSC	Pin holes in (flexible) barrier films	 Incompatibility of encapsulant with electrolyte Edge sealant failure 	 Dissolution of active later into electrolyte (r). Change of electrocatalytic properties (r) Dissolution from the substrate (r) 	 Dye molecule photodegradation (A) Electrolyte photodegradation (A) Reactions between the dye and electrolyte (A) Oxidation and reduction reactions at the TiO₂ surface (A)
OPV	Pin holes in (flexible) barrier films	Edge sealant failure	 Chemical degradation or oxidation of electrode (f, r) Electrode diffusion (f, r) Electrode cracking (f, r) Electrode delamination (f, r) 	 Chemical or photo degradation of the electron donor/acceptor (A) Loss of percolating paths due to blend reorganisation (A) Change in energy levels (A, I)) Metastable film morphology (A) Hole/Electron transport layer diffusion or decomposition (I) Degradation of the quality of the absorber and electrode interface (I)
PSC	 Information not available 	Information not available	 Au or Ag corrosion due to iodine migration (r) Electrode diffusion in to charge selective layers (r). Oxide formation leading to unfavourable interface with charge selective layers (r) 	 Loss of absorption due to absorber layer degradation (intrinsic, moisture or photo- induced) (A) Migration of dopant from interlayer to absorber layer (A) Phase separation (A) Crystallographic changes (A) Change in energy levels (A, I) Hole/Electron Transport layer degradation (I) Dopant diffusion into active layer (I) Change of uniformity of interlayers (I)

Abbreviations: CdTe, cadmium telluridel; CIGS, copper indium gallium selenide; DSC, dye sensitised solar cells; LETID, Light and elevated temperatureinduced degradation; LID, light-induced degradation; OPV, organic photovoltaic; PID, potential-induced degradation; PSC, perovskite solar cell; PV, photovoltaic.

2.1 | Crystalline silicon (c-Si)

Silicon, the second most abundant element on the earth's surface, is the most developed semiconductor material for PV applications and dominates the market.⁵ Being one of the oldest PV technologies, its degradation mechanisms have been studied extensively.⁶⁻⁹ In addition to common environmental and voltage stresses, the c-Si systems can also suffer from mechanical loads because silicon wafers are relatively stiff and brittle. This section examines the degradation mechanisms and features, such as cracks, snail trails and hotspots, that could be induced by these stresses. We also discuss degradation modes that can be observed at c-Si PV systems, including potential-induced degradation (PID), light-induced degradation (LID) and light and elevated temperature-induced degradation (LETID).

2.1.1 | Cracked cells

The reduction of silicon wafer thickness aims to decrease the cost of silicon-based PV cells and modules. Nevertheless, the smaller thicknesses decrease the robustness of solar cells against mechanical loads and may cause cell cracking.¹⁰ Cells that crack during the production process can be detected and eliminated. However, it is not possible to entirely avoid the formation of microcracks on the PV cells; therefore, it is crucial to quantify their long-term effects on the performance of PV modules. Microcracks and imperfections increase the risk of breakage during the production cycle and can propagate further during the lifetime of PV modules.^{11,12}

Microcracks may form in several stages, namely, during (1) ingot cutting, (2) production of cell and module, (3) transportation and installation and (4) operation of PV system due to environmental factors such as temperature cycles, wind, snow and hail.¹²⁻¹⁴ Cracks cause to disconnect the electrical conductivity in cell regions which leads to reduction in the short-circuit current and the increase of the series resistance, resulting in output power reduction of PV modules up to 2.5% in c-Si solar cells.^{15,16} The position, length and orientation of microcracks influence this power reduction.¹⁶

K. Schulze et al. have reported up to 20% power losses based on degradation analysis of more than 250 PV modules after 15 years operation which were affected by cracks in combination with delamination and ethylene vinyl acetate (EVA) browning.¹⁷ In other studies,^{18,19} experimental results have proven that the cracking reduced the fill factor (FF) and output power up to 4% and 3%, respectively.

Cracks and microcracks are distinguished based on their size: A crack with a width up to $30 \,\mu\text{m}$ is classified as a microcrack.²⁰ Cracks occur in different shapes and sizes.²¹ Cracks in c-Si PV cells and modules are further classified using other criteria such as severity and position^{15,22} (see Table 2). Star-shaped cracks consist of several

line cracks originating from an induced point. Line-shaped cracks are also initiated due to laser-cutting of wafers.²³ Figure 1 shows the classification of cracks according to their orientations in silicon PV cells.

2.1.2 | Snail trails

Crystalline-Si PV modules in the field may develop local line-shape discolorations, so-called snail trails, over the cells after a period of months to a few years; see Figure 2. Closer inspection shows that the discoloration occurs on the silver paste only. Snail trails form in the presence of cell cracks and depend on the packaging polymers (e.g., EVA acetic acid formation).^{25,26} Moisture ingress seems to be the cause of silver line corrosion. Chemical reactions at the silver paste-encapsulant interface may lead to the formation of silver-containing nanoparticles above the silver lines.²⁷ The line conductivity is only little reduced in the process. Optical transmission loss is also negligible as the discoloration happens above the silver lines; the encapsulant away from the silver line remains unaffected.²⁷

While the snail trail itself does not have a direct or significant effect on cell or module performance, it is an indication of moisture ingress, commonly caused by mechanical stress-induced loss of module hermeticity and cell cracking. These are performance risks. Consequently, some reports indicate that the impact of snail trails on PV performance is negligible,²⁸ but other studies conclude that output energy produced by PV modules can be reduced up to 20% due to snail trails.^{29,30} It appears that in some cases, positive correlations between the occurrence of snail trails and power loss are misinterpreted as causal, whereas instead they have a common origin. To observe snail trails in an accelerated test, a combination of mechanical load, UV exposure and temperature elevation is recommended.^{31,32}

 TABLE 2
 Cracks classification based on direction, position, size, shape and severity¹⁴

Direction Position Size Shape Severity	
Diagonal +45° and -45°FacialMacrocracksLine-shapedMode A:+45° and -45°SubfacialMicrocracksStar-shapedNo significant power lossesParallel to busbarsMicrocracksStar-shapedMode B:Perpendicular to busbarsPartially isolated and electrically inactive, causing po degradation and hotspotMultiple directionsVertexMode C: Completely isolated and electrically inactive, causing degradation and hotspot	ower g power



FIGURE 1 Classification of cracks according to their orientations in silicon photovoltaic (PV) cells: (A) no crack, (B) perpendicular, (C) parallel, (D) dendritic, (E) multiple directions, (F) $+45^{\circ}$, (G) -45° (reprinted from Papargyri et al.¹⁴)

2.1.3 | Hot spots

A hot spot is a high-temperature area on the PV module which may cause serious damage on the solar cells and other elements on the modules (see Figure 3). It forms when one or more solar cells generate less current than the string current of a PV module. A hot spot can occur in a PV module due to various causes, such as cells mismatching, partial shading or interconnection failures.^{8,35–37} Indeed, when the cell is affected by partial shading, it results in a short-circuit condition or a reverse bias.^{37,38}

The cell temperature can also be nonuniform. Hot spots can occur within a cell in case of manufacturing defects, such as deformations at the p-n junction, a local impurity imbalance or metallurgical shunts.^{1,39-43} Cells with such hot spots are normally identified and rejected during cell testing, but they can also form over time due to cell and module degradation.

2.1.4 | PID

High potential differences over insulators, in the kilovolt range, tend to cause such insulators to fail. Small leakage currents and local discharging may occur under these circumstances. PV modules have high potential differences between the cells and the grounded frame. PV



FIGURE 2 The photograph of snail trails on photovoltaic (PV) modules²⁴



FIGURE 3 Thermal infrared (IR) photograph of a photovoltaic (PV) module affected by hotspots^{33,34} 6 WILEY PHOTOVOLTAICS

modules are daisy-chained to improve power efficiency and lower the system cost. In that case, the potential difference between cells and frame may reach \sim 1000 V, depending on the module, combiner and inverter ratings. This may soon even go as high as 1500 V, as some manufacturers are pushing to reduce balance of system (BoS) cost.

