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Encapsulating perovskite solar cells for long-term stability and prevention of lead toxicity

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ABSTRACT

Lead halide perovskite solar cells (PSCs) have achieved remarkable efficiencies comparable to those of their established silicon counterparts at a very fast pace. Moreover, solution-processable facile technologies offer low-cost, low-temperature, scalable fabrication of these solar cells. Numerous studies have focused on improving the performance, stability, and processing of PSCs. However, potential lead toxicity and poor long-term stability impede their commercialization. In recent years, several studies have developed novel encapsulants for PSCs that can simultaneously improve stability and impede potential lead leakage. Although improvements have been made on both fronts, no solution to date could achieve a level of stability and leakage prevention that could result in a market breakthrough. Here, we analyze PSC encapsulation and lead leakage prevention techniques undertaken in recent years. While most of the related studies focused on improving either stability or toxicity, we note that both can be solved together with a suitable encapsulant that is impermeable to both moisture and Pb²⁺ ions. In addition, the lack of a unified standard stability testing protocol has led to stability testing under a variety of temperatures, humidities, and environmental conditions. Therefore, the urgency for a standard protocol for stability and lead toxicity testing cannot be overlooked.

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INTRODUCTION

The demand for clean energy is on the rise every year, and solar cells provide more green energy than any other suitable large-scale energy source.^{1–3} Unlike most other renewable energy sources, solar cells are capable of meeting current energy demands.^{4–7} The current solar energy market is dominated by silicon PV, which is a mature technology. Si solar cells have almost reached saturation in terms of energy production and durability.^{8,9} However, this technology suffers from excessive material usage, high energy costs, and high-temperature processing, resulting in a longer payback period for this class of PV.^{10–12} Researchers across the world are looking for alternative PV technologies to meet future energy demands, either side by side or in tandem with Si solar cells.^{13,14} Perovskite solar cells (PSCs) are the forerunners to potentially gain a large share of the future PV market owing to their superior power conversion efficiencies (PCE), facile low-cost processing schemes, suitability in a wide range of

applications, and low material usage.^{15–20} PSCs have made the fastest progress among all PV technologies due to their bandgap tunability, long carrier lifetime, high carrier mobility, and high absorbance over a wide range of solar spectra.^{17,18,21–23} These fitting properties enabled researchers to obtain record efficiencies in both single junction and tandem applications.^{19,24–26} Most importantly, bandgap tunability of PSCs has opened areas for tandem cells with almost any other PV, resulting in perovskite–silicon, perovskite–organic, perovskite–CIGS, and perovskite–perovskite tandem structures.^{24,26–28} Moreover, additive engineering is still being studied quite extensively in PSCs to yield better-performing devices that might exceed the PCE for silicon PV at the module level.^{29–31}

Despite such promising prospects, the market breakthrough of PSCs is impeded by two major challenges: poor long-term stability and potential lead toxicity.^{32–34} Stability has been a long-lasting problem for PSCs, which primarily arises from the decomposition of the perovskite film into its precursors.^{35,36} In addition, perovskite films are also degraded by moisture, oxygen, light, heat, mechanical stress, and intrinsic factors.^{37–43} The absence of a standard testing protocol has resulted in different nonstandard protocols being used in recent studies. For any PV technology to commercialize, three boxes need to be ticked, often referred to as the golden triangle for any PV as demonstrated in Fig. 1(a).^{44,45} Currently, PSCs have achieved two of these three goals, except for their lifetime. Figure 1(b) shows how PSCs compare with silicon PV in terms of cost, efficiency, and lifetime. Although they already outperform Si PV in terms of cost and efficiency, the lifetime is nowhere near that of already matured silicon PV technology.^{46–51} Therefore, progress in this area is crucial for further development of perovskite PV.

Moreover, perovskite material is made of around 50% w/w lead (Pb), posing a threat of lead toxicity if these Pb²⁺ ions are leaked out. This is particularly concerning when the modules are made for large-scale energy production. However, through proper encapsulation, lead leakage can be largely prevented.^{33,34} In order to do so, an extra lead-absorbing film needs to be added to the regular encapsulation and packaging materials.³⁴ This film will be responsible for capturing the

lead ions in case the modules have a crack or total breakage. Ideally, this needs to be a polymer or resin with a moderate Young's modulus, preventing a creak or break.

Owing to the fast progress in this field, it is tough to keep up to date with state-of-the-art research and developments. Moreover, the variation of stability testing reported in numerous studies recently makes it harder to follow and select the best method to analyze and further develop PSCs. In addition, the degradation mechanisms of fully inorganic perovskites are different, which calls for encapsulation methods uniquely tailored for those. Also, there are several recent studies, mostly with recently developed materials on lead capture of PSCs when the covering glass breaks. Therefore, a comprehensive review of the recent studies on encapsulation of PSCs with excellent results on stability, efficiency, lead capture, or sustainability will certainly help to understand the field and its current level of progress. Furthermore, it will help to understand how a suitable encapsulation can stabilize PSCs without having a major effect on their PCE, side by side inhibiting the possibility of lead leakage in extreme environmental conditions. Remarkable progress in recent years may encourage researchers to focus on developing more suitable materials to encapsulate PSCs.

FACTORS AFFECTING THE STABILITY OF PSCS

PSCs are degraded by multiple factors, both intrinsic and extrinsic.^{52–55} Most of the encapsulants reported thus far provide protection only against extrinsic factors, such as moisture and oxygen. However, they cannot protect against internal degradation by ion migration and material degradation into their precursors.^{43,56–58} The degradation mechanism of PSCs are discussed below in relation to encapsulation strategies that might inhibit those processes.

Moisture

Continuous exposure to ambient humidity can rapidly degrade PSCs. The organic cations (methylammonium iodide and formamidinium iodide) are very hygroscopic and can easily breakdown into their precursors.^{52,59} Water molecules form weak hydrogen bonds with these organic molecules, forming a hydrated perovskite phase.^{60,61}

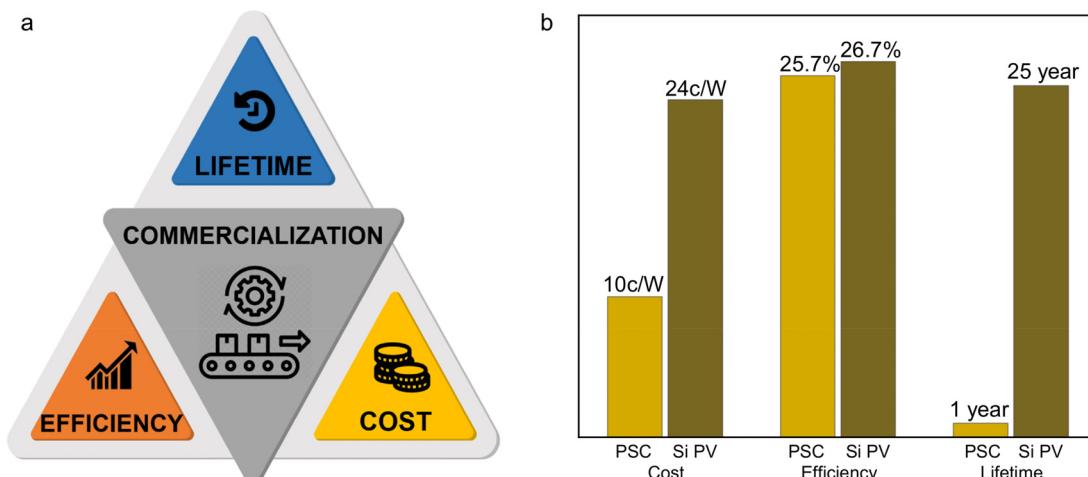
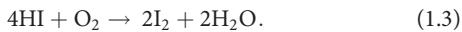


FIG. 1. (a) Illustration of the Golden Triangle for any PV leading to commercialization. Reproduced with permission from Raman *et al.*, Renewable Sustainable Energy Rev. 151, 111608 (2021). Copyright 2021 Elsevier B.V.⁴⁴ (b) Comparison of the three aspects of the golden triangle between PSCs and Si PV.

This in turn weakens the structural bond of the perovskite, which is reversible. However, continuous exposure to water molecules causes an irreversible chain reaction [Eqs. (1.1)–(1.3)], which severely degrades the perovskite film.⁶²



Without encapsulation, PSCs cannot withstand humid conditions for too long and are unable to pass the damp heat test, which involves 85% relative humidity (RH).^{63,64} Zheng *et al.* studied the effect of humidity levels on the continuous performance of PSCs and observed rapid degradation when the RH was raised to more than 70%.⁶⁵ The normalized efficiency, normalized absorption, and color change trend of the perovskite film at different humidity levels are depicted in Fig. 2. The visual appearance of the yellow-phase perovskite is a good indication of the degree of degradation.^{66,67}

Light and Heat

As a solar energy harvester, PSCs are expected to be under light for a major portion of the day. When heated under continuous light, the device can reach temperatures up to 85 °C.^{68–70} However, light and high temperatures resulting from continuous exposure to sunlight can

degrade PSCs, especially under humid conditions. The chemical reactions that break perovskite molecules in humid conditions are accelerated in the presence of light and at high temperatures.^{71,72} Figure 3 shows the normalized performance parameters of a PSC at different operating temperatures. Rapid degradation of J_{SC} and FF is observed even from 85 °C, resulting in severe degradation of the PCE. However, at low temperatures, the devices are much more stable.

