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Sustainable Approaches to Address Lead Toxicity in Halide Perovskite Solar Cells: A Review of Lead Encapsulation and Recycling Solutions

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ABSTRACT

The increasing global concerns about energy shortages and environmental pollution are driving the development of materials for clean energy conversion. Among various materials, lead halide perovskite solar cells (PSCs) have emerged as promising candidates for next-generation photovoltaic (PV) technologies. However, the use of toxic lead in high-efficiency perovskite devices raises sustainability concerns, particularly due to the risk of environmental contamination from lead leakage. Given the projected growth of the perovskite photovoltaic market, effective management of lead toxicity is essential for the safe deployment of this technology. This review explores the latest developments in lead encapsulation strategies, including both external and internal encapsulation materials, aimed at mitigating lead leakage and enhancing the safety and sustainability of perovskite photovoltaics. Additionally, it also discusses various recycling solutions necessary to establish a sustainable closed-loop lead management system. These approaches not only recycle lead but also reclaim other materials, promoting the circular use of resources and advancing the competitiveness of perovskite PV technologies.

1 | Introduction

Global concerns regarding energy shortages and environmental contamination are driving the development of materials for clean energy conversion. Many materials have been developed for their ability to capture solar energy, due to their eco-friendliness, sustainability, and widespread accessibility, particularly in photovoltaic devices. Among these materials, metal halide perovskite solar cells (PSCs) have garnered significant interest and demonstrate tremendous potential as the next generation of photovoltaic (PV) technologies. This is primarily due to factors such as their ability to achieve high power conversion efficiencies (PCEs), simple fabrication processes, and cost-effective manufacturing.

The light-absorbing layer in PSCs is based on an inorganic–organic metal halide compound that forms a crystal structure known as perovskite. These perovskite materials adopt the chemical formula ABX_3, where A represents a monovalent cation, such as methylammonium (MA^+), formamidinium (FA^+), cesium (Cs^+), or rubidium (Rb^+); B is a divalent metal cation, such as lead (Pb^{2+}) and tin (Sn^{2+}); and X is a halogen anion, encompassing Cl^-, Br^-, and I^-. In the PSC architecture, the perovskite layer is integrated with various charge-separate layers and conductive electrodes. In the past 10 years, remarkable certified PCEs have increased to 26.7% for single-junction PSCs and 33.9% for perovskite-silicon tandem solar cells [1]. Moreover, the stability of PSCs has also been significantly enhanced [2–6], meeting various industry-standard tests, including long-term maximum power point tracking under continuous one-sun illumination, damp heat (85% humidity, 85°C), and temperature cycling tests (−40°C to 85°C). These achievements demonstrate that PSCs have the potential to become the next-generation low-cost photovoltaic technology.

Despite significant improvements in the PCE and stability of PSCs, the use of toxic lead—a critical component for commercialization—raises sustainability concerns for future photovoltaic applications, as shown in Figure 1. The leakage of lead from damaged cells poses a risk of contaminating soil and groundwater. Consequently, these lead-containing compounds may be deposited into the ecosystem through rainwater or air, causing adverse effects on both living organisms and the environment. According to previous reports, a solar PV capacity of 1 GW utilizing perovskite modules with 20% efficiency would contain approximately 3.5 tons of lead, based on the most advanced perovskite materials and assuming a perovskite film thickness of 500 nm [7]. Extrapolating to a broader context, if only 20% of the anticipated 8500-GW PV market in 2050 is adopted by perovskite PV, the cumulative lead content in perovskite modules could reach up to approximately 6000 tons.

Addressing Pb toxicity is the foremost challenge for the practical integration of perovskite photovoltaics, as the presence of lead poses significant environmental and health risks. To ensure the safe deployment of this promising technology, it is crucial to develop strategies that effectively manage and mitigate these risks. The most effective approach involves the use of stable and efficient lead encapsulants, which can contain and control lead leakage throughout the lifespan of the photovoltaic cells. Accordingly, this review focuses on the latest advancements in both external and internal lead encapsulation materials, evaluating their design, performance, and potential to prevent lead leakage and enhance the long-term safety of perovskite photovoltaics. Additionally, this review introduces various lead recycling strategies that aim to recover lead with high purity and efficiency. The integration of recycling processes into the lifecycle of perovskite photovoltaics not only reduces the reliance on raw lead but also ensures that the environmental footprint of these cells is minimized. By combining encapsulation with robust recycling programs, the industry can better manage lead throughout the entire product lifecycle, paving the way for safer and more sustainable perovskite photovoltaic technologies.

2 | Hazard Assessment of Toxic Lead

Unlike widely commercialized silicon solar cells, the lead toxicity in PSCs raises sustainability concerns. Perovskite solar modules designed for outdoor use can be damaged or broken by various natural factors, such as hail, snow, wind stress, or fires during operation. This damage may lead to the release of components containing lead, resulting in the leakage of lead-related substances. Considering its chemical properties, lead compounds such as $PbCO_3$, PbO, $Pb(OH)_2$, and $PbSO_4$ are typically deposited on soil surfaces and in groundwater following a leakage event. These hazardous elements accumulate in the environment, propagating through the food chain and circulating within the atmosphere, soil, water, plants, animals, and ecosystems. Consequently, lead, a top-ranked hazardous substance, can enter the human body through inhalation, ingestion, and skin absorption.

FIGURE 1 | Illustration of the key factors influencing the commercialization of perovskite solar cells.

Research indicates that 20% to 80% of ingested lead can be absorbed by the human body, potentially disrupting normal protein synthesis and leading to various health issues. For example, lead ions can interact with glutathione sulfhydryl groups and other functional groups within cell membranes, increasing oxidative stress levels [8]. Additionally, lead can form chemical bonds with protein sulfhydryl groups, inhibiting the activities of δ-aminolevulinic acid dehydratase, glutathione peroxidase, and protein kinase C [9]. These reactions disrupt the balance of magnesium, calcium, and zinc in the body, impeding cellular communication and neurotransmitter absorption. This disruption can reduce nerve excitability and impair memory storage. Additionally, children are particularly vulnerable to lead exposure during early developmental stages. Evidence from animal models and human genetic research indicates that exposure to lead significantly increases the risk of central nervous system and cognitive disorders in children.

The World Health Organization (WHO) and other organizations in different nations have reached a consensus on the necessity of mitigating lead pollution, leading to the implementation of stringent environmental guidelines for lead usage. For example, the European Union enforces lead mass concentration limits of below 10 μg/L in drinking water and 50–300 mg/kg in soil [10]. Consequently, preventing lead leakage and conducting thorough environmental safety assessments are essential prerequisites for the commercialization of PSCs.

3 | Importance of Lead in PSCs

Among all types of perovskite technologies, lead halide perovskites have demonstrated remarkable success as photoabsorber materials in solar cells, achieving high device performance with a world-record efficiency of 26.7% [1], comparable to Si solar cells. However, the toxicity of lead in perovskites poses a significant challenge for practical applications.