Degradation phenomena in PV modules related to this high voltage are termed PID. Discolouration, delamination, microcracks, shunts and even stacking faults are observed in c-Si leading to significant power and efficiency losses. The EL images after the PID stressing show that the degradation is strongest at the frame edges. The standard PID test procedure follows stressing the module with an external bias of 1000 V in a climate chamber of 60°C (or 85°C) and 85% relative humidity (RH) (IEC 62804-1).

Two main mechanisms have been identified in PID of c-Si: (1) shunting (PID-s) and (2) surface polarisation effect (PID-p). In PID-s local shunts are formed over the emitter. The current understanding of PID-s is that sodium ions from the cover glass migrate to the cell due to the external bias and get reduced to metallic sodium in the n + emitter. Here, they decorate pre-existing stacking faults to form a conductive path between the n-doped emitter and p-doped base.44

On the other hand, PID-p results from a surface polarisation effect due to the accumulation of net charge in the dielectric stack between frame and cell, a typical case of high voltage creepage. This accumulated charge changes the surface field of the cell, leading to a reduction of short-circuit current and open circuit voltage.⁴⁵

LID and LETID 2.1.5

LID and LETID are two phenomena that can be observed in c-Si systems and result in significant reduction of minority carrier lifetime in the bulk of c-Si wafers and the solar cells.46,47

LID can occur even at low light exposure at room temperature and the formation of a boron-oxygen defect in the silicon wafer is the main degradation mechanism.⁴⁶ To mitigate the effects of LID, it has been proposed to decrease the oxygen content or substitute boron by other dopants such as gallium.⁴⁷

LETID is a specific degradation type first observed on PERC-type multicrystalline Si PVs in the field.⁴⁸ Later works showed that mono-Si cells also suffer from degradation under the combination of light and temperature stress. In comparison with LID, which occurs in a short period time of initial exposure to sunlight, LETID develops more slowly. The consequences are severe with high efficiency and power losses.49

As in the case of LID, boron-oxygen complexes can be observed after LETID, but are not the main root cause for degradation.48 Hydrogen redistribution phenomena are currently considered responsible.⁴⁹ Preventive measures that can be taken for the mitigation of LETID include use of silicon wafers with low oxygen content, dielectrics with little hydrogen and low firing temperatures.

2.2 CdTe

CdTe technology dominates the thin film PV market based on the relatively low cost for manufacture, increases in module efficiency (18%), small temperature coefficient (0.25%/°C) and large scale of manufacture.³⁹ The latter has been largely due to one manufacturer, First Solar, who now produces in excess of 5 GW per year. The history of CdTe module deployment is much shorter than that for c-Si so less is known about the long-term performance.

Wendlandt et al.⁴⁰ reported the range of measured degradation rates for CdTe modules to be 0.2%-4%/year per year with a median value of 0.5%/year. The range can partly be attributed to variations in the manufacturing method and module sealing but is largely due to the nonlinear development of CdTe module degradation with time.

The IEA-PVPS Report (2014)³¹ mentions the main CdTe-specific failure mechanisms as being:

- Front glass breakage that can cause short term failure;
- Back contact degradation that causes longer-term loss of performance.

These are detailed in the following subsections. In addition, we will treat PID in CdTe modules.

2.2.1 Front glass breakage

A consideration for CdTe PV technology is the constraint imposed on the manufacturing process by the superstrate configuration where the front glass is used as the substrate for depositing the thin films in the PV device. This prohibits hardening or tempering of the front glass (the superstrate) because it has to endure a series of temperature cycles during the deposition of the thin film coatings and heat treatment. The lack of hardening or tempering makes CdTe modules more susceptible to failure due to front impact (see Figure 4).^{50,51}

2.2.2 Back contact degradation

The role of copper in CdTe modules has been an important factor for reducing back contact series resistance and doping the CdTe absorber layer.⁵² However, too much copper applied to the back surface will result in diffusion to the front junction and cause an increase in carrier recombination and loss in Voc. Controlling the amount of copper applied to the back surface of CdTe is therefore crucial to obtain high efficiency devices. Artegiani et al.⁵³ present evidence that just 0.1 nm of Cu suffices. The drop in PV module performance over the first 2-3 years of deployment is attributed to copper diffusion to the front junction. As Perenoud et al.⁵² have pointed out, the solubility of copper in CdTe is low; Cu concentrates at the crystal grain boundaries, providing a fast diffusion pathway to the front junction.



FIGURE 4 A hot spot due to cracked front glass (not readily spotted by visual inspection) detected using aerial infrared thermography^{50,51}

The temperature coefficient of CdTe is -0.25% per °C temperature rise, half that of c-Si. That makes CdTe an attractive choice in warm climates; however, the high operating temperatures could enhance the diffusion of copper. Strevel et al.⁵⁴ have observed that a 4%–7% power loss over the first 1–2 years before a linear degradation factor of -0.7%/year is established. The initial drop goes faster at higher operating temperatures. Strevel et al. state that the initial power output of the module is underspecified to deal with this initial degradation.

In a series of controlled laboratory heat cycling tests on experimental CdTe solar cells, Bertoncello et al.⁵⁵ have attributed degradation to two different mechanisms. The first is copper diffusion, and the second is oxidation. One of the observations was an increase in series resistance which was attributed to loss of copper from the back contact accompanied by a conversion of low resistance Cu₂Te to high resistance CuTe. The diffusion of the excess copper through the CdTe to the CdS buffer layer causes loss of short wavelength external quantum efficiency. The oxygen ingress during accelerated heat testing caused the formation of TeO at the back contact, increasing resistance. However, in a paper on As-doped CdTe, an air anneal resulted in enhancement of the V_{oc}^{56} The oxidation degradation might occur specifically to Cu-doped back contacts.

A radical solution to copper-related degradation is to replace Cu by another element; preferably one with a higher solubility, to obtain higher acceptor concentrations. With As doping, this concentration has been shown to exceed 1×10^{16} cm⁻³.^{57,58} Arsenic is a slow diffuser in CdTe solar cells and does not appear to diffuse into the buffer layer so it should improve the long-term stability. Experimental arsenic-doped CdTe modules have now undergone the standard thermal stress test subjected to conventional Cu-doped modules.⁵⁷ Remarkably, the Asdoped modules show an initial rise in efficiency over the same period that the Cu-doped module shows the steep decrease mentioned earlier. This is followed by an efficiency constant over time, bearing the prospect of an improved long-term performance.

2.2.3 | PID

Like c-Si modules, PID in CdTe modules is also strongest on the negative string end.⁵⁹ Modules from a 2.3 MW CdTe plant that had been

operational for 6 years showed 43% power loss compared with the nominal power on the negative end and 17% power loss on the positive string end, the latter probably unrelated to PID. PID degradation came with visible transparent conducting oxide (TCO)-corrosion around the clamps and edge region. In some cases, the reverse bias protection had failed.

As with c-Si solar modules, the mechanisms governing PID in CdTe modules are not yet fully understood. Leakage currents develop due to the large potential difference between the grounded frame and the cells, particularly those at negative potential. The glass superstrate for CdTe modules is typically a soda-lime glass, and sodium can migrate towards the cell and reach the junction region degrading cell performance by introduction of recombination centres. This typically degrades the V_{oc} and FF of the cell. The mechanism of Na transport and the subsequent cell degradation depend on the moisture conditions. In a dry climate, the sodium migration described above will operate and can be reversed by subjecting the modules to a reverse bias. Moisture ingress, however, will result in irreversible degradation of the module as the reduced sodium will react with the moisture to produce atomic hydrogen which will then react with the SnO₂ based TCO. This is the mechanism that leads to visible TCO degradation near the edges of a module (see Figure 5).

2.3 | Copper indium gallium diselenide (CIGS)

CIGS is now one of the most mature thin film PV technologies with rapid growth of instalments and production capacity thanks to their low fabrication costs, short energy payback time and most importantly due to their freedom of size, shape and flexibility, which makes them suitable for integration in various infrastructures.⁶¹ Jordan et al. reported that CIGS modules installed in the 21st Century demonstrated low median power degradation rates of 0.5% per year.⁶² The majority of the modules showed rates between 0% and 1% per year, while some modules actually improved during outdoor operation. A small quantity of outlier modules showed worse field behaviour.

Degradation in CIGS PV systems can be induced by various stress loads including humidity, partial shading and biases. CIGS solar devices are formed as a multilayered material stack and responses to

such stress loads differ for each layer. Figure 6 demonstrates the cross-sectional schematic of the material stack in a typical CIGS solar device and illustrates the degradation mechanisms.