Inorganic perovskites degrade due to phase transformation at different temperatures. For example, $\text{CsPbBr}_{3-x}\text{I}_x$ has a photoinactive δ -phase and three photoactive phases (cubic α -phase, tetragonal β -phase, and orthorhombic γ -phase) depending on the annealing and operating temperatures.⁷³ The cubic α -phase has a suitable bandgap for photovoltaic applications, which is formed at 130 °C. However, when operating at lower temperatures, they tend to turn back to β and γ phases, which are more stable at the operating temperature of PSCs. Therefore, a proper encapsulation for the inorganic perovskites would be able to resist the reaction that shifts the film from α -phase.

Photo-induced degradation is less severe for PSCs, and fortunately, the damage is reversed when kept in the dark.^{74,75} Interestingly, the J_{SC} and V_{OC} increase during the first few minutes of light soaking, greatly improving the device efficiency. Studies suggest that photo-induced PCE improvement is associated with the reduction of iodine vacancies and interstitial Frenkel pair defects.⁷⁶ However, after several minutes, these defects are all filled, and degradation begins, which can reach 10%–30% during the day if other factors (moisture, oxygen) are

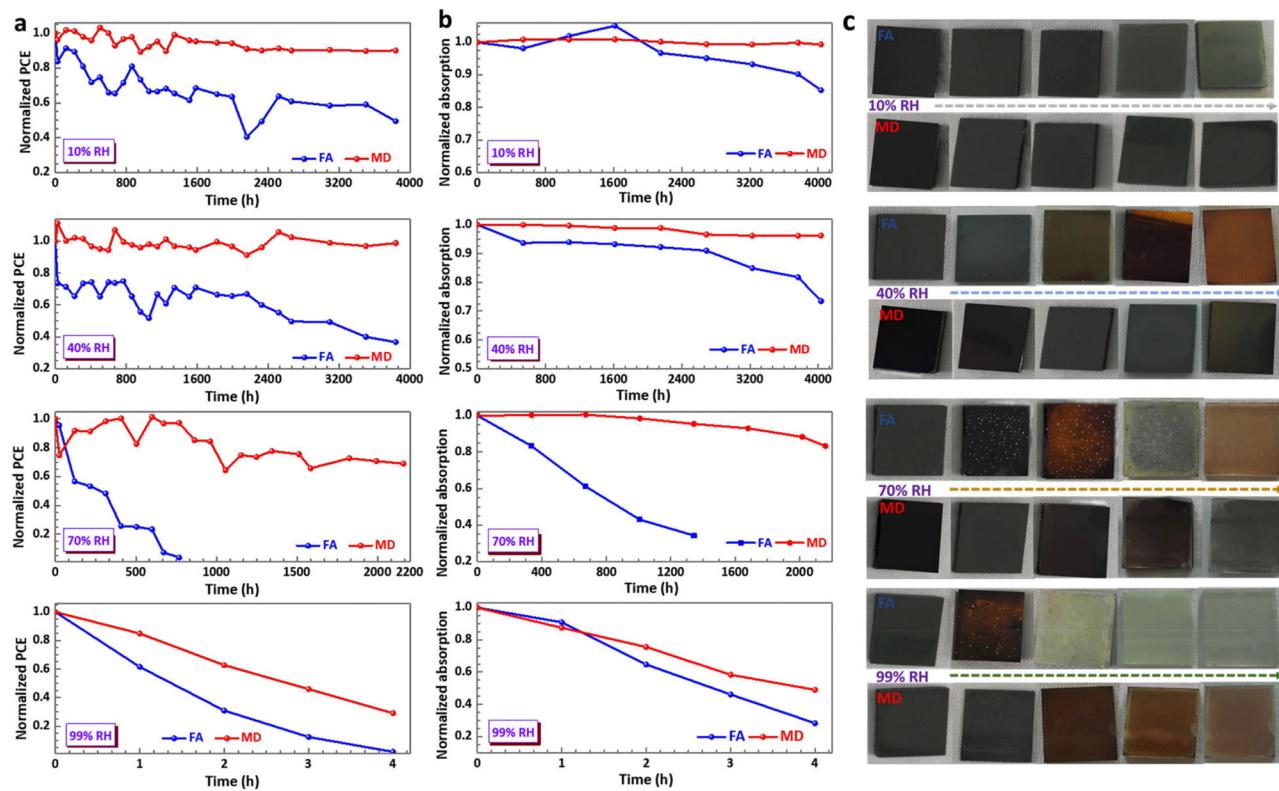


FIG. 2. (a) Normalized PCE, (b) normalized absorption, and (c) visible color change of formamidinium (FA) and mixed-dimensional (MD) perovskite film at different humidity conditions. Reproduced with permission from Zheng *et al.*, Sol. RRL 3, 1800276 (2019). Copyright 2019 John Wiley & Sons, Inc.⁶⁵

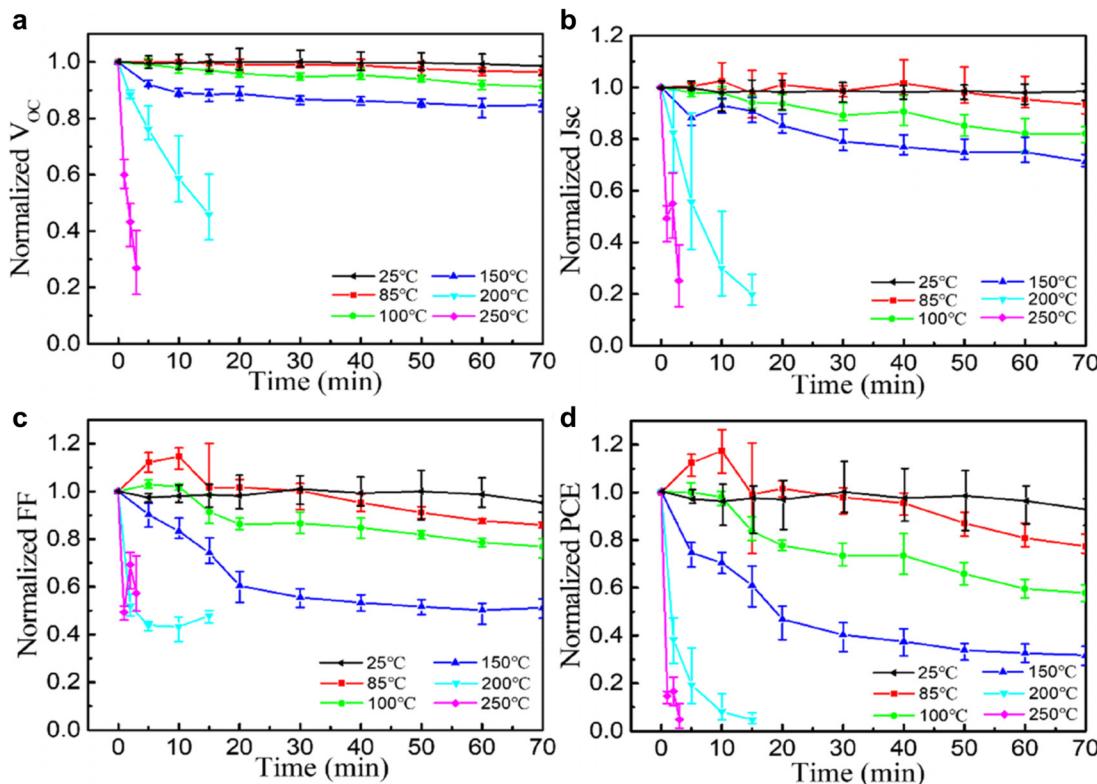


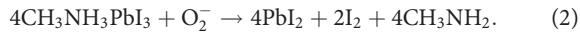
FIG. 3. Normalized performance parameters (a) V_{oc} , (b) J_{sc} , (c) FF, and (d) PCE of $(\text{FAPbI}_3)_{1-x}\text{MAPb}(\text{Br}_{3-y}\text{Cl}_y)_x$ -based PSCs at different temperatures from 25 to 250 °C. Reproduced with permission from Meng *et al.*, *J. Mater. Sci. Mater. Electron.* **32**, 12784–12792 (2020). Copyright 2020 Springer Nature Limited.⁶⁸

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neutralized. Studies have shown that this degradation is reversible at night, when the cells are nonoperational.^{77,78}