To eliminate the risk of using lead in PSCs, the most effective solution is to replace lead with non-toxic materials in PSCs. Several alternative metal ions, such as Sn^{2+}, Ag^{+}, Bi^{3+}, Sb^{3+}, and Ti^{4+}, have emerged as less toxic options for constructing lead-free PSCs. Among these alternatives, tin (Sn) stands out as the most promising element for lead replacement due to its similar external electronic structure and ionic radius [11, 12]. Furthermore, tin-based perovskites offer the advantage of tunable optical bandgaps within the range of 1.2–1.41 eV, encompassing the highly promising optimal bandgap of 1.34 eV, which

aligns with the highest Shockley–Queisser limit. However, Sn^{2+} materials exhibit poor stability, as they tend to oxidize to Sn^{4+} under ambient air conditions. As a result, Sn-based devices necessitate fabrication within a controlled nitrogen glovebox environment, leading to increased production costs compared to Pb-based PSCs. Recent advancements have improved the efficiency of Sn-based PSCs to 15.38% PCE through perovskite precursor additive engineering [13]. However, the efficiency of Sn-based PSCs still falls behind that of the Pb-based counterparts. An alternative approach to mitigate lead toxicity involves partially replacing lead with tin to create Pb-Sn mixed PSCs. Unfortunately, these Pb-Sn mixed devices exhibit inferior performance and reduced stability compared to Pb-based PSCs.

Additionally, due to its inherent thermodynamic stability, small carrier effective mass, and low exciton binding energy, the Pb-free double perovskite has garnered significant interest as a prospective environmentally friendly photovoltaic material [14]. An innovative approach involves substituting two Pb^{2+} ions with a univalent cation and a trivalent cation to create a tunable double perovskite with the formula $A_2B^{I}B^{III}X_6$. For example, $Cs_2AgBiBr_6$ is a well-established double perovskite composition used in PSCs, demonstrating commendable resistance to moisture, light, and thermal influences. However, the PCE of this double perovskite device remains below 7%, which is significantly lower than that of Pb-based PSCs [15].

Thus, given the challenges in replacing lead to achieve high-efficiency and stable PSCs, encapsulating lead to prevent potential leakage has emerged as an effective strategy to mitigate its environmental and health impacts.

4 | Mechanism of Perovskite Decomposition and Lead Leakage

When perovskite modules endure hail, the intense external mechanical force can induce device cracking. These cracks allow water infiltration into the modules, resulting in perovskite decomposition. Consequently, Pb-containing byproducts may leach into the soil and rivers through rainfall. Perovskite materials exhibit sensitivity to moisture and are susceptible to decomposition because of their hygroscopic characteristics. Previous work reveals that highly polarized H_2O molecules can establish robust hydrogen bonds with organic cations, disrupting the distribution between A-site cations and the $[PbI_6]^{4-}$ octahedral structure. This disruption ultimately results in the irreversible deterioration of the perovskite crystal structure. As shown in Figure 2, the $MAPbI_3$ ($CH_3NH_3PbI_3$) perovskite composition decomposes into PbI_2 and CH_3NH_3I in a humid environment [16]. The CH_3NH_3I can undergo further deprotonation by water molecules to produce HI and CH_3NH_2. Subsequently, exposure of perovskite films to air can trigger the decomposition of HI, resulting in the formation of I_2 and water molecules through a reaction with oxygen, as summarized in Figure 2a. The presence of I_2 adversely affects the degradation process of the perovskite layer. Typically, iodine accelerates the dissociation of water molecules into OH^- and H^+, effectively serving as deprotonated groups for MA^+ ions. Consequently, this detrimental reaction negatively impacts the nucleation and growth of perovskite crystal grains.

In addition to damage from rain or hail, lead leakage from perovskite photovoltaics can occur due to elevated temperatures caused by fires or intense sunlight. Elevated temperatures often result in the collapse and decomposition of the perovskite structure, releasing lead components into the environment. The corresponding decomposition process is illustrated in Figure 2b. For example, under high-temperature conditions, $CH_3NH_3PbI_3$ perovskite undergoes degradation, leading to the formation of PbI_2 and CH_3NH_3I, which subsequently decompose into CH_3NH_2 and HI gas (Figure 2b). In contrast to MA^+, FA^+ ($NH_2CH{=}NH_2^+$) serves as an alternative A-site cation in the perovskite ABX_3 structure. FA^+ ions are larger molecules with smaller dipole moments. This characteristic results in a stronger binding energy between FA^+ and $[PbI_6]^{4-}$ octahedral structure, leading to higher stability in FA-based perovskites due to reduced halide ion migration. However, despite this improved stability, $FAPbI_3$ still undergoes decomposition when the temperature exceeds 230°C, as illustrated in Figure 2c. Additionally, a simulated experiment conducted by Boyen et al. to assess the fire safety of PSCs demonstrated the presence of PbI_2 in the smoke generated during combustion [17]. This finding suggests that lead compounds may disperse into the air, contaminating the surrounding environment along with the smoke.

Besides the aforementioned factors, the degradation process of perovskite materials induced by continuous sunlight, especially under ultraviolet (UV) radiation, cannot be omitted. UV light can trigger the dissociation of organic cations within the perovskite composition, resulting in both perovskite phase collapse and chemical decomposition, as shown in Figure 2b. The PbI_2 in the residual film can undergo decomposition into Pb (0) and I_2 [18]. Consequently, the formation of I_2 under light conditions can induce further degradation of perovskite films, thereby promoting the photo-decomposition reactions with different ions (I^- and $CH_3NH_3^+$) in the perovskite films, resulting in additional I_2 generation. This irreversible process accelerates the precipitation of hazardous lead compounds.

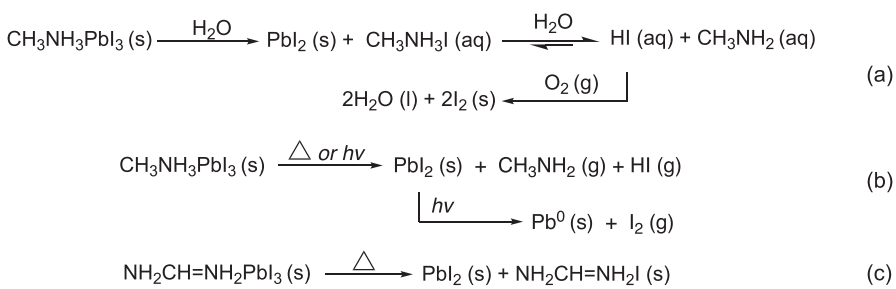

FIGURE 2 | The degradation process of perovskite materials under (a) moisture and (b, c) heat and light conditions.

5 | Lead Encapsulation Materials

While replacing lead in current PSCs remains challenging, effective encapsulation strategies can significantly reduce the risk of lead leakage. Recent advancements utilize external encapsulants that encase entire PSCs, acting as physical barriers to prevent atmospheric penetration and minimize lead leakage. Some of these materials are further enhanced with Pb-chelating agents, offering improved protection by actively binding lead ions. In contrast, internal encapsulation involves materials integrated within the PSC layer stack, which chemically interact with lead in the perovskite to capture it or inhibit ion migration, as illustrated in Figure 3. This section will discuss both external and internal lead encapsulation materials used in PSCs.

5.1 | External Lead Encapsulation Materials

External encapsulation (as illustrated in Figure 3a) is a packing strategy designed to shield PSCs from the surrounding atmosphere, protecting them against harsh environmental conditions and therefore significantly increasing the solar cell's operational stability. More importantly, it also minimizes lead leakage in the event of damage or breakage to the perovskite solar modules.

5.1.1 | Physical Encapsulants

Currently, various types of external physical encapsulants, many of them borrowed from Si and other PV technologies, have been tested on PSCs. Polymers are widely used for external encapsulation in PSCs due to their flexibility, good barrier properties, ease of processing, mechanical protection, chemical compatibility, transparency, cost-effectiveness, and lightweight nature. For example, cross-linked ethylene vinyl acetate (EVA) is a common external encapsulation material for PSCs due to its outstanding elasticity and strong adhesion to glass, which effectively protects PSCs from the external atmosphere. Additionally, the impressive transparency of EVA ensures a negligible impact on solar cell device performance [19]. However, prolonged exposure of EVA in photovoltaic modules to light and heat initiates a degradation process, producing acetic acid and polyenes. The oxidation of conjugated polyene forms polyene and α, β-unsaturated carbonyl groups, causing a red shift in absorption and turning the EVA sheet yellow or brown. This color alteration significantly reduces light absorption and adversely affects device performance.