This section then looks into these mechanisms and features in detail under following subsections:

- Decrease of conductivity of TCO front contact and molybdenum oxidation of the scribes due to water ingress
- Alkali element migration promoted by internal (under illumination) and external biases (PID)
- Wormlike defect formation due to partial shading.

2.3.1 | Reduction of contact conductivity due to water ingress

A literature review⁶³ revealed that nonpackaged CIGS devices show strongly varying degradation rates under damp heat conditions



FIGURE 5 Images of a CdTe module (a) before and (b) after 1043 h of voltage stress of -1000 V in a chamber of 85°C/85%RH, the latter exhibiting transparent conducting oxide (TCO) corrosion⁶⁰

(85°C/85% RH). The most impacted device parameters were the FF and the open circuit voltage. These devices were on the other hand mostly stable when exposed to dry heat conditions, which was also the case for packaged devices exposed to damp heat. This indicates that adequate packaging, both flexible and rigid, is sufficient to keep the devices stable. Nevertheless, in case of insufficient water protection, like a damaged edge seal or a broken front sheet, humidity could enter a CIGS device. This can have a negative impact on especially the conductivity of the front contact and the monolithic interconnection.

In CIGS devices, several types of TCOs are used as front electrode. Sputtered aluminium-doped zinc oxide (ZnO:AI) is the most common material, while sputtered tin doped indium tin oxide (ITO) can be implemented as well.

In the case of unpackaged CIGS solar cells, thus allowing water ingress, increased resistivity of ZnO:Al is often found to be a major cause for efficiency loss. The initial resistivity of the ZnO:Al is used to define cell width, so even minor resistivity increases will directly impact the device performance.⁶³ Damp heat-related resistivity increase of ZnO:Al is primarily caused by a decrease of carrier mobility due to grain boundary degradation. This is typically caused by the diffusion of 'foreign' species, like water and CO₂, from the environment into the grain boundaries,^{64,65} where the potential barrier can then increase.^{66,67} The resistivity increase was reported to be largely reversible by annealing in vacuum⁶⁸ or in a reducing atmosphere at elevated temperatures.⁶⁹

The more expensive ITO is generally more stable than ZnO:Al in the presence of humidity and elevated temperatures. Degradation of ITO can be caused by the migration of water and alkaline species into the layer. Temperature-humidity stress of this material was further shown to cause recrystallisation and local concentrations of In and Sn.⁷⁰

Another effect that can occur in the presence of humidity is the degradation of molybdenum. This material can oxidise if directly exposed to (liquid) water and oxygen, especially under elevated temperatures. Oxidation can first lead to the formation of black and blue stains on the metallic molybdenum surface, which can contain molybdenum oxide (MoO_2/MoO_3 , potentially with sodium or selenium⁷¹). These materials can be badly conducting and/or poorly reflecting.^{72,73}



FIGURE 6 Schematic representation of a typical copper indium gallium selenide (CIGS) solar device and the degradation mechanisms that can occur in an unpackaged CIGS solar cell due to damp heat exposure and internal or external biases

This oxidation can mainly affect the scribes of monolithically interconnected devices, while it will not likely occur in the covered molybdenum back contact in the bulk of the material, due to the lack of direct water. In case of the second scribe (often referred to as 'P2'), where a Mo/ZnO:Al contact is responsible for the current transport between solar cells, increased resistance of the scribe has been observed in model systems exposed to damp heat conditions.74-77 Possible explanations are the introduction of an oxide layer at the Mo/ZnO:Al interface as well as increased resistivity of ZnO:Al in this scribe.⁶³ Moreover, oxidation of the P3 scribe was also observed, for example, on positions that have been damaged by the scribing process. As long as some conductive molybdenum is present, this is not per se a problem: If the layer is only partly degraded, the current can still laterally bypass via a nondegraded part.⁷⁷ However, in extreme cases, the molybdenum in the P3 can completely disappear, leading to the loss of connection between the cells.

2.3.2 Alkali element migration and PID

Alkali elements, in particular sodium, are highly available in CIGS solar devices as the cover glass and the substrate glass are both typically soda-lime glasses containing more than 15% Na₂O. The efficiencies of CIGS devices are known to improve by sodium doping through defect passivation at grain boundaries; however, the presence of alkali elements can be detrimental depending on their quantity and distribution within the solar stack.

Theelen et al.^{78,79} demonstrated that combined exposure to damp heat and illumination (leading to a small bias voltage over the cell) of unpackaged cells led to migration of the alkali elements sodium and to a lesser extent potassium. These elements could end in the p-n junction and ZnO:Al front contact and have a negative impact on the shunt resistance.

The migration of alkali elements can also be induced due to an external voltage stress and can lead to PID as in the case of c-Si and CdTe PV systems. When compared under the same testing conditions, it was demonstrated that CIGS thin film PV modules have higher resistance to PID than multicrystalline Si and a-Si modules.⁸⁰

PID in CIGS PV systems depends on the migration behaviour of sodium. Sodium either migrates from the substrate glass and accumulates at CIGS/CdS interface deteriorating the p-n junction or migrates to ZnO:Al front contact layer from the cover glass causing its corrosion or delamination.^{80,81} The accumulation of sodium can result in reduction in charge carrier concentration and built-in voltage, TCO corrosion and a degree of shunting, resulting in a significant drop in open circuit voltage V_{oc} and FF.⁸² A lower intensity of EL is observed at the degraded parts, which are mostly at the edge cells closer to the frame (see Figure 7).⁶⁰

Partial shading 2.3.3

The impact of partial shading strongly depends on the design of the module. Commercial CIGS modules can be divided into two classes. The first class consists of separated large cells with a current collecting grid in either series or parallel connection. Such modules may commonly experience (changing) partial shading, as they are very attractive for integration in, for examples, vehicles, textile and facades. However, this module design allows the use of bypass diodes, so the impact of partial shading can be minimised. Rigid monolithically interconnected modules form the second class. They consist of series connected long, narrow cells (e.g., 1200 mm \times 5 mm). In general, these modules maintain a very good output power when partly shaded,⁸³ especially when it comes to predictable row-to-row shading.⁷⁸ However, negative long-term effects can occur when the orientation of the cells is such that one or more cells are completely shaded, while other cells are illuminated. These modules generally do not contain bypass diodes.

For the monolithically interconnected modules, very harsh partial shading can thus present a risk, due to reverse bias exposure. An example of an undesirable situation is the use a cleaning robot, which can completely cover one or more cells while the modules are still operational. Such shadings can result in the nonreversible formation of wormlike defects. These long and winding defects have a width of tens of micrometres and can have a length of multiple centimetres (see Figure 8). In these defects, the CIGS absorber material has

(C) (B)

FIGURE 7 Optical images of copper indium gallium selenide (CIGS) solar modules (A) before and (B) after potential-induced degradation (PID) test of 1043 h - 1000 V in a chamber of $85^{\circ}C$ and 85% RH and -1000 V. Transparent conducting oxide (TCO) corrosion is shown by the blue arrows, and (C) is electroluminescence (EL) image of the state of (b)⁶⁰





FIGURE 8 A close-up image of wormlike defects in two interconnected cells of a CIGS module exposed to partial shading stress.⁸⁸

recrystallised and formed into a thick semiporous and likely conductive structure. Due to the volume expansion, a 'ridge' of elevated material^{84–86} is formed. At these positions, the ZnO:Al front contact is still intact but is lifted from its original position. The appearance of the wormlike defects leads to the formation of localised shunts in the devices, negatively affecting the module output. Although the performance loss of one wormlike defect can be minor, repeated exposure to harsh partial shades will lead to multiplication of the losses.^{87,88} Alongside to wormlike defects, also nonpermanent changes in device performance were observed due to (mild) reverse bias exposure.⁸⁹ Since these effects were often reversible, for example, for small cells under illumination,⁹⁰ they are not often studied.