Oxygen and ultraviolet (UV) light

The commonly used hole transport layer (HTL) in PSCs is 2,2',7,7'-tetrakis(N,N-di-p-methoxyphenylamine)-9,9'-spirobi-fluorene (spiro-OMeTAD).⁷⁹ When exposed to oxygen, the hole transport level of this HTL becomes deeper when doped with oxygen, increasing the hole transport barrier.^{80,81} This degradation is induced by decreased hole extraction at the perovskite/HTL interface as a result. However, this damage is reversed as shown by Purev-Ochir *et al.* when kept in a nitrogen-filled environment for a long time.⁸² Oxygen also reacts with perovskite in the presence of water, which is an irreversible reaction. In the absence of water (and light), this reaction has a positive enthalpy. However, the enthalpy becomes positive when water is present, making the reaction energetically favorable.⁸³ The formation of superoxide (O_2^-) is a critical step for the whole reaction. It is postulated that iodine vacancies play a critical role in the formation of superoxides from oxygen molecules.^{84–86}



In addition, the ETL/perovskite interface of high-efficiency PSCs with TiO_2 or SnO_2 ETL is prone to UV and oxygen-induced degradation due to photocatalysis. In particular, TiO_2 ETL is severely degraded

when exposed to continuous UV illumination.^{87,88} To improve the stability, Wei *et al.* used ZnTiO_3 as the ETL, which brought better photo-stability.^{89,90} Devices with ZnTiO_3 ETL retained about 90% of their peak PCE after 100 h of continuous UV exposure in contrast to only 55% retention by the control device with TiO_2 ETL.⁹⁰ Therefore, the ETL and ETL/perovskite interface evidently impact the UV stability of PSCs. However, some encapsulation materials use UV filters that might mitigate the impact of continuous UV exposure on PSC device stability.⁹¹

POTENTIAL LEAD TOXICITY

Nearly 50% (w/w) of perovskite films are lead (Pb), which poses a considerable threat to living beings and the environment.^{92–94} Pb toxicity from PSCs can occur in two stages, during operation and at the end of the life cycle. While the latter can be controlled with proper recycling and waste management, the leaching out of lead during the operation of these solar cells is of great concern.^{95,96} For the safe and pollution-free operation of Pb-containing PSCs, the role of good encapsulants cannot be overstated. In a normal operational environment, glass encapsulation is good enough to keep lead in check. However, in the case of cracks and breaking of the panel, lead from underneath perovskite film can come out.^{97–99} The solubility of Pb^{2+} ions in water is a dangerous consequence. To address this challenge, lead-absorbing materials (films, tapes, covering) should accompany regular encapsulants.^{100–103} Lead-absorbing materials should be able to

control leakage under harsh environmental conditions, such as heavy rainfall, and maintain the lead concentration in water below the United States Environmental Protection Agency (EPA) standard of $15 \mu\text{g/L}$.¹⁰⁴

ENCAPSULATION OF PSCS

Encapsulating PSC devices with a suitable material is the commonly employed method for tackling extrinsic degradation and lead leakage.^{105,106} An ideal encapsulant should be a hydrophobic transparent insulator able to prevent the ingress of moisture (and oxygen) and prevent lead from leaching out.^{40,44} Moreover, the usual energy harvesting from the cells should not be hampered; that is, the efficiency of the cells should remain essentially the same before and after encapsulation. The encapsulant material should also be inexpensive and require less energy for preparation and low-temperature processing while they are applied to the device. Some less critical features of an ideal encapsulant material include blocking UV light, having a reflective index close to that of a good antireflective coating, and the ability to absorb mechanical stress.^{44,63,107–110} Encapsulants can seal the surface, edge or both of the active areas of the devices.^{108,111} Most studies show that sealing both the surface and edges is optimal for device stability and lifetime.^{111,112} Figure 4 illustrates the difference between the edge-sealed and surface-sealed encapsulants.

In addition, a good encapsulant should also be able to prevent lead from the perovskite film from leaching into the environment. Although lead leaching is directly related to the degradation of the film, which most encapsulants prevent, harsh conditions such as continuous rainfall or excess mechanical stress resulting in device breakage introduce additional challenges.^{113–115} While edge encapsulation can

give very similar stability, this strategy is not capable of capturing lead ions if the device is broken. Therefore, a suitable lead-capturing encapsulant should cover both the surface and edges of the perovskite film for optimal protection against lead toxicity. Several recent studies have shown very little lead leakage to be compliant with the limit of $15 \mu\text{g/L}$ set by EPA.¹¹⁶ To address these challenges, numerous studies have been undertaken, and several have achieved promising results. Table I summarizes the promising stability achieved by encapsulating PSCs in the last few years. Recent prominent studies that achieved promising results either in stabilizing the device, preventing Pb leakage, or both are discussed below.

Hydrophobic encapsulation

Moisture degrades perovskite more severely than any other external factor. Therefore, hydrophobicity is the first property to be investigated when selecting an encapsulant for PSCs.^{117,118} However, being hydrophobic is not sufficient to prevent water ingress during rainfall conditions. Zhang *et al.* designed a thiol-functionalized perfluoroalkyl-based superhydrophobic-modified perovskite and Ag electrode surface to stabilize PSCs.¹¹⁹ Unlike conventional encapsulants, this was not deposited as a separate layer, rather it was generated as a self-assembled monolayer on the device surface. The contact angle increased from 90.4° to 120.1° as shown in Figs. 5(b) and 5(c), as a result of surface modification. Thiol functional groups were also able to capture uncoordinated lead ions, preventing leakage of these ions. The thiol functional group was able to capture more than 85% of the uncoordinated Pb^{2+} as depicted in Figs. 5(d) and 5(e). However, stability analysis was performed only for 500 h as shown in Fig. 5(a), which does not validate the long-term stability claim.

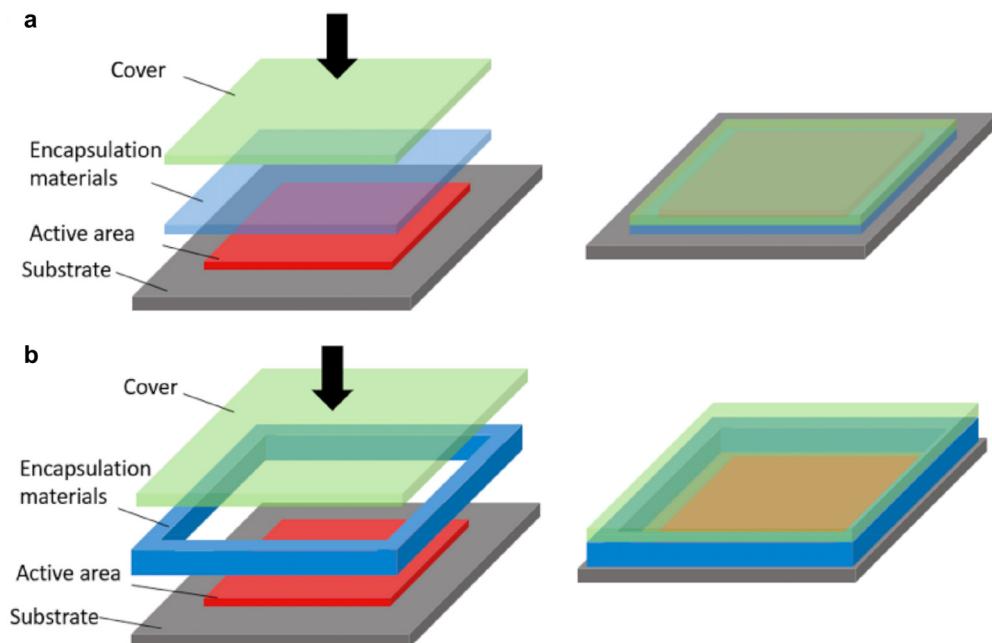


FIG. 4. PSC active device area protected by (a) surface and (b) edge encapsulation. A combination of both surface and edge sealants often results in better protection of the active area. Reproduced with permission from Li *et al.*, J. Power Sources **485**, 229313 (2021). Copyright 2021 Elsevier B.V.¹⁰⁸

TABLE I. Summary of recent studies on the encapsulation of PSCs with reported test conditions and device performance.

| Encapsulation material | Initial PCE | Duration of stability test | Conditions | Retention of peak PCE | Standardized tests performed |
|--|-------------|----------------------------|--|-----------------------|-------------------------------|
| Thiol-functionalized perfluoroalkyl ¹¹⁹ | 21.79% | 500 h 500 h | 85 °C, MPP at N ₂ -filled chamber Room temperature, 85% RH | 90.1% 95% | No |
| PAA ionogel ¹²³ | 22.87% | 1000 h 200 cycles | 85 °C, 85% RH −40 to 80 °C cycling | 95.2% 96.1% | Damp heat, thermal cycling |
| DMDP (front), EDTMP-PEO (back) ^{132,133} | 20.27% | 500 h | MPP at room temperature | 80% | No |
| Multilayer Al ₂ O ₃ and pV3D3 ¹³⁴ | 20.1% | 300 h | 50 °C, 50% RH, dark | 98.9% | No |
| Alucone and Al ₂ O ₃ bilayer stack ¹³⁵ | 17.01% | 2100 h | 30 °C, 80% RH, dark | 96% | No |
| Al ₂ O ₃ and 1H,1H,2H,2H-perfluorodecyl-trichlorosilane stack ¹³⁶ | 19.7% | 500 h | 85 °C, 85% RH | 78% | Damp heat (not long enough) |
| PIB wide blanket ¹¹¹ | 19% | 1800 h 30 cycles | 85 °C, 85% RH −40 to 80 °C cycling | 95% 95% | Damp heat and humidity freeze |
| Resins UVR-C (front), C100 (back) ¹¹⁵ | 25.5% | 500 h | Ambient conditions | 97% | No |
| Sulfonic acid-based resin ¹³⁷ | 20.1% | 500 h | 45 °C, MPP, ambient RH | 83% | No |