Substituting EVA with alternative organic polymers, such as polyurethane (PU), poly(p-chloro-xylylene), epoxy, resin, Surlyn, and UV glue, has shown promise in extending the lifespan of PSCs under harsh conditions and passing industrial aging tests, although their effectiveness in capturing lead remains limited [19–21]. For example, studies have demonstrated that encapsulating PSCs with PU can be conducted at relatively mild temperatures and significantly enhance the cells' thermal stability. This has been observed to maintain 97.52% of the initial efficiency even after exposure to outdoor conditions for over 2000 h [19]. Jiang et al. demonstrated that encapsulating the PSCs with epoxy resin enables self-healing when exposed to temperatures exceeding its glass-transition temperature (Tg). Their study revealed that compared to a control device using bottom ultraviolet-cured resin encapsulation, the device with ER encapsulation reduced lead leakage rates by 375 times [20]. Shi et al. [22] introduced a cost-effective polyisobutylene (PIB)–based polymer that allows encapsulated PSCs to successfully pass the rigorous International Electrotechnical Commission (IEC) 61215:2016 tests for damp heat and humidity freeze. These tests evaluate solar cell modules' capability to endure outdoor conditions through cycles of temperature fluctuations (−40°C to 85°C) and exposure to 85% relative humidity.

Additionally, inorganic materials such as aluminum oxide (Al_2O_3), applied via vacuum deposition techniques, are frequently used as encapsulants for PSCs. Previous studies indicate that applying a thin Al_2O_3 layer as an encapsulant on top of PSCs can improve their stability by acting as a barrier against moisture penetration [23–25]. Furthermore, to enhance the flexibility of the inorganic film and improve the smoothness of the PSC surface, a combination of alternative organic layers (FDTS, pV_3D_3) can be incorporated. This unique structure effectively prevents the penetration of water and oxygen, significantly improving the barrier performance of the encapsulation layer and thereby enhancing the long-term stability of the device [26]. However, the deposition of the inorganic layer using atomic layer deposition (ALD) requires elevated temperatures and a long vacuum process, which can potentially damage the PSCs. Additionally, these encapsulants are not suitable for flexible solar cells.

Alternatively, Valastro et al. [27] recently introduced a sustainable method for capturing lead from solar devices to aid in the commercialization of PSC. This approach involves using solvent-free porous TiO_2 films, which are applied physically to devices as the final production step. Applicable to any Pb-containing device, these transparent TiO_2 sponges are deposited at room temperature and can capture lead that leaks from damaged

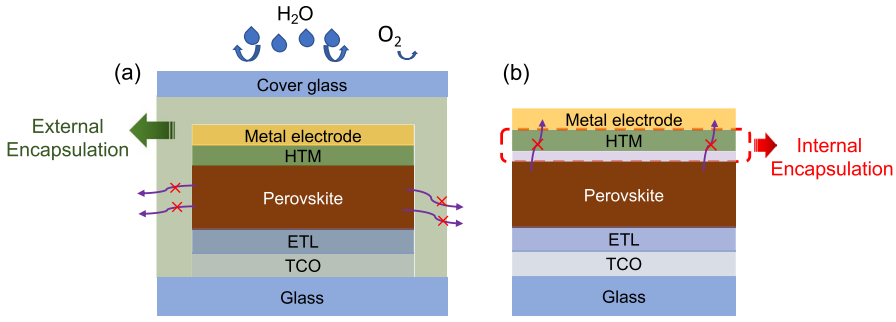

FIGURE 3 | Illustration of PSCs with (a) external and (b) internal encapsulation.

PSCs, preventing environmental release. The TiO_2 sponge can efficiently sequester lead in concentrations ranging from 24 to 63 g/cm^2, equivalent to the amounts contained in $MAPbI_3$ layers with thicknesses from 200 nm in semi-transparent PSCs to 500 nm in opaque PSCs.

5.1.2 | Chemical Encapsulants

To better prevent lead leakage in PSCs, other external lead adsorption encapsulation materials can be employed. These materials, which contain Pb-chelating agents, are applied to the front or back of solar cells to provide effective protection against lead leakage. They have proven to be highly effective in mitigating lead leakage from PSCs under various stress conditions.

For example, Li et al. [28] utilized transparent films like P,P′-di(2-ethylhexyl)methanediphosphonic acid (DMDP) and N,N,N′,N′-ethylenediaminetetrakis(methylenephosphonic acid)-poly(ethylene oxide) (EDTMP-PEO) as external Pb absorption encapsulants. Both DMDP and EDTMP molecules exhibited strong binding energies of 295.6 and 252.6 kJ mol^{-1}, respectively, ensuring a high rate of lead leakage suppression. Comparing these improved encapsulation devices to the original packaging, the lead leakage rate was reduced by over 96%. Importantly, it is noteworthy that the use of DMDP and EDTMP-PEO films had no adverse effects on the photovoltaic performance of PSCs. Later on, they further developed an innovative Scotch-tape-like design utilizing Pb^{2+} absorbents by combining a standard EVA film with a pre-laminated layer of DMDP [29]. This design allows the tapes to be applied to both sides of PSCs in either n-i-p or p-i-n configurations during the standard encapsulation process. To make the tape, they spread a solution containing DMDP on EVA film and allowed it to dry. After mild heating, the DMDP-laminated EVA tape becomes transparent and adheres tightly to the glass surface without affecting the photovoltaic performance. On the backside, the tape binds securely to the metal electrodes. The DMDP layer on the glass side is 5.7 μm thick, which can absorb up to 3.11×10^{-4} g/cm^2 of Pb^{2+}, more than three times the Pb^{2+} present in the PSCs (8.07×10^{-5} g/cm^2). For the back metal electrode, a 10-μm–thick DMDP layer is used, which can absorb 6 times the total Pb^{2+} amount in the device. These Pb^{2+}-absorbing tapes demonstrated an impressive sequestration efficiency (SQE) of over 99.9% in water over a period of 7 days, and they maintained their effectiveness even after 3 months of outdoor exposure and severe physical damage. This method ensures that Pb^{2+} levels remain below the EPA drinking water standard and can be seamlessly integrated with existing encapsulation materials.

Recently, Wang et al. [30] synthesized a cross-linked fluoropropyl methylsiloxane-dimethylsiloxane multiblock polymer (CFDP, synthesis route shown in Figure 4a) with excellent transparency, UV stability, barrier properties, adhesion, and thermal stability. The CFDP encapsulation provided superior heat dissipation compared to typical UV resin encapsulation, preventing performance degradation due to heat accumulation. This was demonstrated by heating perovskite samples to approximately 90°C on a hot plate and then quickly transferring them to a platform at 25°C, with temperature changes tracked using an infrared camera (Figure 4b,c). The CFDP encapsulation showed better heat dissipation than UV resin and POE encapsulations, highlighting its effectiveness as a heat dissipation layer in the target encapsulation strategy for perovskite films. As a result, the devices retained 98% and 95% of their initial efficiency in damp heat and thermal cycling tests, respectively, complying with IEC 61215 standards. Moreover, the flexible encapsulation layer and the strong coordination interaction of C=O groups with Pb^{2+} significantly reduced lead leakage from damaged devices in rain and immersion tests. This encapsulation strategy optimizes thermal management, enhances device stability, and suppresses lead leakage, achieving efficient and sustainable PSCs.