Various studies have reported on design solutions for the mitigation of the impact of (large) reverse biases, especially to prevent the formation of these wormlike defects in monolithically interconnected modules.^{91,92} On the other hand, changes in the composition or thickness of the layers in the cell stack can also impact the cell behaviour under reverse bias.^{84,93-95} More information about the impact and the mitigation of partial shading can be found in the review article by Bakker et al.⁸⁸

3 | EMERGING TECHNOLOGIES

3.1 | Introduction

Emerging photovoltaic technologies aim to drastically reduce the materials cost and the energy payback time as compared with the technologies in mass production. Inorganic materials are being replaced by organic materials where possible and roll-to-roll fabrication techniques are being adopted to fabricate large area PV modules at low cost. Arguably, the use of organic materials induces opportunities for tuning of functional properties, but it also introduces numerous degradation and scale-up issues. Among the wide range of emerging PV technologies, three main classes can be distinguished: DSC, OPVs and PSCs. Among the emerging PV, PSCs outperform in terms of performance and scientific activity, in spite of being a new entry in this category. Thus, in this review, we will mainly focus on PSCs; however, it is important to note that the PSCs field leans heavily on the knowledge and experience built up from the DSC and OPV. Thus, the remainder of this section aims to briefly sketch the reliability status of DSC and OPV cells.⁹⁶

It is important to mention that the greenhouse gas emission estimates of the emerging technologies would improve tremendously (and proportionally) when longer lifetimes are achieved than presently possible.

3.2 | DSC

The DSC was first reported by O'Regan and Grätzel in 1991⁹⁷ and may still have a role to play in energy generation for emerging indoor applications.⁹⁸ The indoor efficiency of DSCs is very impressive (e.g., 28.9% at 1000 lux).⁹⁹ However, the AM1.5G certified performance is still only 12.25% (0.0963 cm² aperture area) and decreases down to 8.8% for a submodule (398.8 cm² device area),¹⁰⁰ as a result of absorption limitations.

3.2.1 | Device architecture

The DSC has several components; the photon absorption occurs within a dye molecule and charge separation, and collection is a result from interfacing materials.¹⁰¹ The remaining components that make up a DSC are the anode and cathode substrates, the anode and cathode electrodes, the electrolyte and the encapsulant (see Figure 9). The anode and cathode substrates can be transparent (e.g., glass, PET and PEN) or opaque (metallic). However, DSCs can be made on substrates that can be engineered to be thin and thus lightweight and flexible.¹⁰² In a conventional DSC, the anode is where light absorption and charge separation occur and thus is referred to as the photoanode. Transparent substrates are typically glass, PET or PEN. For nonmetallic substrates, a deposited TCO layer is necessary. An n-type porous metal oxide is deposited onto the conducting substrate. The metal oxide is subsequently sensitised by adsorbing the dye onto its surface. Similarly, to the anode, the cathode can use a metallic or TCO-coated transparent substrate. Here a catalyst layer is required for efficient electrolyte regeneration. The electrolyte is a redox couple dissolved in an organic or aqueous solvent. A thermoplastic hot-melt or a glass frit¹⁰³ holds the cathode and anode together. This also acts as the electrode spacer to prevent electrode short-circuiting. There are a significant number of up-to-date reviews that describe in depth the DSC (Vlachopoulos and Hagfeldt¹⁰¹ is a recent example—a list of reviews

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can also be found within). The stress factors for the DSC are essentially the same as other technologies although the added complexity is the impact of electrolyte solvent egress and electrolyte corrosiveness (see Figure 10). We next describe the impact stress factors have on the DSC.

3.2.2 | Photoanode

Dye stability is the limiting factor for the photoanode.¹⁰⁵ Water presence can lead to dye desorption by hydrolysis. Water can be present during device sealing or ingress overtime. Using hydrophobic dyes has shown promise in reducing this challenge.¹⁰⁵ Also, a promising alternative is hydrophilic dyes which function in aqueous electrolyte media,¹⁰⁶ thus eliminating the need to prevent water ingress. If UV light is permitted to enter the photoanode, photocatalytic degradation of the dye also occurs mediated by the high bandgap metal oxide.¹⁰⁷

3.2.3 | Counter electrode

For a conventional DSC, the cathode contains platinum which over time degrades either by dissolution or by redox species poisoning (e.g., I^-/I_3^- electrolytes).¹⁰⁸ Of all DSC components, the counter



FIGURE 10 All data are based on the perovskite database project. (A) Initial performance remaining at the end of the stability measurement versus total exposure time for all samples in the database, regardless of perovskite composition, cell architecture, stack sequence, perovskite composition, initial efficiency and testing condition. (B) T80 values for cells measured under 1 sun and max power point tracking (MPPT) conditions as a function of publication year. (C) Percentage of initial performance remaining at the end of the stability measurement versus total exposure time for cells measured under outdoor conditions. (D) Efficiency as a function of cell area¹⁰⁴





electrode (in a conventional photoanode configuration) is where the least research has gone into evaluating stability. Pt based counter electrodes are not economically viable. As such, there are several reported alternatives,¹⁰⁸ ranging from other metals and alloys, conducting polymers, carbon materials, transition metal compounds and hybrids. Polymer-based counter electrodes can be considered the most promising alternative because they can be low-cost, transparent and flexible, while still exhibiting equivalent or superior catalytic activity. One can argue that the counter electrode is the final piece of the

jigsaw puzzle which is a DSC, and thus, its development direction will

3.2.4 | Electrolyte

depend on the dye/electrolyte combination.

The electrolyte is composed of a solvent (organic or aqueous) and a redox shuttle. Volatile organic solvents are inherently difficult to encapsulate. Solvent egress not only degrades device performance but there are also safety (e.g., flammability) and environmental concerns. High boiling point solvents, room temperature ionic liquids, gel electrolytes or even electrolyte substitution for solid-state hole-transporting materials (HTMs) are alternatives.¹⁰⁹ However, these come at the detriment of cost, lower efficiencies or nontransparency (necessary for bifacial configurations). Aqueous-based electrolytes appear to be inevitable for DSC commercialisation because of the potential for easing the encapsulation requirements while also rendering this class of device as safe. Though efficiencies are still below 10%, progress may be rapid.^{110,111}

Traditionally, the iodide/triiodide I^-/I_3^- redox couple has been the choice for high performing DSCs. However, its corrosive nature towards Ag, Cu, Al and stainless steel imposes restrictions.¹⁰⁸ Introducing cobalt redox mediators Co (ii)/(iii) resulted in a significant increase in efficiencies (from certified approximately 11% to the current AM1.5G record of over 14%,¹¹² though uncertified). However, stability is poor and is attributed to photosensitivity.¹¹³ Promising alternatives are copper Cu (ii)/(i) redox mediators,¹⁰⁸ demonstrating superior indoor efficiencies,⁹⁹ and show promising high temperature and light soaking stability.¹¹⁴

3.2.5 | Encapsulation

Device encapsulation is achieved by the anode and cathode substrates and the adhesive material which is used to sandwich these together. The substrates prevent water ingress and also contain the electrolyte. Glass is by far the best material for both purposes. PET and PEN are permeable to water and oxygen ingress and also to the egress of volatile organic electrolyte solvents. How this affect device stability has been covered above. The plastic substrates are also unstable under UV. Metallic substrates require electrolyte corrosion resistance, raising costs.¹⁰⁸

The bonding material must maintain stable physical properties in the device's working temperature range, and withstand the pressure caused by the electrolyte volumetric thermal expansion. The thermal hot-melt is sensitive to UV exposure. The glass frit method although encouraging requires high annealing temperatures (600°C) which has the drawback of not being compatible with most flexible substrates.¹⁰³ Further lamination can be used to fully encapsulate the device to protect it from external contamination and UV exposure.¹¹⁵

3.2.6 | State of the art stability measurements

A recent review by Tiihonen et al.¹¹⁶ was very critical of stability reporting for DSCs. They conclude that the major shortcomings are the inadequate group size for statistical analysis and deficient reporting of measurement conditions. Also, frequently missing are UV intensities and humidity levels. They also point out that when attempts are made to demonstrate stability, success has only been achieved either at moderate temperatures (50 to 60°C) under light soaking or at high temperatures (80°C) but without light soaking. High temperatures combined with light soaking have always led to fast performance degradation. Outdoor ageing testing reports are also limited.¹¹⁶

Until recently, DSC research lacked well-defined protocols for determining device power conversion efficiencies,¹¹⁷ to the detriment of reproducibility. Consensus for stability testing of DSC is still lacking unlike for the perovskite and organic solar cells.¹¹⁶

In summary, no DSC device configuration has proved stability and reliability in accelerated testing to simulate outdoor conditions when all stress factors are present. However, the race is still on to develop sustainable efficient DSCs, which may in part synergistically solve some of the stability problems. Also, a portion of the DSC research community has become very critical of shortcomings in stability studies. This can be viewed as a positive sign.