Lead-absorbing ionogel encapsulant

Ionogels possess tunable mechanical, optical, and chemical properties owing to their liquid electrolyte composition, which allows tuning of these properties and the incorporation of suitable molecules as additives.^{120–122} Xiao *et al.* developed an ionogel-based PSC encapsulant with lead-absorbing monomers and acrylic acid, a covalent cross-linker of methylene-bis-acrylamide, a thermal initiator of azobisisobutyronitrile, and an ionic liquid of tributyl(methyl)phosphonium dimethyl phosphate as shown in Figs. 6(a) and 6(b), abbreviated as PAA ionogel.¹²³ Polyacrylic acid hydrogel networks swollen in water are commonly employed to anchor heavy metal ions with carboxyl groups for water purification.^{124,125} Replacing water with a nonvolatile and thermally stable phosphonium ionic liquid stabilizes the gel by suppressing solvent evaporation. Moreover, they provide more sites for lead capture with phosphate anions.¹²⁵

Interestingly, the ionogel encapsulant also provided mechanical firmness to the modules. The modules were hammered to break and kept for 24 h as shown in Figs. 6(c) and 6(d). The control modules showed visible degradation upon formation of the yellow phase, while the encapsulated cells were considerably more stable. In addition, the authors soaked the broken modules in water to demonstrate lead leakage prevention. For this test, there were control samples (no encapsulant), modules with polyolefin encapsulant, and modules with ionogel encapsulant. Lead leakage of the control and polyolefin encapsulant had little difference. However, the ionogel encapsulants had four orders of magnitude less leakage. This finding demonstrated the superior performance of this class of encapsulants in preventing lead leakage. In addition, the modules passed the damp heat test for 1000 h and thermal cycling test for 200 cycles withstanding more than 95% of their peak efficiency.

Although promising results have been shown for ionogel encapsulants, the main challenge in implementing these is the complexity of the processing and the material cost. The encapsulant thickness is approximately 100 μm, which is more than 200 times the perovskite film thickness; hence, more materials are consumed.¹²⁶ One of the major advantages of PSCs over Si PV is the lower usage of materials; thus, an ideal encapsulant should also be a thin film consisting of low-cost, facile processable materials.

Epoxy resin-based self-healing polymer encapsulation

Epoxy resins are well known for their self-healing properties in electronic applications.^{127,128} Due to their two-part additive properties, these materials are ideal for use as sealants. An epoxy resin, having a mixture of diglycidyl ether bisphenol A, n-octylamine, and m-xylenediamine (4:2:1), was used for encapsulating PSCs in a study by Jiang *et al.*¹²⁹ The resulting encapsulation material is able to self-heal and thus withstand large amounts of mechanical stress.^{130,131} This encapsulation can prevent lead leakage by approximately three orders of magnitude under harsh weather conditions, as simulated by breaking the modules by Jiang *et al.* Figure 7(a) shows a schematic of the epoxy resin encapsulant before and after cutting, and the depth of the cut as a function of time is depicted in Fig. 7(e) at three different temperatures. Self-healing is a temperature-assisted process, and the contact angle of the film with water returns to the initial value due to self-healing, as depicted in Figs. 7(b)–7(d).

Moreover, lead capture by the encapsulation cross-linking network depends on the weather conditions. Raining for shorter durations, such as less than 4 h, does not allow the film to prevent much lead leakage. However, intermittent rain and sunny weather conditions have a large effect on lead leakage from the devices. During sunny hours, the film has enough time to self-heal and form a cross-linking network to capture lead, which would otherwise leach out. However, the stability of these devices has not been reported, which is one of the primary requirements of any PSC encapsulant.

On-device lead-absorbing encapsulant

Two different lead-absorbing films were used to cover the front and back surfaces of PSC modules by Li *et al.*^{132,133} to protect against lead contamination. They used the transparent phosphonic acid containing material P,P'-di(2-ethylhexyl)methanediphosphonic acid (DMDP) for the front surface, and a polymer blended with the lead-chelating agent N,N,N',N'-ethylenediaminetetrakis(methylenephosphonic acid) (EDTMP-PEO) for rear surface encapsulation. The device structure and the chemical structure of DMDP and EDTMP-PEO are depicted in Figs. 8(a) and 8(b). There was minimal change in the transmission of the device due to the addition of a new film on the front surface.

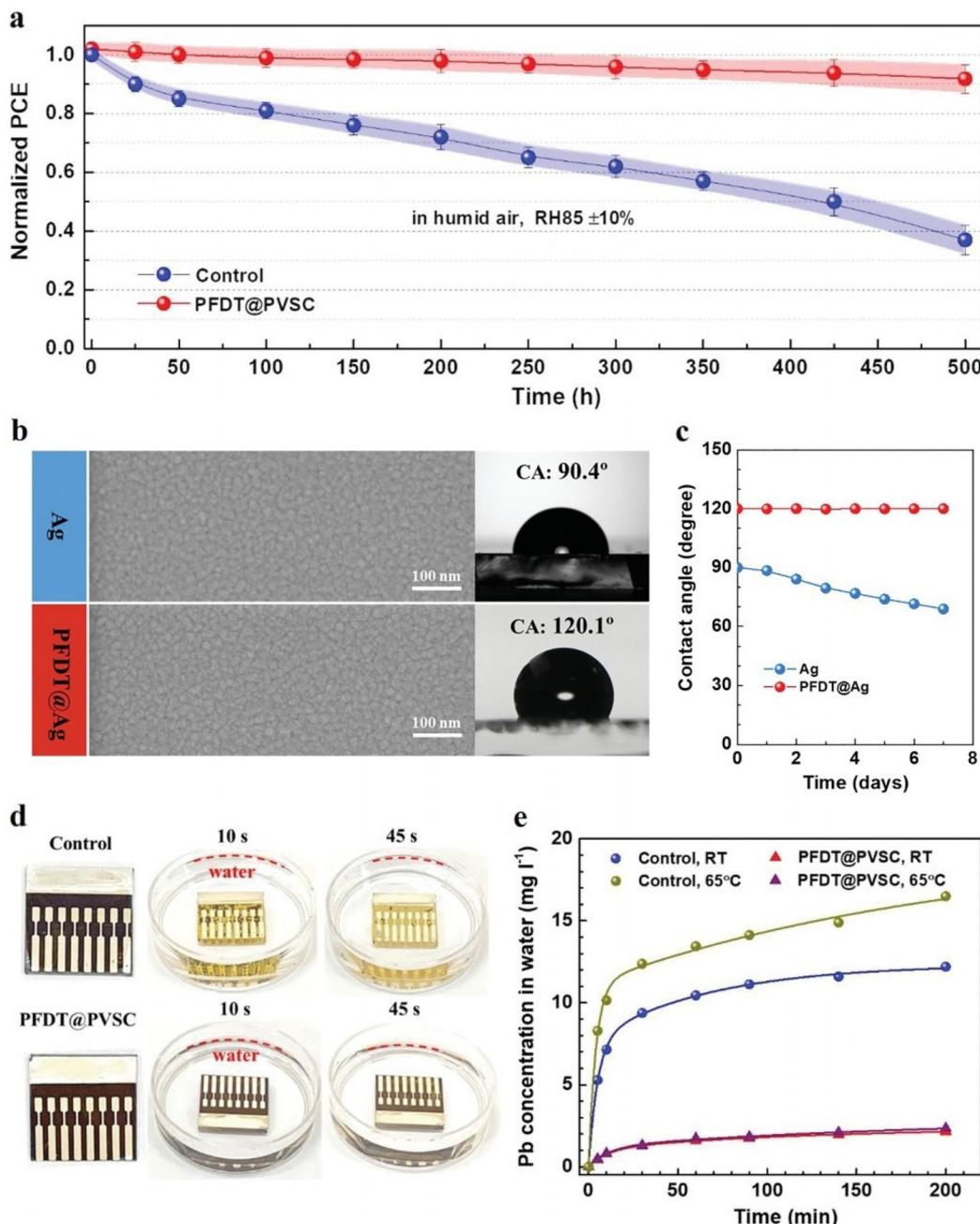


FIG. 5. (a) Device stability of a PSC with a thiol-functionalized perfluoroalkyl-modified superhydrophobic surface and (b) contact angle with and without surface modification. (c) Change in the contact angle with time. (d) Experimental setup of immersing the devices in water to test lead leakage. (e) Pb^{2+} concentration in the water indicating lead leakage from the control and modified surface PSCs at room temperature and 65°C. Reproduced with permission from Zhang *et al.*, *Adv. Energy Mater.* **11**, 2102281 (2021). Copyright 2021 John Wiley & Sons, Inc. ¹¹⁹