Another study from Dipta et al. [31] introduced a versatile nanoencapsulation platform using natural polyphenols for Pb-halide PSCs. The researchers applied a polyphenol-based coating to the rear electrode of the PSC device (Figure 4d), utilizing tannic acid (TA), a natural polyphenol, and titanium ions (Ti^{4+}) as the cross-linking agent (Figure 4e). This approach builds upon previous work involving a gel system composed of TA and group IV transition metals, known as metal–phenolic gels [32]. Unlike other polyphenol-based coatings that use Fe^{3+} or Cu^{2+} under aqueous conditions, the metal–phenolic gel system is solvent independent, can incorporate additional functional compounds, and exhibits high mechanical stability due to the strong bonding between TA's catechol groups and Ti^{4+} ions. Furthermore, by controlling the gelation time, the initial mixture can be transformed into thin films via spin coating. The encapsulated devices achieved a PCE of 20.7%, retaining up to 80% of their peak performance over 2000 h and up to 70% over 7000 h. Additionally, under simulated rainfall conditions, the catechol-rich encapsulant effectively captures Pb ions released from degraded perovskites, maintaining lead levels below the safe drinking water threshold of 15 ppb.

Overall, the integration of external functional chemical materials presents a promising approach to mitigate lead leakage. Beyond their effective lead absorption capabilities, combining these materials with other encapsulation agents could significantly enhance the durability of PSCs against moisture, elevated temperatures, UV light exposure, and other harsh conditions. However, it is crucial to conduct rigorous long-term stability assessments of these small molecular materials and carefully evaluate potential additional manufacturing costs for the future commercialization of PSCs.

5.2 | Internal Lead Encapsulation

In addition to external encapsulation methods, internal encapsulation strategies have also been developed to sequestrate or bond mobile lead ions within the bulk (e.g., grain boundaries) or interfaces of PSCs to prevent lead migration, as illustrated in Figure 3b. Various encapsulants, including functional hole-transporting materials (HTMs), conjugated metal–organic frameworks (MOFs), organic polymers, and some small molecules, have been applied to the surface or within the perovskite absorbing layer. These will be summarized in this section. The properties of these materials show strong binding interactions with lead and are capable of minimizing lead diffusion across the interface, capturing mobile ions and preventing them from migrating to the external environment. Beyond

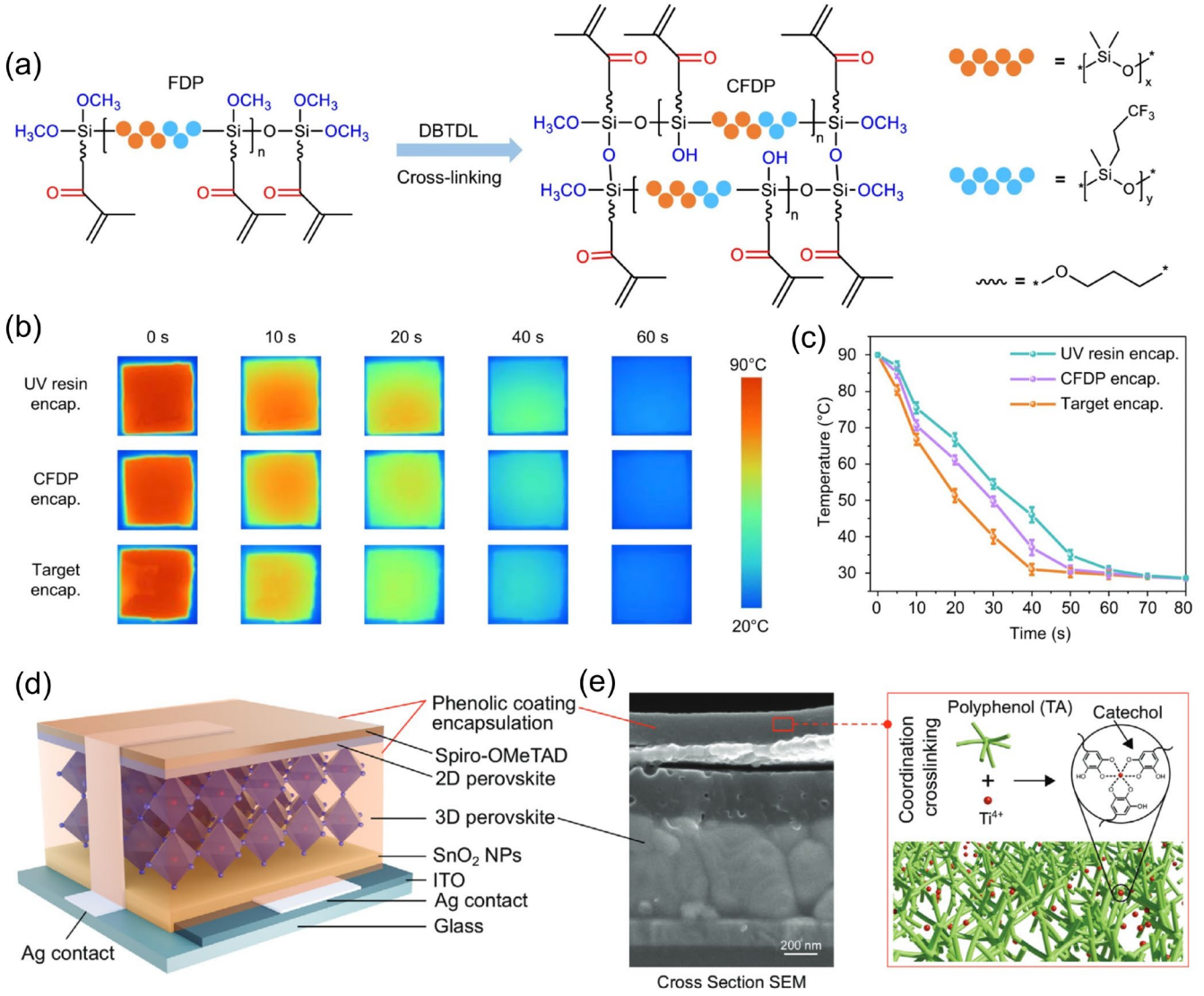

FIGURE 4 | (a) The synthetic route of polymer gel CFDP. (b) IR thermal images of encapsulated perovskite films during a cooling test. (c) The surface temperature change of encapsulated perovskite films versus time. The error bars represent the standard deviation for three samples. (d) A 3D schematic illustration of the full device with the polyphenol encapsulant. (e) Cross-sectional SEM image of the encapsulated device and the molecular view of the encapsulant, cross-linked networks of the TA-Ti^{4+} coating, where catechol groups are present in TA. Adapted and reproduced with permission from ref. [30, 31].

simply encapsulating lead ions, these encapsulants also passivate perovskite defects and suppress nonradiative recombination, thereby enhancing device performance and inhibiting the perovskite degradation internally.

5.2.1 | Functional HTMs

In n-i-p–structured PSCs, HTMs are applied on top of the perovskite layer. In addition to their primary role in hole selection and transport, ideal HTMs should also exhibit robust structural and chemical stability, as well as hydrophobic properties, to protect the perovskite layer from exposure to the external environment. Additionally, when HTMs contain lead-coordinating groups, they can further function as internal encapsulants.