3.3 | OPVs

OPVs offer the possibility of producing flexible, large area, semitransparent, coloured PV modules using low-cost solution processing methods and are attractive for many applications including Building Integrated PVs and indoor modules. From an environmental perspective, they also have the lowest embodied energy of any PV technology.^{118,119} Most modern devices are based on bulk heterojunction cells, and the current record efficiency is 18%¹²⁰ and efficiencies of up to 31% under indoor lighting conditions.¹²¹ Extending the lifetime of OPVs is vital in order to realise their feasibility for commercial applications.

In 2011, consensus standards were developed by the OPV community to provide a common framework to assess stability against, as the IEC standards were considered too harsh to provide meaningful.¹²² A series of interlaboratory studies have been conducted on the stability of OPV devices. These papers highlight the complex relationships between materials, technological steps, degradation protocols and PV properties.

The degradation of OPV solar cells is related to several stress factors which can be separated into both extrinsic and intrinsic factors. Extrinsic factors relate to the degradation which occurs when OPVs are aged in ambient conditions.¹²³⁻¹²⁵ Intrinsic factors relate to the inherent degradation of the constituent materials of an OPV, unrelated to the extrinsic stress factors applied.¹²⁶ Intrinsic degradation can be induced by the extrinsic factors, for example, light, heat, water or water vapour interpenetration, voltage bias and mechanical stress. The intrinsic factors included the metastable morphology, the diffusion of the electrodes and buffer layers into the active material: the extrinsic factors included oxygen and water infiltration, irradiation, heating and mechanical stress.^{123,127,128} There have been several review articles on OPV stability, and it is clear that degradation is also not due to a single intrinsic or extrinsic failure mechanism.¹²⁹ For example, this has been demonstrated by consideration of the combined effect of humidity and temperature on OPV degradation, leading to an interaction effect. When an OPV is stressed by both temperature and humidity, a greater degradation is observed than when each factor is increased individually.¹³⁰ Furthermore, by not considering the interaction effects in other reports, misleading conclusions can be reached due to the significant impact of the interactions on the degradation.¹³¹ The effects of applying multiple stress factors on OPV modules simultaneously using a design of experiment approach were performed to demonstrate predictive ageing of OPVs based on multistress testing using a log-linear life model.¹³² Table 1 gives common intrinsic and extrinsic stress factors considered during OPV stability studies.

There has been a large body of research aimed at improving the stability; this has focused on active material design, device engineering of the active layers, employing an inverted architecture, transport layer optimisation, electrodes and encapsulation optimisation. However, from such a review, ranking of the different intrinsic and extrinsic factors is difficult in terms of severity; machine learning, however, presents a possible methodology for considering such literature sources and using analytical techniques to quantify and rank the significance of each factor.¹³³⁻¹³⁵ Nevertheless, there are a number of examples where high stabilities have been reported. Nonfullerene acceptors (NFA), IDTBR and IDFBR were combined with both a scalable and affordable donor polymer, PBDTTT-EFT (PCE10), and devices were found to be highly efficient owing to changes in the microstructure which reduces charge recombination and increases photovoltage.¹³⁶ Recent results published in Du et al.¹³⁷ show that the lifetime of OPVs based on NFAs can remain within 80% of the initial power conversion efficiency (PCE) after 11,000 h under 1 Sun illumination. In this work, it was stated that the photostability is strongly dependent on the end-groups and side-chains of the NFAs, and the side-chain modification can significantly improve the morphological stability. Nevertheless, the results were obtained in the samples with only 10.4 mm² active area. Despite these highly encouraging recent results, the next challenge is to scale this efficiency and stability to larger area modules.

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One noticeable trait in the OPV community is that outdoor monitoring has been a sparingly adopted approach for testing the stability of OPVs. By testing OPVs in outdoor conditions, multiple stress factors can be applied, and outdoor testing remains one of the best approaches to review OPV stability as OPVs are subjected to multiple stress factors^{138,139} including tests which are not conducted in ISOS consensus standards such as the impact of condensation.¹⁴⁰ One of earliest reports on outdoor testing of OPVs was conducted by Katz et al. in 2007.¹⁴¹ A number of outdoor stability studies were led by the Danish Technical University, and small molecule outdoors stability has been conducted by Josey et al.^{142,143} One of the most significant was the report on a solar park based on polymer solar cells, which investigated the practicality of assembly, the installation, operation and end of life. The analysis showed that a high voltage installation, where solar cells are all printed in series, enabled an installation rate that far exceeds any other PV technology which existed at the time. The installation and deinstallation rates were estimated at 100 m min⁻¹, which exceeds the total manufacturing speed of the polymer solar foil. Using the methods presented, simultaneous installation and deinstallation were possible, providing efficient schemes for decommissioning and recycling. New research directions are discussed by Krebs et al. whereby new and advanced materials must be developed with the potential for large-scale application in solar parks, fast roll-to-roll processing using only abundant materials and finally, the use of flexible substrates using low-cost barriers and adhesives. Finally, Krebs et al. noted how this research field could be directed by life cycle assessment (LCA) and that a short energy payback time could be obtained by choosing a wooden structure (0.32 days), rollbased installation and high voltage connections, although the potential exists to reduce to approximately 1 day with mass

3.4 | PSCs: State of the art

manufacturing.144

Organic-inorganic metal halide perovskites have emerged as a new class of semiconductors to fabricate low-cost efficient PV modules. The term 'organic-inorganic metal halide perovskites' is used to describe a group of compounds, which has a structure similar to CaTiO₃ crystal and is represented by ABX₃, (A is an organic alkyl ammonium cation; B is a metal, for example, Pb and Sn; and X is halide anion).¹⁴⁵ Perovskites are relatively new to the PV community and entered in 2009 as a promising material.¹⁴⁶ Since the initial reports on PSCs with PCE of 3.8%¹⁴⁶ in a dye solar cell, the PCE has witnessed rapid increase. The current certified record PCE is 25.5% for single junction, and >29% is for tandem (perovskite over Silicon) solar cells.¹⁴⁷⁻¹⁴⁹ Perovskites possess extraordinary intrinsic optoelectronics properties such as broad absorption spectrum, high absorption coefficient enabling low binding energy, long charge carrier diffusion length and long carrier separation lifetime, which makes them promising materials for PV. Moreover, they offer flexibility, semitransparency

and lightweight and are easy to synthesise. The precursor materials for the synthesis of perovskites such as methyl ammonium (MA) or formamidinium (FA), lead halides, are low cost, and their processing is relatively simple. Despite the success in research, most of the achievements result from laboratory studies and the higher performance still needs to be validated for commercial PV. Particularly by addressing the reliability challenge, it has the merits to surpass the silicon baseline. There is also an appealing industrial rationale for tandem cells, either together with silicon or CIGS, or in the form of a perovskiteperovskite tandem. The operational stability of the first PSCs¹⁴⁶ was in the order of minutes rather than hours, and this has improved from minutes to days, to weeks, when the liquid electrolyte was replaced with a solid-state hole conductor, compositional engineering of perovskites,^{150,151} passivation of perovskites and rational charge selective layers. Given these developments, it is very unlikely that PSC will soon achieve a 25 year life time warranty which is common for commercial silicon PV modules

Common triggers for failure include heat, moisture and UV light (discussed in the next section) and compositional dynamics, such as ion migration, defect accumulation and phase instabilities of perovskites with mixed compositions. Such factors cause irreversible degradation and should be taken in to account during the measurements. Currently, competitive shelf life of PSCs >1000 h of max power point tracking (MPPT) has been reported. The PSCs Database Project¹⁰⁴ contains most of the PSCs data available in the literature (around 15,000 papers) and aims to collect all future device data in one place. So far, it is the most comprehensive data source available (Figure 10A). The performance of devices at the end of the stability measurement is plotted as a function of total exposure time for all the samples in the database, regardless of perovskite composition, cell architecture, stack sequence, perovskite composition, initial efficiency and testing condition. It can be deduced from Figure 10 that a majority of devices degrades rapidly, and only handful of devices can go >1500 h.¹⁵²

One of the most relevant testing conditions is MPPT under 1 sun illumination, as it mimics real operating conditions. By plotting the T_{80} , that is, the time the devices have lost 20% of its initial power output, against publication year for PSCs tested under 1 sun with MPPT (Figure 10B), substantial progress can be noted. A subsequent analysis suggests that some of the PSCs with high T_{80} values also boast a high initial PCE. Arguably, a comparison of the initial performance against the remaining life after 1000 h of MPPT under 1 sun illumination provides useful insight. In some cases, the poor initial PCE is not worthy for an elaborate understanding of the stability issues. To address such limitations, we plotted (Figure 10C) the initial performance against the remaining performance after 1000 h of MPPT. We noted a cluster of points in the upper right corner representing efficient devices with PCE ${\sim}20\%$, and this functions efficiently even after 1000 h. It is worth mentioning that most of the available data are on small area cells. Similar to other PV technologies, the efficiency tends to decrease when the area goes up due to local defects and material nonuniformity (Figure 10D). Technology learning cycles are necessary to decrease the efficiency gap between small and larger areas.