The films were crushed such that the glass substrate was broken and kept in water at room temperature and 50°C, resulting in approximately 96% less lead leakage from the encapsulated devices [Fig. 8(c)]. Moreover, Fig. 8(d) shows the distribution of lead in different films of the encapsulated PSC modules. Surprisingly, only 1.1% and 1.9% of

the total lead in the device were found in water at room temperature and 50°C, respectively, showing tremendous improvement against lead contamination. In their subsequent study, Li *et al.* made tapes of DMDP (front) and DMDP-laminated EVA (rear) that can be readily applied to any PSC.¹³³ However, this film cannot provide extra

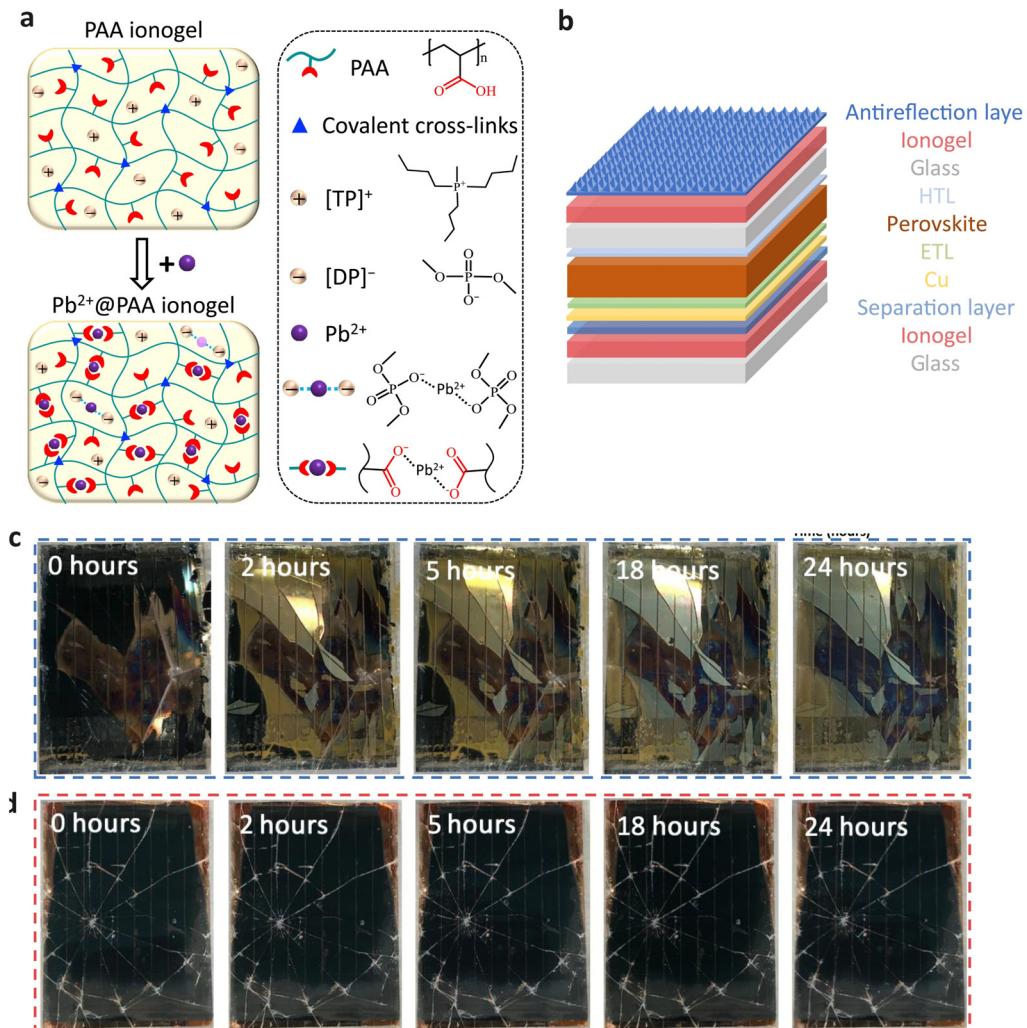


FIG. 6. (a) Structure of the polyacrylic acid ionogel before and after lead capture. (b) Structure of the device with the PAA ionogel covering both sides. (c) and (d) Images of perovskite modules hammered to break without (c) and with (d) the applied ionogel encapsulant. Reproduced with permission from Xiao *et al.*, Sci. Adv. 7, eabi8249 (2021). Copyright 2021 American Association for the Advancement of Science.¹²³

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protection against moisture and oxygen ingress. Therefore, an additional film of ethylene-vinyl acetate (EVA) is used on the rear of the devices to protect them from degradation. In addition, the thickness of the tape is approximately 5.7 and 10 μm for the front and rear surfaces, respectively. Having films of this thickness along with EVA requires materials approximately 100 times the perovskite film (by volume); thus, material usage becomes an issue. The materials used to process the films are also not cost-effective; thus, they contrast two of the critical advantages of PSCs.

Thin-film encapsulants

Thin-film encapsulants of PSCs are not uncommon.^{135,138–140} Lee *et al.* stacked multiple layers of Al₂O₃ and poly(1,3,5-trimethyl-1,3,5-trivinyl cyclotrisiloxane) (pV3D3) on top of the perovskite layer via

initiated chemical vapor deposition (ICVD) and atomic layer deposition (ALD).¹³⁴ They achieved a water vapor deposition rate on the order of 10^{-4} g m⁻² d⁻¹ at an accelerated condition of 38 °C temperature and 90% RH. However, no standard stability test was performed on their work to make a detailed comparison with other encapsulation studies.

Plasma-induced ALD (typically for Al₂O₃) is often used for depositing an encapsulant, which severely damages the perovskite. In addition, Wang *et al.* used ethylene glycol to form an intermediate layer to protect the inside perovskite layer when depositing any encapsulant that needs harsh conditions such as high temperature.¹³⁵ Afterward, encapsulation was performed using ALD Al₂O₃. The device structure and the performance without and with encapsulation are shown in Fig. 9. The water vapor transmission rate decreases by almost 80% when three bilayers of Alucone and Al₂O₃ are employed.