For example, a novel donor–acceptor polymer, alkoxy-PTEG (structure shown in Figure 5a), composed of benzo[1,2-b:4,5:*b′*] dithiophene and tetraethylene glycol (TEG)-substituted 2,1,3-benzothiadiazole, has been introduced as HTM in PSCs [33]. Researchers found that alkoxy-PTEG exhibits high solubility even in non-aromatic solvents like 3-methylcyclohexanone (3-MC) and can prevent potential lead leakage through chelation. Nuclear magnetic resonance (NMR) studies reveal that TEG groups can chelate lead ions with moderate strength ($K_{binding} = 2.76$), a level considered non-destructive to the perovskite lattice, thus preventing lead leakage. Xu et al. [34] proposed an internal encapsulating layer (IEL) based on an in situ cross-linked insoluble polymer called Spiro-NPU (synthesis route shown in Figure 5b). This polymer is formed from Spiro-OH (2,2′,2″,2‴-(((9,9′-spirobi[fluorene]-2,2′,7,7′ tetrayltetrakis(phenylazanediyl))tetrakis(ethane-2,1-diyl))tetrakis(oxy))tetrakis(etan-1-ol)) and 1,5-naphthalene diisocyanate (NDI) monomers. The IEL enhances moisture resistance and lead blockage owing to the spiro block and NCO and carbonyl groups. In situ cross-linked polymers are particularly advantageous due to their better diffusion of small molecules compared to

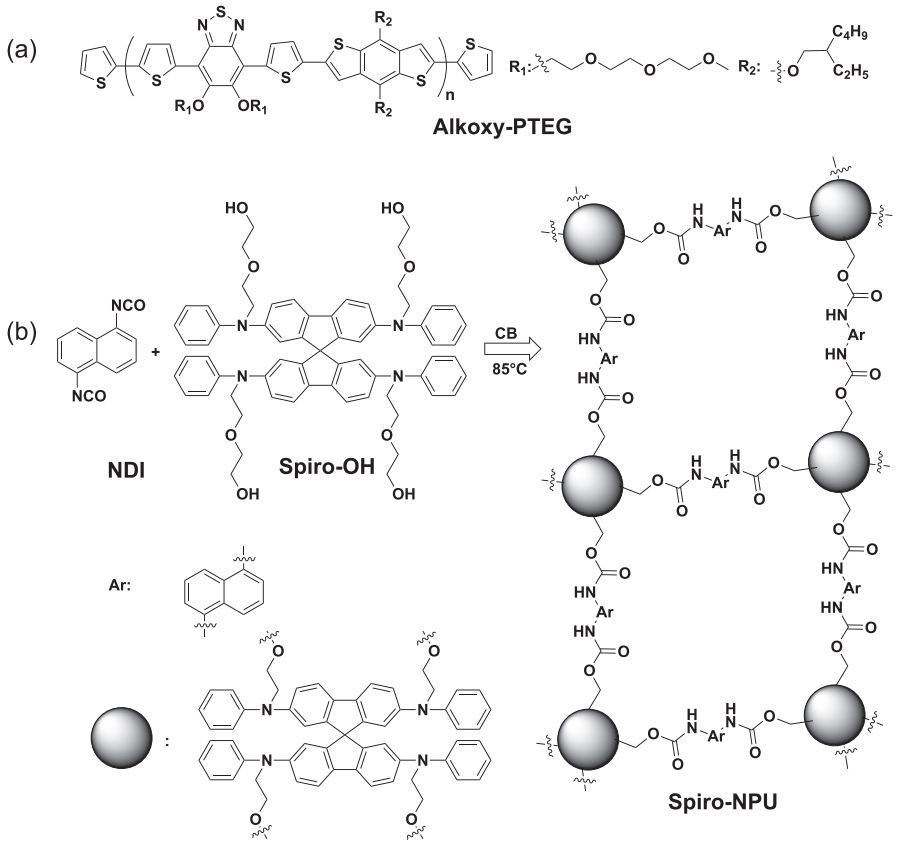

FIGURE 5 | (a) Chemical structures of alkoxy-PTEG. (b) The synthesis of Spiro-NPU.

high–molecular-weight polymers, higher morphology reproducibility via in situ polymerization, and stronger moisture-blocking capabilities. As a result, the Spiro-NPU IEL effectively traps nearly all Pb^{2+} ions within a confined area through strong Pb—O interactions, significantly enhancing lead blockage.

5.2.2 | MOFs

MOFs offer a promising approach for enhancing the performance and stability of PSCs. Due to their tunable porosity and functionalization, MOFs can be employed in PSCs as scaffolds for improved charge transport, as well as for passivating defects at the perovskite interfaces. Their high surface area and customizable pore structures allow for the efficient incorporation of functional groups that can chelate lead ions, thereby reducing lead leakage.

For example, Wu et al. [35] utilized a thiol-functionalized 2D MOF as an electrode extraction layer placed atop the perovskite layer to reduce lead diffusion into the environment. The robust Pb—S bonding interactions in the MOF generated a water-insoluble lead compound, enhancing its lead absorption capacity. This resulted in the capture of over 80% of lead during an acid rain lead leakage test. Additionally, these MOFs exhibited n-type semiconductor properties with well-aligned energy levels, facilitating efficient carrier transport. Consequently, the device incorporating the thiol-functionalized MOF achieved an impressive PCE of 22.02% and demonstrated significantly improved stability.

Huckaba et al. [36] investigated the use of FeBTC/PDA, a porous MOF–polymer composite, in perovskite solar panels. This material, which combines a porous MOF scaffold with a polymeric metal-binding agent, effectively sequesters lead from various solutions, including simulated perovskite solutions, solutions with decomposed perovskite thin films, and real-world solutions from damaged solar cells across a range of temperatures and conditions. The proposed lead adsorption mechanism, featuring representative chelating moieties of the polymer and FeBTC/PDA, is illustrated in Figure 6a.

5.2.3 | Polymers

Polymers are increasingly employed in PSCs for internal encapsulation due to their versatility and effectiveness in enhancing device stability. By integrating polymers into the perovskite layer or as interfacial coatings, researchers can create robust barriers that protect against environmental degradation and moisture infiltration. Polymers can be engineered to include functional groups that interact strongly with lead ions, reducing lead leakage and improving safety. Additionally, their ability to form conformal, flexible layers allows them to cover complex surfaces and interfaces within the device. The use of polymers for internal encapsulation not only helps maintain the integrity and performance of PSCs but also contributes to the overall longevity of the devices by minimizing the impact of external stresses and chemical degradation.