3.5 | PSC: Failure modes and their mitigation

Before discussing about the failure modes of PSC, it is vital to know the structure of PSCs, as this has a bearing on the degradation routes. Typical PSCs consist of at least of five layers with four interfaces (see Figure 11). This comprises of TCO-coated glass substrate as anode (indium or fluorine doped tin oxide [ITO, FTO]), perovskite absorber layer, an electron selective *n*-type layer (e.g., SnO₂, TiO₂ and PCBM) and hole selective *p*-type layer (e.g., Spiro-OMeTAD, CuSCN and PTAA) and cathode (Au, Cu or Ag). We can subdivide PSCs into five classes depending on the placement and nature of the chargetransporting layer, namely, the planar n-*i*-p structure, planar p-*i*-n structure, the mesoscopic n-*i*-p structure, mesoscopic p-*i*-n structure and the triple mesoscopic structure (Figure 11).

The absorber and charge-transporting materials can degrade either alone or while in contact with other layers. In the case of the absorber layer, the volatile nature of the organic cation, halide segregation and ion accumulation are undesirable processes that speed up the device degradation. Ions migration at different interfaces induce different types of recombination losses in the PSCs. Recently, PSCs using mixed-halide.¹⁵⁴ mixed-cation¹⁵⁵-based perovskites and lavered perovskites gave competitive device performance and stability. To further increase the stability and performance, doping of perovskite or interfacial laver was adopted.^{156,157} Interfacial modification was made, and new charge transport materials such as BaSnO₃,¹⁵⁸ CuGaO₂,¹⁵⁹ and PTAA¹⁶⁰ were explored in *n*-*i*-*p* and *p*-*i*-*n* configuration. Several stress factors have been used to identify the failure modes in these PSCs; however, their effect on the device stability is not fully quantified by IEC standards. In the below section, the possible stress factor will be discussed along with the known degradation mechanism and possible mitigation.

3.5.1 | Main degradation/stress factors

Humidity is one of the main stress factors, owing to the moisture sensitive nature of organic cation present in hybrid perovskite. The perovskite crystals can be hydrated when in contact with the humid air; however, this hydration process is reversible. Due to poor thermal stability of hydrated perovskite, it can decompose irreversibly rapidly and or in the presence of light. This suggests that investigation of combined stress factors, such as light, temperature and humidity, is paramount, experimentally and complemented by modelling to understand the significance of IEC damp heat tests (85°C/85% RH) to predict 25 years' operational stability for PSCs modules.

Atmosphere composition (moisture and oxygen)

Perovskite layers are liable to degradation under exposure to moisture and air; to note here moisture, oxygen and UV radiation are indispensable for the degradation process. The mechanism of $CH_3NH_3PbI_3$ degradation in the presence of H_2O is as follows:

$$CH_3NH_3PbI_3 + H_2O \leftarrow \rightarrow CH_3NH_3PbI_3 \cdot H_2O$$

FIGURE 11 Annotated diagram of a perovskite solar cell (PSC) with four device configurations: Mesoscopic structure, planar structure, triple mesoscopic structure and tandem structure with lowerbandgap subcell. In mesoporous structure, a thin mesoporous scaffold (typically TiO₂ or Al₂O₃) infiltrated with absorber material is present between a charge extraction layer and the polycrystalline absorber layer¹⁵³



 $4CH_3NH_3PbI_3\cdot H_2O \longleftrightarrow CH_3NH_3)_4PbI_6\cdot 2H_2O + 3PbI_2 + 2H_2O$

 $CH_3NH_3)_4PbI_6\cdot 2H_2O \rightarrow 4\,CH_3NH_3I + PbI_2 + 2H_2O$

$$CH_3NH_3I \leftarrow \frac{H_2}{2} \rightarrow CH_3NH_2 + HI$$

The equilibrium of these reactions leads the co-existence of the salt with the Pbl₂, CH_3NH_3I , CH_3NH_2 and HI. The HI can be decomposed further by the following reactions.

$$4HI + O_2 \longrightarrow 2H_2O + 2I_2 \text{ (in the presence of } O_2\text{)}$$

$$2HI \longrightarrow H_2 + I_2$$
 (photochemical)

Consuming the HI driving the whole decomposition process forward.

Also, the salt is decomposed at elevated temperature by dissociation according to the reaction.

 $CH_3NH_3Pbl_3 - \rightarrow CH_3NH_2\uparrow + HI\uparrow + Pbl_2$.¹⁶¹

Similar mechanisms occur in FAPbI₃. MA, FA and HI are volatile at elevated temperatures. The bromide (Br) anion-based perovskites are relatively stable but follow the similar degradation mechanism.

Solar visible and UV illumination

PSCs also suffer from photo-induced degradation, and its origin is relatively not well understood and this also related to a number of failure modes. The mechanism of UV degradation is distinctive under different environments. Although the degradation under illumination at low temperatures in perovskite has shown to be insignificant,^{162,163} it increases significantly in the presence of H_2O and O_2 or in contact with other materials. When PSCs are exposed to light in the presence of oxygen only, the photo-generated electrons react with the O_2 to form superoxide (O2⁻).¹⁶⁴ This superoxide oxidises the perovskite to Pbl_2 , I_2 and $CH_3NH_2^-$ (Figure 12). The I_2 further oxidises the perovskite. Another unique aspect is that in the absence of H_2O and O_2 , the degradation of PSCs by UV radiation is partly reversible under 1 sun illumination.¹⁶⁵ The UV degradation also impacts other layers in the device.¹⁶⁶ Specifically, the most common electron-transport layer, TiO₂, is a typical photo-catalyst for oxidising organic materials¹⁶⁷ with a bandgap of 3.20 eV (~400 nm wavelength). It photocatalyzes the decomposition of hybrid perovskite at their interface.¹⁶¹ The degradation mechanism in the interface of perovskite/TiO₂ consists of two stages¹⁶⁸ (see again Figure 11). Moreover, charge generation under light illumination and subsequent trapping on the surface of perovskite has been shown to initiate the moisture-induced irreversible degradation to PbI₂ CH₃NH₂ and HI vapours.¹⁶⁹ The exact mechanism is still obscure, though it is suggested that organic cation could become loosely bound to PbI_6^{4-} octahedra after light exposure.¹⁷⁰

TiO₂ itself is susceptible to degradation under UV and compromises the durability of PSCs.¹⁷¹ Increased UV stability with the addition of an additional interlayer (Al₂O₃, Sb₂S₃, MgO and CsBr) at the perovskite/TiO₂ interface¹⁶² or replacement of TiO₂ layer with other material was reported.¹⁷² The classical HTMs in *n-i-p* structure is Spiro-OMeTAD, and it can suffer light-induced oxidation alone or in the presence of perovskite.¹⁷³ Another reason for the photo-induced degradation in PSCs is the deterioration of the chemical bonding between HTM and Au at the interface, causing insufficient hole



1st Stage in the presence of O₂ CH₃NH₃Pbl₃ → CH₃NH₃Pbl₃*

The CH₃NH₃Pbl₃ carries photoinduced electrons and holes. It transfers the e⁻ to O₂to give superoxide O₂⁻.