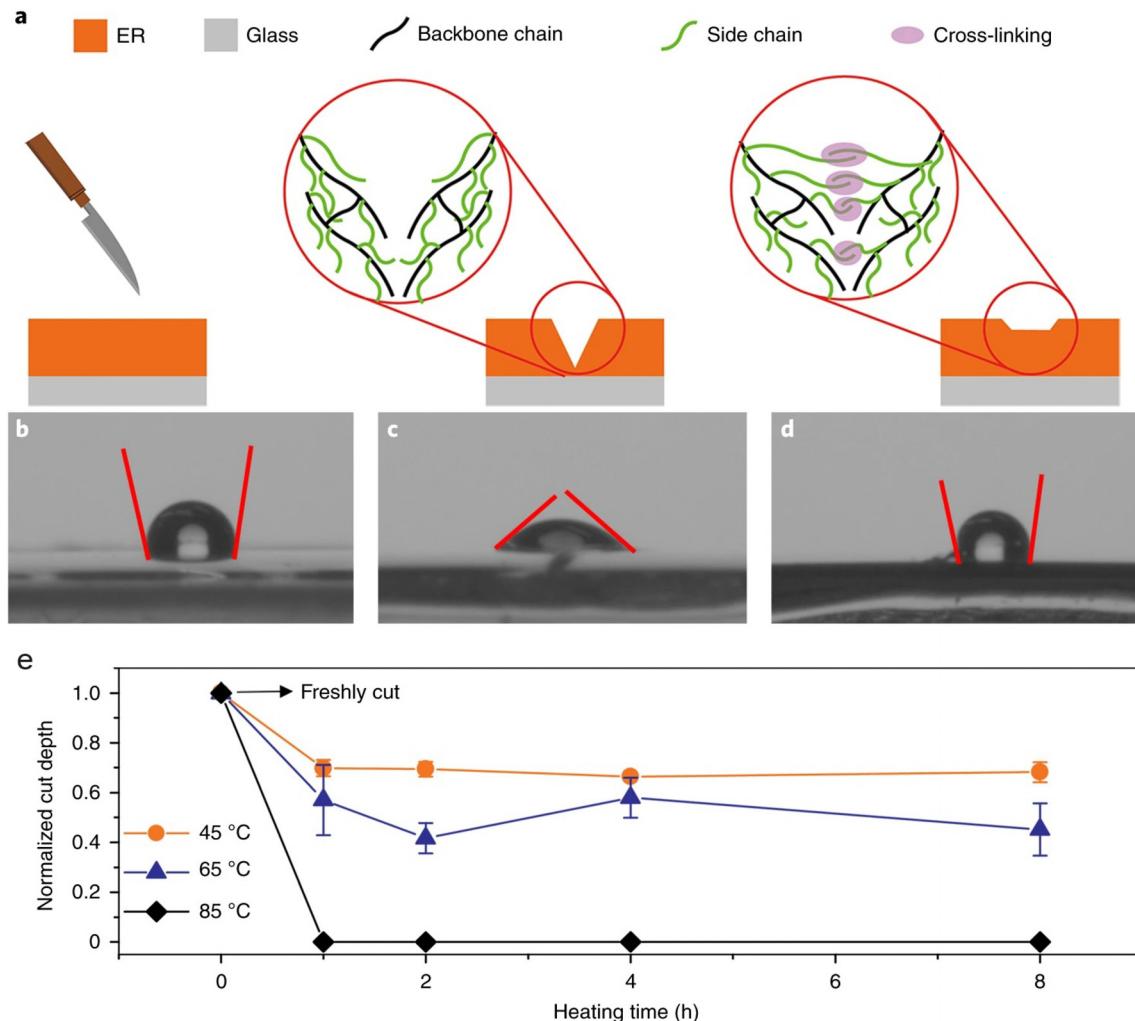


FIG. 7. (a) Schematic showing the self-healing process of the epoxy resin encapsulant. (b)–(d) Images showing the water contact angle measurement on the pristine ER film (b), the film after cutting (c), and the film after cutting and heating at 45 °C for 4 h. (e) The normalized cut depth as a function of heating time at three different temperatures (45, 65, and 85 °C) for the epoxy resin film. Reproduced with permission from Jiang *et al.*, Nat. Energy 4, 585–593 (2019). Copyright 2019 Springer Nature Limited.¹²⁹

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Although the results are promising, the absence of any standard stability testing makes direct comparison with other reported encapsulation techniques difficult.

Mixed resin-based encapsulation

Resin materials have gained considerable popularity for encapsulating PSCs due to their ability to reflect lead internally.^{141–144} Chen *et al.* reported a sulfonic acid-based lead-absorbing resin that scaffolds perovskite films on the glass side.¹³⁷ This scaffold perovskite–resin structure is capable of absorbing lead even under rainfall conditions, keeping the lead level in water below 11.9 ppb. Interestingly, incorporating this layer that connects electrically with the device does not affect the device performance. In a different study, Li *et al.* employed a mixture of cation exchange resin (C100) and an ultraviolet resin (UVR-C) to encapsulate PSCs to absorb leached lead ions.¹¹⁵ Lead

leakage is mainly prevented by cation exchange between the Na^+ ions of the C100 resin and the Pb^{2+} ions. The coating on both sides of the PSC was also accompanied by a 1.1 mm glass substrate with resin films. The lead leakage was less than 5 ppm under simulated rain conditions, passing the standards set by the Resource Conservation and Recovery Act Regulation (RCRA).^{145,146} Figure 10 shows the structure of the lead-capturing resin encapsulant and compares the cost of C100 with that of other encapsulant materials. Surprisingly, the cost of C100 resin is approximately 80 times lower than that of the next cheapest encapsulant (Surlyn films).

Encapsulation strategies for inorganic halide PSCs

All inorganic PSCs are gaining popularity due to their increased intrinsic stability compared to hybrid perovskites.^{147–150} However, they also suffer from thermal and structural degradation and pose the

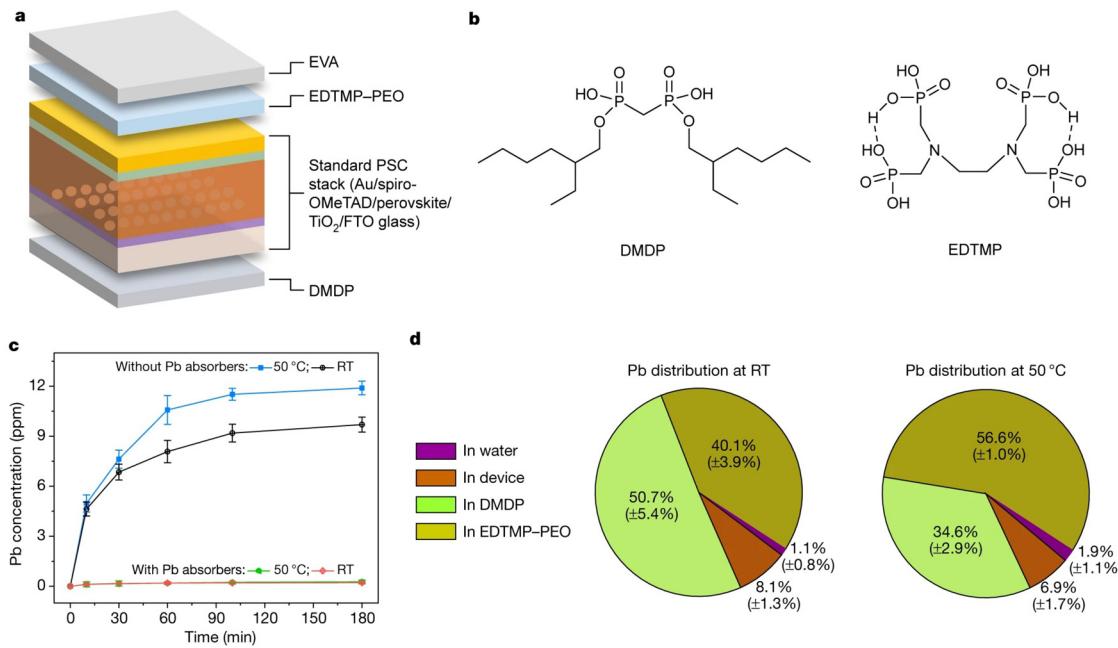


FIG. 8. (a) Structure of the PSC with both the front and back surfaces encapsulated. (b) Chemical structure of DMDP and EDTMP-PEO. (c) Pb concentration in water after different broken PSCs are immersed in it for up to 180 min. (d) Distribution of the device Pb after they are broken and immersed in water. Reproduced with permission from Li *et al.*, Nature 578, 555–558 (2020). Copyright 2020 Springer Nature Limited.^[32]

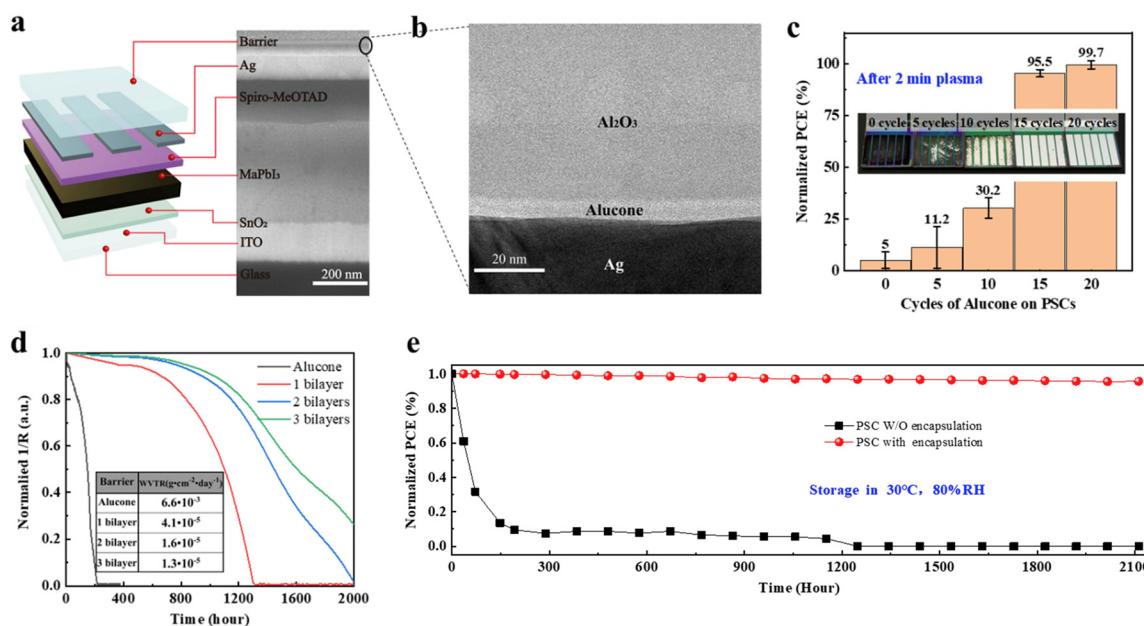


FIG. 9. (a) Schematic illustration of the encapsulated PSC with a cross-sectional SEM image. (b) Zoomed view of the encapsulation layer from the cross-sectional image. (c) Effect of the number of cycles of Alucone on the PSC. (d) Normalized water vapor transmission rate when multiple layers of Al₂O₃ are stacked on an Alucone. (e) Normalized PCE of the PSCs with and without encapsulation at 30 °C and 80% RH. Reproduced with permission from Wang *et al.*, Nano Energy 69, 104375 (2020). Copyright 2020 Elsevier B.V.^[35]