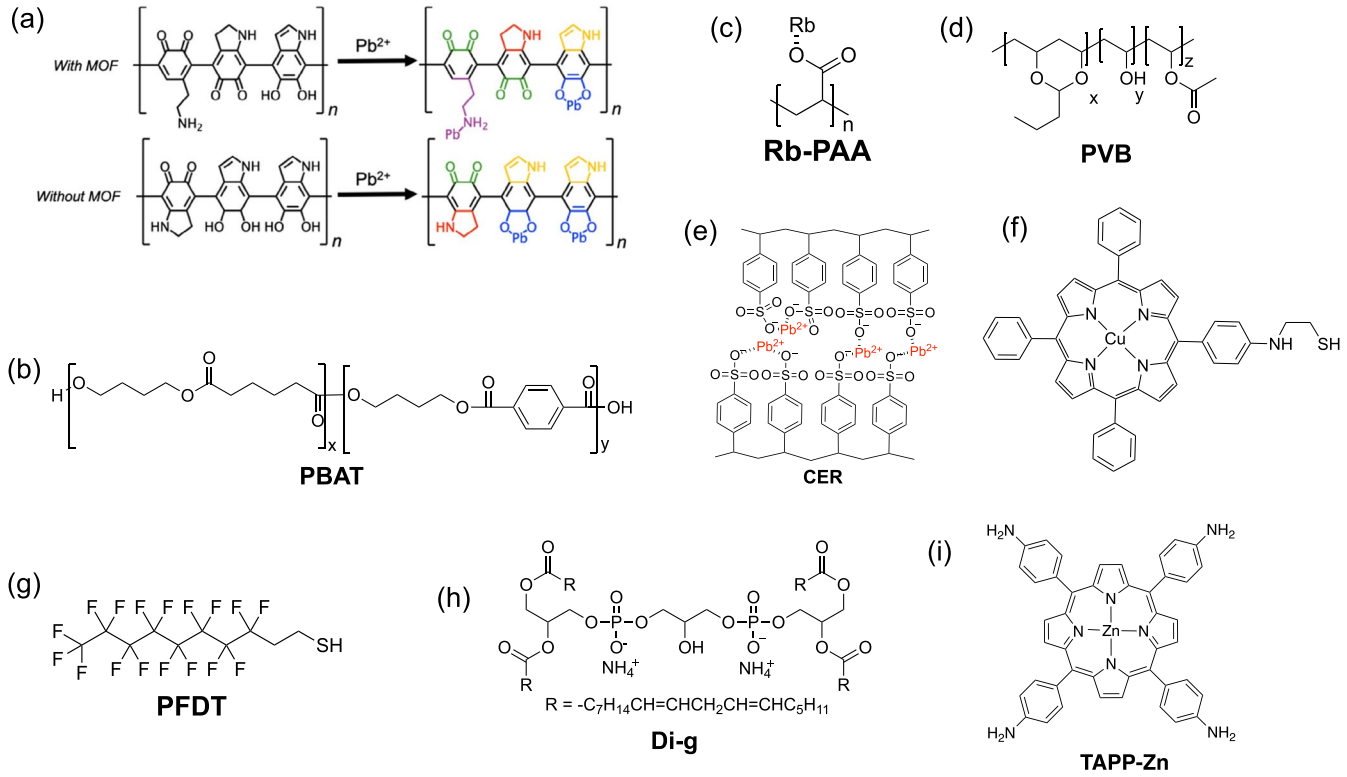

FIGURE 6 | (a) The proposed adsorption mechanism showing representative chelating moieties for the polymer and FeBTC/PDA. Chemical structures of (b) PBAT, (c) Rb-PAA, (d) PVB, (e) CER, (f) thiol-functionalized perfluoroalkyl molecule, (g) PFDT, (h) Di-g, (i) TAPP-Zn. Adapted and reproduced with permission from ref. [36].

For example, Cao et al. [37] incorporated an eco-friendly poly(butylene adipate-co-terephthalate) (PBAT, Figure 6b) polymer into perovskite films, utilizing its carbonyl groups and benzene rings to passivate uncoordinated lead cations and neutral iodine defects. This addition effectively controls the crystallization process, leading to reduced trap density, suppressed nonradiative recombination, and improved charge carrier transport. Importantly, the PBAT polymer network interacts strongly with lead cations, preventing them from dissolving in water and reducing Pb^{2+} leakage by nearly 98% when the polymer-coated perovskite film is submerged. Zhao et al. [38] developed a rubidium (Rb)-functionalized poly(acrylic acid) (Rb-PAA, Figure 6c) to regulate the crystallization of $FAPbI_3$ perovskite films. Their study revealed that the combined effects of the alkali cations and poly(acrylic acid) anions effectively passivate defects on the surface and grain boundaries of the perovskite films. Consequently, the Rb-functionalized poly(acrylic acid) significantly enhances the PCE of $FAPbI_3$ PSCs, while the strong interaction between PAA ions and lead cations effectively prevents lead leakage.

Recently, a convenient internal encapsulation strategy was proposed by Dong et al. [39], involving the introduction of a multifunctional polyvinyl butyral (PVB, Figure 6d) polymer at the perovskite/electron transport layer interface. This PVB layer is pivotal in reducing ion migration, optimizing energy levels, and preventing moisture infiltration. In addition to improving device efficiency and stability, the incorporation of this material effectively reduces lead leakage by forming strong interactions with lead cations in the perovskite films.

Additionally, Huang's group [40] reported an abundant, low-cost, and chemically robust cation-exchange resin (CER, Figure 6e) as an external encapsulation material to prevent lead leakage from damaged PSCs under severe weather conditions. CERs demonstrate both a high adsorption capacity and rate for lead in water, attributed to their strong binding energy with lead ions within the mesoporous structure. Integration of CERs with carbon electrodes and layering them onto the glass surface of modules has minimal impact on device efficiency while reducing lead leakage from perovskite mini-modules by 62-fold to 14.3 ppb in water. Simulations indicate that lead leakage from damaged large-area perovskite solar panels treated with CERs can be further reduced to below 7.0 ppb, even under the worst-case scenario of every sub-module being damaged.

5.2.4 | Other Small Molecules

Other small molecules have been extensively used as additives or surface passivation materials to address defects within the bulk or on the surface of perovskites, thereby improving the efficiency and stability of PSCs. Additionally, some small molecules containing lead-coordinating functional groups have been explored as internal encapsulants. For instance, Li et al. [41] developed a copper porphyrin (Figure 6f) containing thiol and secondary amine groups as a passivation material. The incorporation of this stable porphyrin molecule increased the device efficiency to 21.24%. Moreover, they found that the strong interaction between the thiols and lead cations can effectively mitigate lead compound leakage in perovskite. Zhang et al. [42]

designed superhydrophobic surfaces using thiol-functionalized perfluoroalkyl molecules (Figure 6g) to chemically modify the lead halide perovskite film and metal electrode through a vapor-assisted self-assembly process. Due to their passivation effect and hydrophobic properties, the resulting inverted PSCs achieved a high PCE of 21.79% and demonstrated significantly improved long-term stability. Notably, the dense and stable PFDT array demonstrated a strong synergy in substantially reducing lead leakage from degraded PSCs. The lead concentration in the control devices stored in soaked water reached 12.23 mg/mL at room temperature after 200 min and increased further when heated to 65°C. In contrast, the treated devices exhibited a reduced concentration of 2.05 mg/mL.

Meng et al. [43] introduced diphosphatidylglycerol (Di-g, Figure 6h) as a surface passivation material for flexible perovskite films. Di-g effectively optimized surface defects and trap states, enhancing device efficiency to 20.29% while providing excellent environmental and mechanical stability. Its amphipathic nature allows it to bind tightly to divalent lead ions and offer interfacial water resistance, resulting in a 96% reduction in lead leakage compared to reference films.

To address the issue of lead leakage in narrow bandgap Pb—Sn perovskite compositions, widely used in high-efficiency perovskite tandem solar cells, Liu et al. [44] incorporated 5,10,15,20-Tetrakis-(4-aminophenyl)-porphine-Zn(II) (TAPP-Zn, shown in Figure 6i) into the Sn-Pb perovskite layer. This material forms strong coordination interactions with Pb^{2+}, which modulate crystallization kinetics and promote the formation of a Pb-rich surface. Additionally, the introduction of TAPP-Zn helps passivate defects and enhances charge separation. As a result, the modification enabled a 23.28% efficiency in single-junction PSCs and a 27.03% efficiency in tandem PSCs. Furthermore, TAPP-Zn acts as an internal encapsulant, effectively preventing lead leakage and reducing the Pb leakage rate to a significantly low level of 10.5 mg m^{-2} h^{-1}. Even in severely damaged PSCs, Pb sequestration efficiency remained as high as ≈99%.