1st Stage in the presence of TiO₂ When TiO₂ is exposed to UV-light, oxygen

vacancy-Ti³⁺ transform into active Ti⁴ * trap states

2^{nd} Stage in the presence of O₂ The superoxide O₂ oxidize the perovskite to Pbl₂ CH₃NH₂ and I₂. The I₂ keep

2nd Stage in the presence of TiO₂

The Ti⁴⁺ extract electrons from the I⁻ ions converting into Ti³⁺ and oxidizing I⁻, to I₃⁻ which deconstructs the perovskite crystal.

FIGURE 12 The mechanism of perovskite solar cell (PSC) degradation under continuous ultraviolet (UV) radiation in the presence of O₂ and/or TiO₂

extraction.¹⁷⁴ Subsequently, the resistance and carrier recombination increase resulting in degraded PV performance. Formamidinium lead iodide (FAPbI₃)-based PSCs exhibit better photostability than of MAPbI₃¹⁷⁵ and are now being explored.¹⁷⁶

Temperature

For possible commercialisation of PSCs, long-term stability at 85°C (representing an elevated temperature on a roof on a hot summer day) is necessary in order to compete with different PV technologies. Decomposition can occur during the fabrication process, during the annealing of the perovskite (>100°C). Classical MAPbl₃-based PSCs are stable to temperatures up to 60°C, and at temperatures >80°C, the degradation is rapid and irreversible¹⁷⁷ due to its phase transformation. Deterioration occurs mainly in the bulk of perovskite,¹⁷⁸ and also, thermal-induced deterioration at the interfaces and in the charge selective layers (HTL or ETL)⁴⁹ can take place. Different pathways were reported for thermal degradation at variable temperatures.

MAPbl₃ undergoes a tetragonal-to-cubic phase transition at a relatively modest temperature of approximately 56°C,.¹⁷⁹ The impact of this phase transition on the device performance and long-term stability is not well understood. The thermal degradation of CH₃NH₃Pbl₃ can occur even at lower temperatures (80°C) under an inert atmosphere if exposed for extended time (>60 min).¹⁸⁰ The decomposition reaction is¹⁸¹:

 $CH_3NH_3PbI_3 \rightarrow NH_3 + CH_3I + PbI_2$

At high temperatures (>350°C), the decomposition of $CH_3NH_3PbI_3$ also involves a small amount of the methane (CH_4) formation.¹⁸² The formamidinium-based perovskites are more stable at low temperatures. At low temperatures, the reversible reactions a and b occur, but at temperature >95°C, it decomposes irreversible according to the reaction c, Scheme 1.¹⁸³

In reports, it is also suggested that at high-temperature gold diffusion can occur from the electrode through the HTL to the perovskite layer and deteriorates the performances of PSCs,¹⁸⁴ while >80°C large voids are created in the Spiro-OMeTAD layer¹⁸⁵ and all these compromise reliability.

Mechanical and electrical loads

PSCs exhibit poor resistance to fracture and are considered extremely fragile in the presence of applied loads.¹⁸⁶ The mechanical stability of the PSCs is strongly depended by the architecture used and the other ancillary layers (HTL, ETL, electrodes, etc). Experiments showed two points of failure for PSCs¹⁸⁷: firstly, the adhesion of charge-transporting materials to the perovskite layer and secondly, the cohesive failure of these auxiliary charge-transporting materials. The perovskite layer itself does not exhibit significant resistance to fracture due to the brittle, salt-like crystal structure. Mechanical stability can be improved by choosing the appropriate architecture, materials and preparation methods, though flexible devices capable of withstanding hundreds of bending cycles¹⁸⁸ were reported.

Charge extraction layers and electrode

The most common electron transport materials in n-i-p structure are based on metal oxides such as TiO₂, ZnO and SnO₂. The TiO₂ is the most extensively studied but shows degradation under UV irradiation, similar to the typical HTM (Spiro-OMeTAD). Additionally, HTL layer produces pinholes during spin coating, and through these pin holes, H₂O and O₂ can permeate and decompose the MA- or FA-based perovskite. Meanwhile, mobile ions also migrate from the perovskites and degrade the HTL; see Figure 13.

Further, in *n-i-p* structure, commonly used metal electrode is gold while for *p-i-n*, silver, copper and aluminium are being used. These electrodes can be corroded by I_2 or I^- that migrates from the perovskite layer through the HTL. The migration of ions towards the









FIGURE 13 Mechanism of perovskite solar cells (PSCs) degradation through the pin holes of Spiro-OMeTAD

electrode side is boosted by the presence of humidity due to partial hydrolysis of perovskite. The silver electrodes although are costeffective as compared with gold can cause a fast degradation as silver can react with perovskite to form AgX (X = Cl, Br or I depending on the perovskite used), causing short circuits and degradation of PSCs¹⁸⁹ apart from Ag oxidation. Similarly, Al reacts with the I₂ to give AlI₃. Though Au shows high corrosion resistance to iodine, it could also be corroded in the presence of I₂ and I⁻ to form AuI₂ and AuI₃.¹⁹⁰

4 | LESSON LEARNED FROM MATURE AND EMERGING PV TECHNOLOGIES

One of the aims of this review article is to identify lessons learnt from mature technologies in order to aid the development of next generation PV technologies. It is clear for the commercial viability of any PV technology that reliability is key and it should not be separated from upscaling and other product development aspects. Therefore, also for lab-scale PV technologies, it is important to explore how reliability can be optimised already under laboratory conditions. In the following, we will address the long-term stability of third generation PV technologies, OPV, DSC and PSC. For OPV and DSC, power conversion efficiencies of resp. >18% and 14% have been reported for lab-scale devices. In the case of liquid electrolyte-based DSC, the limitation also arises from the series resistance of the DSC components, which limits the efficiency. Thus, both electron and hole transportation should be improved to decrease the series resistance and increase FF. Theoretically, achievable efficiencies in solid-state DSC are higher and the configuration in solidstate suggests improved stability.

The advent of perovskite-based solar cells that stems from solidstate DSCs and its power conversion efficiencies has surged at a rapid pace. However, by evaluating reliability issues, it is not clear whether this technology will be able to reach to 25 + years of operational stability required for commercial relevance. In the last decade, operational stabilities of such devices rise from minutes to months and if that development continues, perovskite-based photovoltaics may soon become a contender in the field. Nevertheless, mature technologies such as silicon never had such inherent instabilities under exposure of light, so it is possible that tens of years of lifetime is not achievable. The recent developments in the device architecture, compositional engineering, rational charge selective materials, additives and watertight encapsulation can pave way for production with long lifetime. Besides the fabrication of laboratory-sized cells, ageing tests should be performed on mini-modules and modules level and reporting of data should be made under actual working conditions to identify the failure mechanism for possible mitigation steps. The maturation of this technology is reflected by the fact the community has devised standard testing protocols. In this direction, a close collaboration of interlaboratory device testing will be of significant importance to understand and resolve the degradation mechanism.

For many new technologies, the module components will be similar to those for mainstream technologies, such as the encapsulation and interconnect (e.g., TCO) materials. Therefore, the understanding of these aspects of module design and testing can be readily applied to new solar cell materials. However, the sources of instability for new PV technologies are numerous and include materials intrinsic properties (e.g., absorber layer and electrode) and extrinsic

environmental conditions (humidity, light, temperature, oxygen/water vapour and thermal changes), among others. Like in the early stages of c-Si, CdTe and CIGS technology development, most research on emerging technologies focuses on advancements of the cell technology. However, many degradation and failure modes are related to other module components and the not solar cells. In particular, the polymeric materials used as encapsulants and backsheets can play a significant role in cell and module degradation. Either low molecular degradation products can interact with other module components; for example, EVA generates acetic acid which among other effects corrodes cells and metallisation. Because they are permeable, the polymers also determine the availability of water vapour and oxygen in PV modules. Thus, the BOM of any emerging technology should be carefully selected, considering permeation properties and possible incompatibilities.

Aside from module packaging, which is similar between c-Si and thin film, the mature thin film technologies (CdTe and CIGS) provide more lessons for emerging technologies. Any cell degradation modes commonly found in thin film cells (such as pinholes, reverse bias, shunting, TCO corrosion) would be the main topics of concern for an emerging thin film PV technology.