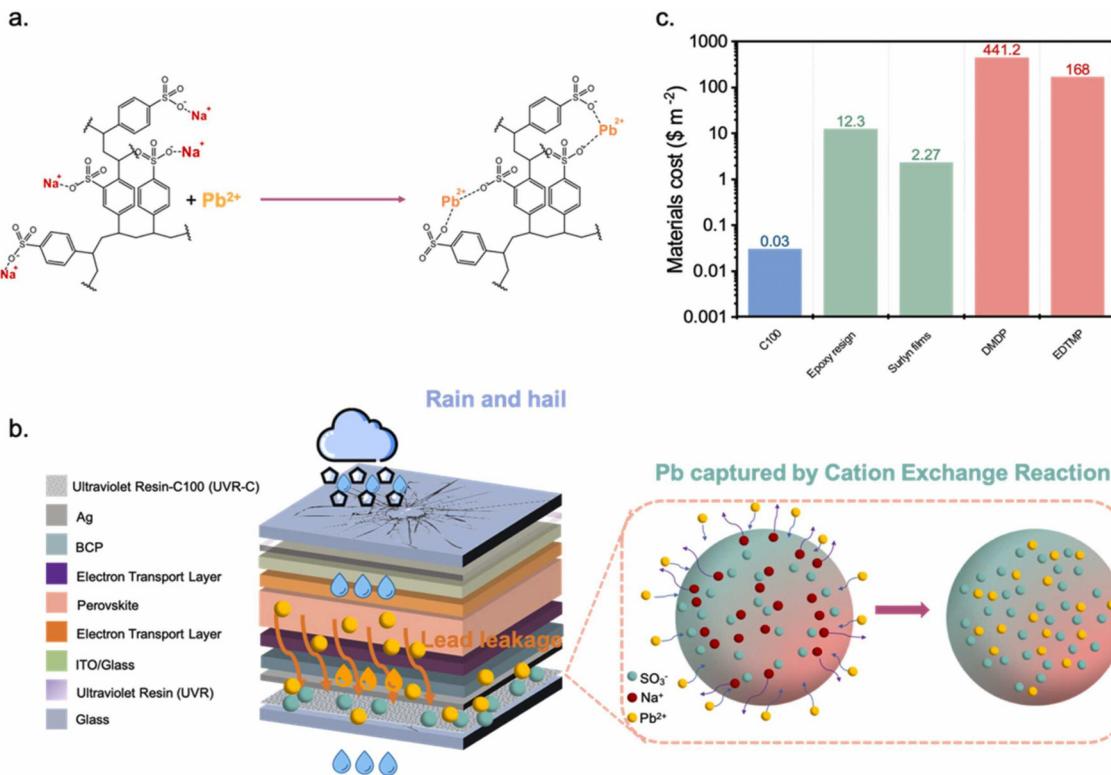


FIG. 10. (a) Chemical reaction showing the trapping of Pb^{2+} ions by cation exchange with the Na^+ ions of the C100 resin. (b) Device structure showing the encapsulation technique on both sides of a PSC using additional glass on each side. (c) Material cost of the C100 resin, compared with that of different other encapsulant materials used in PSCs for lead capture. (d) Schematic showing the reaction through which lead capture occurs inside the device. Reproduced with permission from Li *et al.*, Nano Energy 93, 106853 (2022). Copyright 2022 Elsevier B.V.¹¹⁵

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threat of lead toxicity. Thermoplastic polyurethane (TPU) was reported as a good encapsulant for inorganic PSCs due to its optical transparency, flexibility, and elasticity. TPU was used by Shi *et al.* at room temperature to bind CsPbBr_3 QDs and $\text{CsPbBr}_3@\text{Cs}_x\text{PbBr}_6$ NCs, resulting in a significant decrease in thermal degradation, which they showed by photoluminescence intensity.¹⁵¹ In a very interesting study, Lin *et al.* encapsulated the perovskite thin film with mesoporous SiO_2 to form a complex, which prevented thermal degradation of a wide bandgap range of perovskites.¹⁵²

Commercially available encapsulants

The encapsulants available for PSCs on the market are polymer type tapes. The most popular commercially available encapsulant for PSCs is EVA [Fig. 11(a)],^{153,154} which has also been used as an interface material in some studies.^{155–158} EVA materials possess excellent transparency, moisture impermeability, and facile processing steps.^{159,160} In addition, EVA can be used to seal both the surface and edges of devices. Similarly, polyisobutylene [PIB; Fig. 11(b)] is another commercially available encapsulant for PSCs.^{111,161} Blanket sealing of PSC modules with PIB has been shown to pass both the International Electrotechnical Commission (IEC) 61215:2016 Damp Heat and Humidity Freeze tests.¹¹¹ They provide excellent protection against external factors, such as moisture, oxygen, UV light, and heat.

However, neither of these two materials provides much impedance to the leaching of lead ions. In addition, the thicknesses of both EVA and PIB are more than $100 \mu\text{m}$, which is more than 200 times the perovskite thickness, and a significant amount of material is used. Therefore, the current commercial encapsulants suffer from two major problems, lead leakage through the encapsulants, and large material usage.

STANDARD TESTING PROTOCOL

For a long time, PSCs were processed and reported without unified standard testing protocols, such as IEC 61215. As a result, the

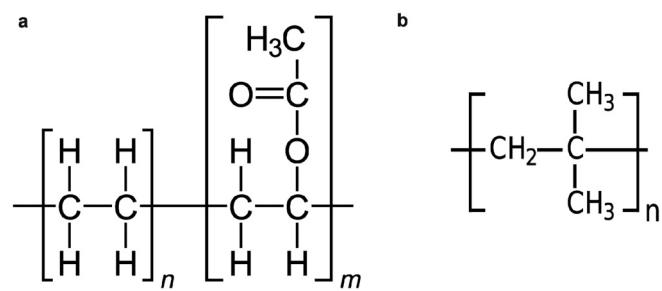


FIG. 11. Chemical structures of (a) EVA and (b) PIB, which are two commercially available encapsulants for PSCs.

reported performance in those studies varied widely from laboratory to laboratory and is not fully reproducible within the research community.^{162–165} Additionally, there are no standardized tests for assessing lead leakage in PSCs, nor are there established methods that could be adapted from other photovoltaic (PV) technologies. Consequently, the criteria for “harsh” or “rainfall” conditions lack uniformity across studies, leading to confusion about which guidelines should be followed when evaluating newly developed materials for PSC encapsulation against lead leakage. Therefore, it is imperative for the research community to establish a standardized protocol for lead leakage testing in the near future. The testing protocols for stability involve using masks for PV data acquisition, reporting hysteresis, and reporting conditions for long-term stability tests.^{166–169} Most importantly, accelerated aging tests are crucial for assessing the performance of any encapsulation.¹⁶⁹ Over the years, three tests have been identified and often conducted for any new encapsulant material or method to compare against other technologies, namely, the damp heat test, thermal cycling test, and humidity freeze test. For any study reporting new encapsulation materials or methods, the cells are expected to pass all three tests.

Damp heat test

The damp heat test is the most common accelerated aging test and involves exposing encapsulated PSCs to 85 °C and 85% relative humidity simultaneously for 1000 h.^{64,168,170,171} The cells are placed in a damp heat chamber where these conditions are moderated. The damp heat test particularly challenges the lamination of the cells along with their edge sealing against humidity. Different types of defects, including delamination, corrosion on the surface or edges, and metal grid dislocation are detected by the damp heat test. To pass this test, a PSC must retain >95% of its initial PCE after 1000 h in damp heat conditions.^{64,170} Any encapsulation technology passing the damp heat test is expected to protect the solar cell for 20 years. However, due to internal degradation of PSCs, the lifetime can be lower, that remains to be determined.

Thermal cycling test

Another crucial accelerating aging test for PSCs is the heat cycle test, which involves cycling the temperature 200 times from –40 to 85 °C, dwelling at extreme temperatures for 10 min, as shown in Fig. 12.^{172–174} The cycles are continuous, with a maximum of 6 h for each cycle. This test is performed to address the thermal fatigue of PSCs and determine whether the cells degrade within the maximum limit of the operational temperature (85 °C). During the thermal cycling test, current needs to be applied to the cells, which was not applied in some of the reports. The current is crucial because the current during the operational period can create local hot spots that might exceed 85 °C. The cells must retain more than 95% of their initial PCE to pass this test. The modules passing the thermal cycling test are expected to withstand real-world temperature changes for 20 years.

Humidity freeze test

This test involves high temperature and humidity (damp heat testing conditions) followed by a temperature of less than 0 °C. The cells dwell at damp heat conditions for at least 20 h, and then at –40 °C for at least 30 min for 10 cycles. One cycle can reach up to 24 h (or 1 day) as shown in Fig. 13, with temperature change rates and minimum dwell times. This test is enforced to challenge PSCs against rapid humidity and temperature changes. Modules passing this test should sustain the rapid changes in humidity and temperature during operational conditions for 20 years. However, for PSCs, the operational lifetime is not determined yet, which is expected to be lower. Unfortunately, this test is not often reported in PSC encapsulation studies.