6 | Lead Recycling

As discussed above, encapsulating PSCs has proven effective in preventing lead leakage from degraded or damaged cells, which could facilitate the widespread application of perovskite solar technology. Once perovskite PVs reach global commercial scale, thousands of tons of lead compounds will be required to produce efficient and stable modules [45]. The responsible disposal of end-of-life perovskite modules emerges as a critical concern, posing potential harm to the ecosystem and impeding further perovskite PV applications. Consequently, the development of sustainable lead recycling technologies to avoid the production of potential lead waste becomes imperative. The investigation of these technologies will further mitigate lead toxicity concerns by establishing sustainable approaches alongside lead encapsulation methods. Additionally, the recycled lead compounds can be reused to manufacture new perovskite modules, addressing environmental issues while also reducing raw material costs, thereby increasing competitiveness in the photovoltaic market.

Chemical dissolution–precipitation is one of the common strategies to recycle and purify lead compounds from PSCs. This method involves dissolving the perovskite layer in an appropriate solvent, followed by chemical treatment to precipitate lead as a solid compound, such as PbI_2 and $Pb(OH)_2$. The advantages of this approach include its relative simplicity and the ability to achieve high recovery rates. For instance, Zhang et al. [46] used N,N-dimethylformamide (DMF) to dissolve PSCs and obtain a lead-containing lixivium. $NH_3{\cdot}H_2O$ was then added as a precipitant to extract Pb ions from the lead-containing lixivium, leading to the formation of $Pb(OH)_2$ precipitate with a 99.9% extraction rate. Subsequently, the addition of HI as a purifying agent reacted with $Pb(OH)_2$, producing pure PbI_2. The recycling process is illustrated in Figure 7a. Schmidt et al. [47] used hot water to extract lead compounds from PSCs. Upon cooling the extractant, crystalline PbI_2 precipitated with high yield and good purity, as illustrated in Figure 7b. Additionally, due to the coordination interaction between amine and Pb^{2+}, Feng et al. [48] developed butylamine as the solvent to exact lead compounds. The formed coordinated intermediates can be easily purified and recrystallized to pure $MAPbI_3$ crystal to fabricate new PSCs, thus completing a closed-loop recycling process. Recently, Wu et al. [49] employed an anti-solvent, ethyl acetate, to recycle $MAPbI_3$ from a GBL (γ-butyrolactone) solution. From this method, 87% lead compounds have been recycled, and the recycled $MAPbI_3$ was reused to fabricate PSCs, which showed comparable performance in current–voltage measurements to those fabricated using new materials.

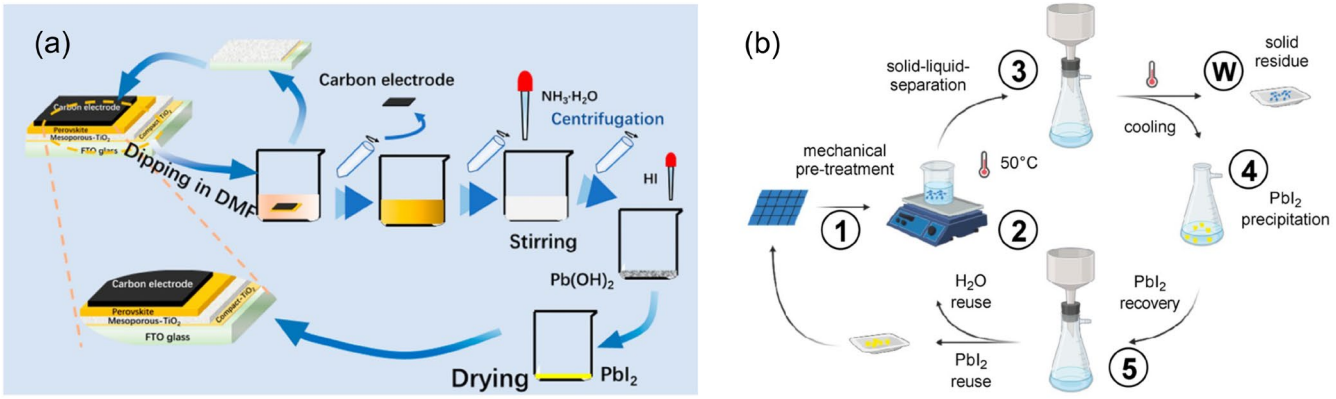

FIGURE 7 | (a) Illustrated cyclic utilization process of lead from carbon-based PSCs with $NH_3{\cdot}H_2O$. Adapted with permission from ref. [46]. (b) Closed-loop lead recycling process with hot water. Adapted with permission from ref. [47].

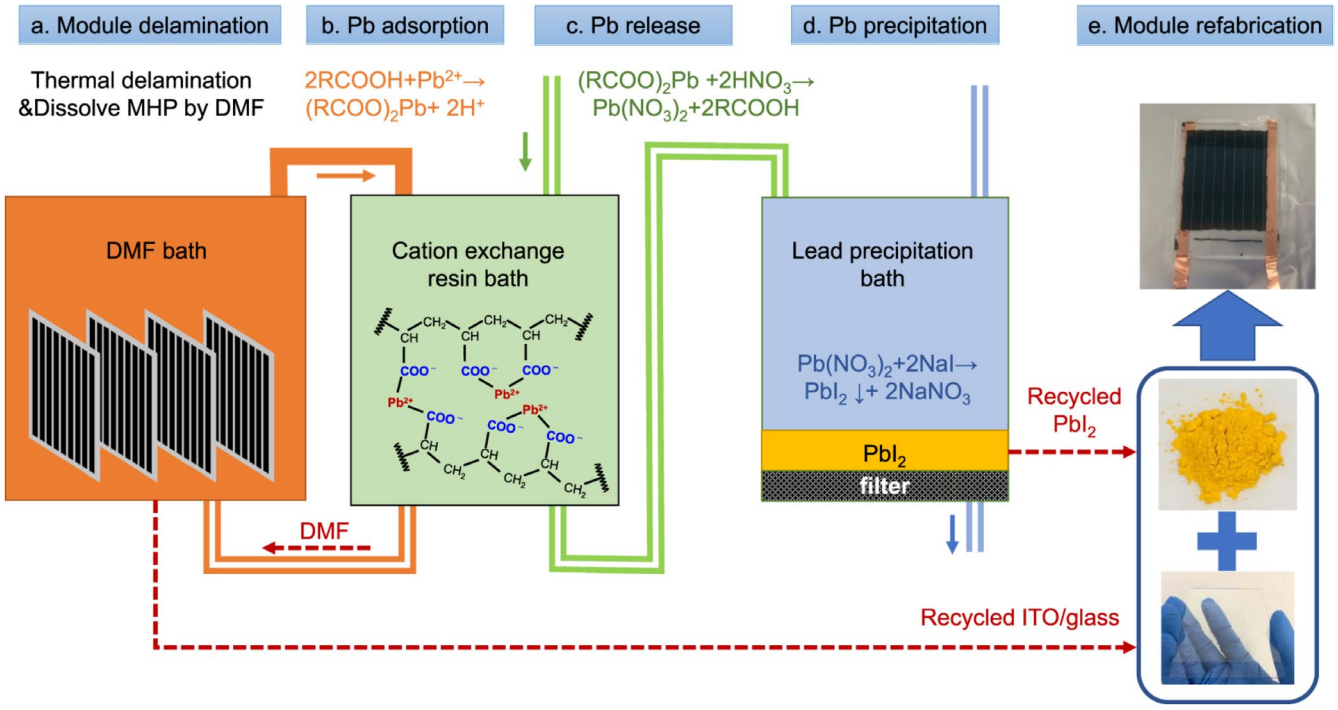

FIGURE 8 | Illustration of the recycling process from encapsulated perovskite solar modules. (a) Encapsulated perovskite solar modules were delaminated, and MHP was dissolved in DMF. (b) Lead ions in DMF were removed by carboxylic acid cation-exchange resin. (c) The adsorbed lead ions on resin were released to the aqueous solution by resin-regeneration process via HNO_3. (d) Precipitation of PbI_2 by pouring NaI into $Pb(NO_3)_2$ containing solution. (e) Module refabrication based on recycled materials. Adapted with permission from ref. [7].