Understanding why failures happen is key for improvement in reliability. A detailed understanding of the various failure modes occurring during in-field operation of the solar cells is key to minimising or eliminating performance losses. Failure modes and effects analysis (FMEA) should be used and widely applied to reliability growth protocols. An objective approach to improvement is required, where researchers apply strong test programmes with measurements of all failure modes which can be used to assess actual failure obtained from accelerated and outdoor testing. Key to this will be the material/root cause failure analysis that in some cases needs operando techniques to decipher the kinetics.

Together with field tests, accelerated lifetime (ALT) tests are of fundamental importance to reduce the time to market for a new PV technology. Ideally, this requires through understanding and verification that ALT testing indeed reproduces and amplifies only the failure modes observed under real operational conditions. While indoor stability studies are more prevalent in the literature, outdoor tests provide an opportunity to understand materials and device degradation under field conditions. Furthermore, outdoor studies from the whole PV community show that failure modes are either reduced or increased in severity under outdoor conditions and that different failure modes are observed outdoors which are not observed from indoor ALT tests.

The case of organic and dye-sensitised solar cells has highlighted that standards issued by the International Electrotechnical Commission can overstress the devices leading to failure modes that do not necessarily occur in the field. Therefore, specific tests are required to properly estimate the potential failure rates in emerging PV technologies. Most studies on perovskite stability to date have been focused on considering one or maximum two stresses and/or the alteration of one or two layers. Although a stability study under one applied stress can provide meaningful information, it does not provide information about the likely outdoor stability during which multiple stresses are simultaneously applied. Multistress is a good methodology to ensure that acceleration factors remain high, without overstressing the device. Other PV technologies can no longer rely on just the main module qualification standards (IEC 61215, IEC 61730). It is worth pointing out that it is possible to modify, expand or add tests to IEC standards to address new failure modes.

Standard high temp/humidity/light accelerated life testing (ALT) simply does not stimulate the types of complex material systems found in third generation technologies with organic layers, hybrid barrier layers, sealing layers and so on. One of the impediments for undertaking both outdoor testing and ALT of new PV materials has been the issue of scalability. Specifically, transferring knowledge gained about performance from tests on a small device or single material or component to a complete system (module). For example, metaanalysis by the authors shows that from 500 research papers on perovskites published between 2017 and 2019, the vast majority of devices reported on were small-sized laboratory-scale devices with >94% having an active area of 0.2cm² or less. There is no straightforward means of linking performance of small-scale devices to those full-sized modules, and as the devices get larger, the higher sheet resistivity of the transparent electrode (typically ITO or FTO) leads to increased series resistance. With the scaling of devices, quality control becomes more important and fundamental changes to processing factors such as solvents, solution formulations, material selection and device design have to be made to accommodate; all of these are likely to influence the stability of the final product and limit the usefulness of small-scale stability tests. On the contrary, on scaling from cells to modules, edge effects become less significant.

Certain stress factors are not well studied in emerging PV, such as soiling, chemical pollutants and, to some extent, mechanical loading. The latter is particularly interesting; next generation PVs modules are often on flexible substrates so the types of mechanical stress are different to one would expect from the module in Figure 5. While it is commonplace to see tests on repeated bending, prolonged flexing, under load such as wind might yield different failure mechanism. Indeed, the mounting format is likely to play a role in the mechanical stability. Longer-term soiling and chemical pollution might have an influence. The mismatch in thermal expansion coefficients and low fracture energy of layers in PSCs raise a concern as to whether devices can withstand mechanical stresses from temperature fluctuations. Large mismatches in CTE between adjacent materials could build up stress and lead to delamination during temperature cycling, which presents a direct path for moisture ingress to the solar cells. In addition, the metal oxide barrier layers used with flexible substrates might degrade under sustained chemical pollutant exposure.

Encapsulants need to be chosen carefully to be optically transparent, flexible enough to absorb any fluctuation in strain energy during temperature cycling, electrically insulating to mitigate PID, to have a reasonably low water vapour transmission rate and to not release byproducts that would be harmful to the electrical contacts and solar cell absorber (i.e., acetic acid and EVA). One specific challenge faced with next generation modules is the high voltage when large numbers of cells are connected in series, leading potentially to PID issues or arcing. As an example, a 2.1 m long PSC module with 1 cm wide serially connected cells would have an open circuit voltage of around 184 V,¹⁹¹ compared with around 46 V for an example same sized c-Si module. Clearly ensuring these higher operating voltages do not have an impact if the substrates are thin films is a vital research area. Another critical aspect is shear forces that build up in molten or liquid encapsulants during module production, which eventually could break the absorber materials.

Stability of the encapsulants and edge sealants (if applicable) is required to minimise this. There is very little work in next generation PVs on edge sealants, and this is also an area that needs more research to be done one them.

It is clear that step changes in improved encapsulation and module packaging are required for next generation technologies, simply by considering their internal stability issues. Many encapsulation strategies in literature are at low TRLs or too expensive for scalable use. Moving production to scale will add quality-engineering issues that are presently unknown; the EVA issues stipulated in Section 2.2 show how a stable material can give problems as companies move to mass manufacture. Clearly as the drive to low cost, mass manufacture starts for emerging technologies; issues with packaging will become more commonplace.

5 | CONCLUSION

The aim of the review paper was to describe technological specific degradation modes of the different PV technologies. The paper not just introduced degradation modes found in mature PV technologies (c-Si, CdTE and CIGS) but also provided a review of known failure modes and areas of future research for emerging technologies such as DSC, OPV and PSCs. The review paper discussed how known failure mechanisms and developmental issues that were discovered in mature and commercially available PV technologies might impact emerging PV technologies and if learning lessons from mature technologies might speed up development of these emerging technologies.

Degradation modes typical for c-Si PV are cell cracks, snail trails and hotspots as well as PID, LID and LETID. PID is also found in CdTe and CIGS modules. Especially CIGS modules are also sensitive to partial shading and water ingress. Partial shading can result in the nonreversible formation of wormlike defects. In these defects, the CIGS absorber material has recrystallised and formed into a thick semiporous and likely conductive structure. Water ingress decreases the conductivity of TCO front contact.

The review made it clear that the long-term stability of third generation PV technologies (OPV, DSC and PSC) remains a challenge and needs to be addressed for achieving rapid commercialisation. These technologies replace inorganic materials by organic materials where possible and adopt roll-to-roll fabrication techniques. However, even though the use of organic materials induces opportunities for tuning of functional properties, it also introduces numerous degradation and scale-up issues. Firstly, unlike the mature solar cell technologies, the organic absorber components are prone to oxidation itself. Additionally, material compatibility and unintended side reactions of electrode, absorber and interlayer materials are a big challenge for emerging cell technologies.

Like in the early stages of c-Si, CdTe and CIGS technology development, most research on emerging technologies focuses on advancements of the cell technology. However, many degradation and failure modes are related to other module components and not the solar cells. In particular, the module packaging can play a significant role in cell and module degradation, so the BOM of any emerging technology should be carefully selected, considering permeation properties and possible incompatibilities.

Aside from module packaging, which is similar between c-Si and thin film, the mature thin film technologies (CdTe and CIGS) most probably will provide more lessons for emerging technologies. Any cell degradation modes commonly found in thin film cells (such as pinholes, reverse bias, shunting and TCO corrosion) would be the main topics of concern for an emerging thin film PV technology.

Understanding why failures happen is key for improvement in reliability. A detailed understanding of the various failure modes occurring during in-field operation of the solar cells is key to minimising or eliminating performance losses. One of the impediments for understanding the long-term behaviour of emerging solar cell technologies has been the issue of scalability. The vast majority of devices reported on were small-sized laboratory-scale devices with >94% having an active area of 0.2cm² or less. There is no straightforward means of linking performance of small-scale devices to those full-sized modules, especially as edge effects become less significant. Also, certain stress factors are not well studied in emerging PV, such as soiling, chemical pollutants and to some extent mechanical loading. The latter is particularly interesting as next generation PVs modules are often on flexible substrates, and there are different mechanical stress distributions to be expected.

Summarised, it is clear for the commercial viability of any PV technology that reliability is key and it should not be separated from upscaling and other product development aspects. Therefore, especially for emerging lab-scale PV technologies, it is important to explore how reliability can be optimised already under laboratory conditions.

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