Other stability tests

Apart from standard stability tests, operational stability tests and storage stability tests are important for PSCs. During operation, the cells are exposed to continuous light throughout the day, during which both reversible and irreversible degradation can occur due to light

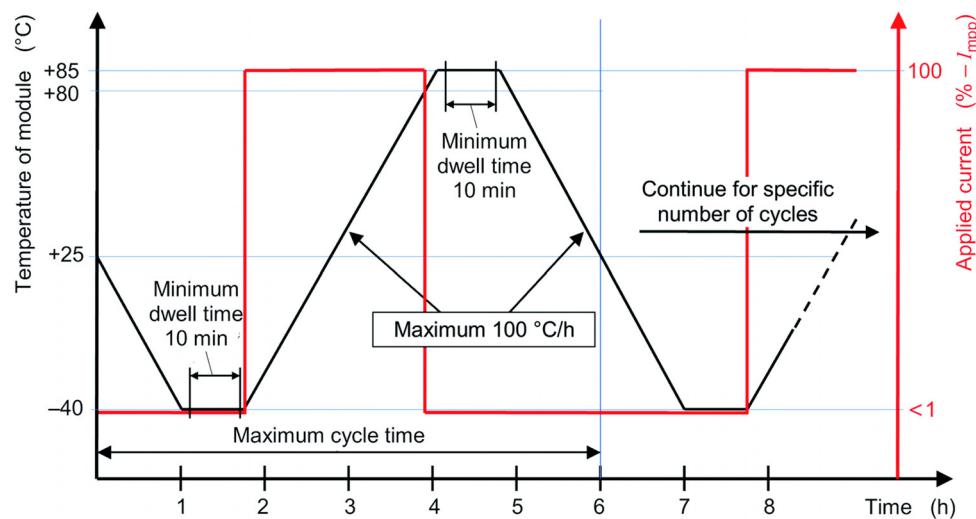


FIG. 12. Temperature and applied current variation along with important timing parameters for a thermal cycling test according to IEC 61215 standards. Reproduced with permission from Holzhey *et al.*, J. Mater. Chem. A **6**, 21794–21808 (2018). Copyright 2018 Royal Society of Chemistry.

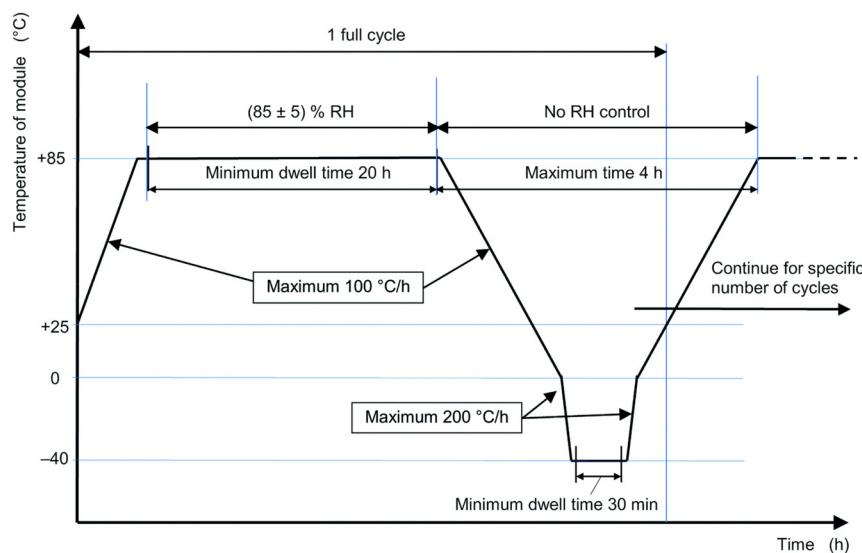


FIG. 13. Temperature and humidity variation in a typical humidity freeze test. One cycle corresponds to 20 h at damp heat conditions; and a maximum of 4 h for temperature change and freezing conditions. Reproduced with permission from Holzhey *et al.*, *J. Mater. Chem. A* **6**, 21794–21808 (2018). Copyright 2018 Royal Society of Chemistry.¹⁷⁴

exposure. To estimate these photo-induced degradations, a continuous operational stability test (AM1.5G illumination, 25 °C, 60% RH) of at least 1000 h is needed for any newly reported encapsulant.^{175,176} The devices are expected to withstand more than 90% of their peak PCE at the end of 1000 h of operation. Moreover, after resting in the dark following the operational conditions for 24 h, the devices should recover most of the lost PCE, which was recoverable damage. Storage stability tests can be performed in a dark and nitrogen-filled environment. Therefore, the only degradation element that can occur is internal degradation. All the important tests for PSCs are listed in Table II. However, at this stage, only three crucial tests (damp heat, thermal cycling, and humidity freeze) are frequently reported along with operational stability tests.

Lead leakage test

For encapsulated PSCs, Pb²⁺ ions can leach out if the encapsulation breaks or due to micro air gaps on the edges of the encapsulant. In both cases, water can penetrate inside the device and dissolve these ions. Therefore, two tests are necessary to determine the capacity of any PSC encapsulant to prevent lead leakage. First, the encapsulated modules must not leak any lead when continuously immersed in water. Considering rainfall conditions in most parts of the world, this

test can be performed for 5–7 days, and the lead content of the used water should be reported as $\mu\text{g}/\text{cm}^2$ such that the impact of the amount of water used is removed. This test will challenge the ability of encapsulation to prevent inward and outward influx of water.

Furthermore, to simulate the condition of broken panels, the modules need to be cracked and immersed in water. Here, the additional lead-absorbing layer must absorb a major portion of lead before leaking out. This will test the ability of the additional lead-capturing film to prevent lead from leaching out. Though there is no standard duration for this test to be adopted, the authors believe that 3–4 h in immersed conditions will be sufficient to determine the lead-capturing ability of the encapsulant in cracked or broken conditions.

CONCLUSION

Although PSCs are an increasingly popular solar cell research area, the future of this PV technology is still uncertain due to several crucial challenges. Two of the major challenges are the durability of the modules in the long term and lead leakage. Most of the studies on PSCs are based on improved PV performance, which is already at a decent level. Conversely, the number of studies undertaken to prevent lead leakage of PSCs is relatively low. In addition, most studies on

TABLE II. Summary of all the standard testing protocols PSCs are required to pass before commercialization.

| Test | Conditions | Duration |
|-----------------------------|--|------------|
| Outdoor exposure test | 60 kW h m ⁻² light | 2100 h |
| Hot-spot endurance test | 1000 W m ⁻² irradiance at maximum heat dissipation condition | 5 h |
| UV preconditioning test | 15 kW h m ⁻² UV irradiance (280–400 nm) at 60 °C | 100 h |
| Thermal cycling test | 200 cycles from –40 to 85 °C with applied current (I_{SC}) | 6 h/cycle |
| Humidity freeze test | 10 cycles from –40 to 85 °C, 85% RH to –40 °C | 24 h/cycle |
| Damp heat test | 85 °C and 85% RH | 1000 h |
| Hail test | 25-mm-diameter ice ball at 23 m s ⁻¹ , directed at 11 impact locations | N/A |
| Static mechanical load test | Three cycles of uniform mechanical load (front and back) specified by the manufacturer | 1 h/cycle |

encapsulation that show improved stability do not perform lead leakage tests, even though they are related. Therefore, it is of utmost importance to develop encapsulants that can solve both of these challenges while minimizing the associated costs.

There have been several reports on the novel encapsulation of PSCs, as summarized in this study. Several of them claimed to have solved both stability and lead leakage issues. Many of these studies did not follow standard accelerated aging tests (damp heat test, thermal cycling test, humidity freeze test). As a result, it is difficult to compare their performance and select the optimum solution. Moreover, there is currently no standard lead leakage test available for PSCs, leading to the use of a wide range of strategies in different studies. Most of the studies show the parts per $\times 10^6$ (ppm) level of lead in water when the device is broken as an indication of lower lead leakage. However, the ppm is related to the amount of water used, the area of the device immersed, and the duration of the test. It is urgent for the community to develop standard lead leakage tests for PSCs such that all studies can adhere to these tests. This will also help researchers and readers compare the ability of the encapsulants to prevent lead leakage. Moreover, newly developed encapsulants should also be reported with their ability to inhibit lead leakage under extreme environmental conditions. The remarkable progress made in recent studies has made the research community believe that a suitable PSC encapsulant capable of protecting the device and preventing lead leakage is not too far from our sight.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Shahriyar Safat Dipa: Conceptualization (lead); Formal analysis (equal); Investigation (equal); Software (equal); Writing – original draft (lead). **Md. Arifur Rahim:** Formal analysis (equal); Software (lead); Supervision (equal); Validation (equal); Writing – review & editing (equal). **Ashraf Uddin:** Project administration (lead); Supervision (lead); Validation (equal); Writing – review & editing (equal).

DATA AVAILABILITY

Data sharing is not applicable to this review as no new data were created or analyzed in this study.

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