Additionally, various lead absorbents have also been investigated to separate and recycle lead compounds [50–52]. The strong coordination interactions with lead cations efficiently absorb lead compounds within the materials. After cation exchange or other processes, recycled lead compounds are separated from the solution. For example, Chen et al. [7] realized a lead recycling process with an acidic cation exchange resin, as illustrated in Figure 8. The interaction between Pb^{2+} and functional groups in the resin facilitates lead separation by displacing H^+ ions. The isolated lead compound is subsequently redissolved in aqueous nitric acid (HNO_3) to yield soluble lead nitrate, $Pb(NO_3)_2$, which, upon the introduction of sodium iodide (NaI), undergoes precipitation to form lead iodide. By the employment of CER, the total lead recycling efficiency can reach up to 99%. More importantly, the PSCs fabricated from the recycled PbI_2 show photovoltaic performance comparable to devices crafted from fresh raw materials. Moreover, Park et al. successfully synthesized and utilized a new iron-incorporated hydroxyapatite with excellent lead absorption capacity, attributed to its negatively charged surface. Following the recycling process, 99.97% of the lead was obtained to produce lead iodide [51].

A cross-linking supramolecular complex composed of 2-hydroxypropyl β-cyclodextrin (HPβCD) and 1,2,3,4-butane tetracarboxylic acid (BTCA) has been explored as a precursor additive for fabricating high-efficiency PSCs [52]. This HPβCD-BTCA complex effectively mitigates lead leakage from severely damaged PSCs, which retain 97% of their initial efficiency after 522 h of dynamic water scouring, with less than 14 ppb of lead contamination in the water. Additionally, the exceptional lead-capturing ability of the HPβCD-BTCA complex makes it a promising candidate for recycling lead iodide complexes from end-of-life devices. ICP-MS measurements show that the recycled PbI_2 derived from these composites has a purity of approximately 98.9%. New PSCs fabricated with 2.5 wt% of the recycled Pb-containing additives, or using recycled PbI_2 as raw material (instead of commercially purchased PbI_2), exhibited PCEs approaching 20%.

It is worth mentioning that during the lead recycling process, other materials such as conductive glass substrates, organic molecules, and metal electrodes can also be recycled. This not only reduces the cost of manufacturing new PSCs but also promotes the circular utilization of various materials, minimizing the production of different wastes. This, in turn, expedites the progress of clean and renewable perovskite photovoltaic technologies.

7 | Summary and Future Outlook

7.1 | Summary

Addressing lead leakage to mitigate the toxic lead concerns in PSCs is crucial for enhancing their stability and environmental safety. This review highlights recent advancements in both lead encapsulation and lead recycling strategies.

External encapsulation focuses on protecting the entire PSC through physical or chemical barriers. Physical encapsulants act as shields, safeguarding the PSCs from environmental exposure and minimizing lead leakage in the event of damage or breakage. However, these materials face limitations, such as saturation from metal cations in rainwater, damage from UV radiation, and insufficient protection when the devices are

compromised. To overcome these challenges, there is a growing need for chemical encapsulants that offer high lead-adsorption capacity and durability against environmental stresses. Some advanced materials incorporate Pb-chelating agents, which actively bind lead ions and provide additional protection against lead leakage in damaged or broken cells. Internal encapsulation strategies involve embedding materials within the perovskite layer or at the device interfaces to sequester lead ions and prevent ion migration. Techniques utilizing functional HTMs, polymers, MOFs, and small molecules with lead-coordinating groups have demonstrated effectiveness in reducing lead leakage and enhancing device stability. These internal encapsulants improve the perovskite's resistance to degradation while actively capturing lead ions, addressing both stability and sustainability concerns. The integration of internal and external encapsulation strategies offers a comprehensive approach to mitigating lead leakage, with each method contributing to the overall sustainability and performance of PSCs.

In addition to encapsulation, lead recycling technology is essential for further reducing the toxic effects of lead in end-of-life perovskite devices and modules. Efficient methods have been explored for recycling lead compounds and reusing them in the production of new PSCs. These recycling strategies not only extend the life cycle of PSCs but also lower manufacturing costs, thereby accelerating the commercialization of perovskite photovoltaics.

7.2 | Future Outlook

To overcome the limitations of current encapsulation methods, future research should focus on developing materials that address both internal and external challenges effectively.

For external encapsulation, there is a need for materials that offer enhanced resistance to UV radiation, mechanical abrasion, and saturation from environmental contaminants. These materials should provide long-term protection and maintain their Pb-adsorption capabilities over extended periods.

For internal encapsulation, advancing the design and synthesis of materials with superior lead-binding properties and improved stability is crucial. Researchers should explore new polymers, MOFs, and small molecules that can better integrate with perovskite layers and effectively capture lead ions without compromising device performance. Additionally, optimizing the application and deposition methods to ensure consistent and effective coverage will be important.

Integrating internal and external encapsulation strategies could provide a comprehensive solution, with internal methods preventing lead migration within the device and external methods mitigating lead leakage from damaged cells. The development of industry standards, rigorous maintenance protocols, and recycling programs will also be essential to ensure the safe deployment of PSCs.

Additionally, a critical component of the future outlook is the establishment of efficient lead recycling processes. As PSCs move toward commercialization, the management of lead waste will be vital. Developing recycling systems that can effectively recover lead from end-of-life PSCs will mitigate environmental risks and reduce the need for virgin lead, supporting a more circular economy in solar technology. Research into low-energy and cost-effective lead extraction and purification methods from perovskite modules will be necessary. These recycling programs should be integrated into the manufacturing and deployment pipelines to ensure the sustainable use of lead and other valuable materials in PSCs.

Moreover, as the perovskite photovoltaic sector grows, strengthening policy formulation and industry standards around recycling, reuse, and environmental protection will be essential. Establishing clear standards for recycling and treating perovskite photovoltaic waste, ensuring that discarded components meet environmental protection requirements, and defining the recycling responsibilities of manufacturers across the product life cycle can all help minimize the environmental impact associated with PSC production and disposal. These standards and protocols should be designed to address both the production and operational phases of PSCs to safeguard environmental health and optimize resource utilization.

Overall, continued innovation and optimization of encapsulation techniques, combined with robust lead recycling strategies and supportive regulatory frameworks, will be vital in addressing the environmental and performance challenges associated with PSCs. This holistic approach will pave the way for their sustainable and widespread adoption in the renewable energy sector.

Author Contributions

Original draft preperation, J.S. and B.Y.; Supervision, B.Y.; Draft revision, H.P. All authors have read and commented on the manuscript.

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Conflicts of Interest

Henrik Pettersson is the CEO of Dyenamo AB.

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