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Recent Advances in Suppressing Sn^{2+} Oxidation in Sn–Pb-based Perovskite Solar Cells

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Abstract. Tin-lead (Sn-Pb) perovskite, a new solar energy material with superior energy-related characteristics, is the material of choice for the next generation of high-efficiency solar panels. Its single big flaw, however, is that the tin is unstable and readily "rusts" or oxidizes, a process that causes defects, which harms the electrical characteristics, constitutes waste of energy, reduces the power output of the solar cell and speeds up its degradation. The answer to this problem is essential. The purpose of this review paper is to survey the various latest ways of inhibiting this tin oxidation. To this end, first the paper discusses the chemical reasons for the instability of the tin. It then goes through the latest approaches in which there are three major approaches: changing the initial formulation of the chemicals so as to offer protection to the tin from the start, addition of some special protecting substances which will act as antioxidants to prevent against or overcome the defects, and lastly the strengthening of the union between the layers making up the solar cell. In analyzing these, it is expected that the problems lying ahead will be brought into sharp focus and this will indicate the field of future research on tin-lead perovskite solar cells, leading to the production of stable high efficiency (Sn-Pb) perovskite solar cells.

Keywords: Perovskite solar cells; Sn–Pb perovskite; Sn^{2+}oxidation; Interface engineering.

1. Introduction

The transition to an environmentally and economically sustainable energy system worldwide is essential and urgent owing to increased demand for energy consumption and climate change [1]. Solar energy is one of the most important components of this transition. Although commercial crystalline silicon photovoltaics (PVs) are among the most efficient energy-converting devices available and currently possess efficiencies of nearly the theoretical Shockley–Queisser efficiency (~29.4 %), their relatively high manufacturing and production costs encourage the search for the next generation of high efficient energy-converting devices which possess properties with economically feasible manufacturing methods [2].

This next generation of energy-converting devices in the form of metal halide perovskite solar cells (PSCs) seems to be one of the most promising options available and has gone from an efficiency of 3.8 % in 2009 as the first report to over 26 % in 2020 [3]. The flexibility in composition of the perovskites is a key property, because a wide range of bandgaps may be synthesized, e.g., ~1.2 to 2.3 eV, and this degree of variability in bandgap may economically optimize light absorption and allow for the manufacture of tandem devices with Si or with other perovskites for improved utilization of the solar spectrum.

From the viewpoint of materials, mixed tin-lead (Sn-Pb) perovskites have an interesting profile owing to their very low bandgap of (1.18 - 1.30 eV) suitable for all-perovskite tandem configurations as the most efficient lower absorber layer able to harvest the near infrared which will not be able to be harvested by the wider band gap upper cell. These single junction tin-lead PSCs have recently shown to be achieving efficiencies of over 21% whilst all-perovskite tandem cells have exceeded 25%, clear indications that these materials have potential and a bright commercial future. However, there is here a clear bottleneck in overcoming the problem of instability linked with self-oxidation of Sn^{2+} to Sn^{4+}, an easy enough thermodynamically viable process accelerated by oxygen, humidity and light. Even in inert atmospheres the residual oxidants in precursor solutions and interfaces are such that they

promote the universal degradation associated with Sn-Pb PSCs. This oxidation process is easy and causes deep trap states associated with the irremediable p-type self-doping effect and this leads to an increased rate of non-radiative recombination occurring. This oxidation leads to a vacancy deficiency of Sn as well as liability to excess holes leading to the corresponding phenomena associated with self-doping plus oxidation of Sn^{2+} leading to a remarkable degradation of open circuit voltage in subsequent efficiencies as well as fill factors and long-term stability. It would thus seem that central to this Sn-Pb based PSC commercial viability is evasion of self-oxidation of Sn^{2+}.

This review addresses the sources and consequences of oxidation of Sn^{2+} in Sn-Pb-based PSCs. We summarize the various strategies adopted in the literature to solve this problem: precursor design, additive development and device design, which are related to the degradation path of the material. The aim of this review is to give guidance to future work in order to find ways of achieving higher efficiency and stability in Sn-Pb PSCs.

2. Sn^{2+}/Sn^{4+} Oxidation Mechanisms and Impacts

2.1. Thermodynamic Drivers and Multifold Pathways of Sn^{2+}Oxidation

The inherent instability of Sn^{2+} arises from its electronic structure. In contrast to lead, tin does not experience the stabilization of the energy of the 6s-orbital arising from the lanthanide contractions which in Pb is a significant energy lowering. As a consequence, Sn has a higher energy of orbital, lower electronegativity, and less s-p splitting and hence a much-reduced ionization potential a consequence of which the energy of ionization of the 5s electrons is thereby favorable [9]. This is reflected in the standard reduction potentials of Sn^{4+}/Sn^{2+} (+0.15 V) which make Sn^{2+}extremely susceptible to further oxidation on the part of even small quantities of oxygen [10].

At such predisposition oxidation of Sn^{2+} proceeds by complicated interrelated processes in a self-catalytic cycle [11]. This includes oxidation by oxygen directly to halogen vacant sites, an I_2 mediated cycle (I_2 cycle) when hydrolysis of SnI_4 ($SnI_5 + 2H_2O \rightarrow SnO_2 + 4HI$) liberates HI which is oxidized to I_2 again acts as a vigorous oxidant, solvent induced oxidation (e.g. by DMSO to the acid solutions of the precursor), photoxidation wherein oxygen appropriates photo-excited electrons to form vigorous superoxydes, (O_2^-) etc. These factors stressed the polymorphic and auto-catalytic nature of the Sn^{2+}degradation as shown in Figure 1.

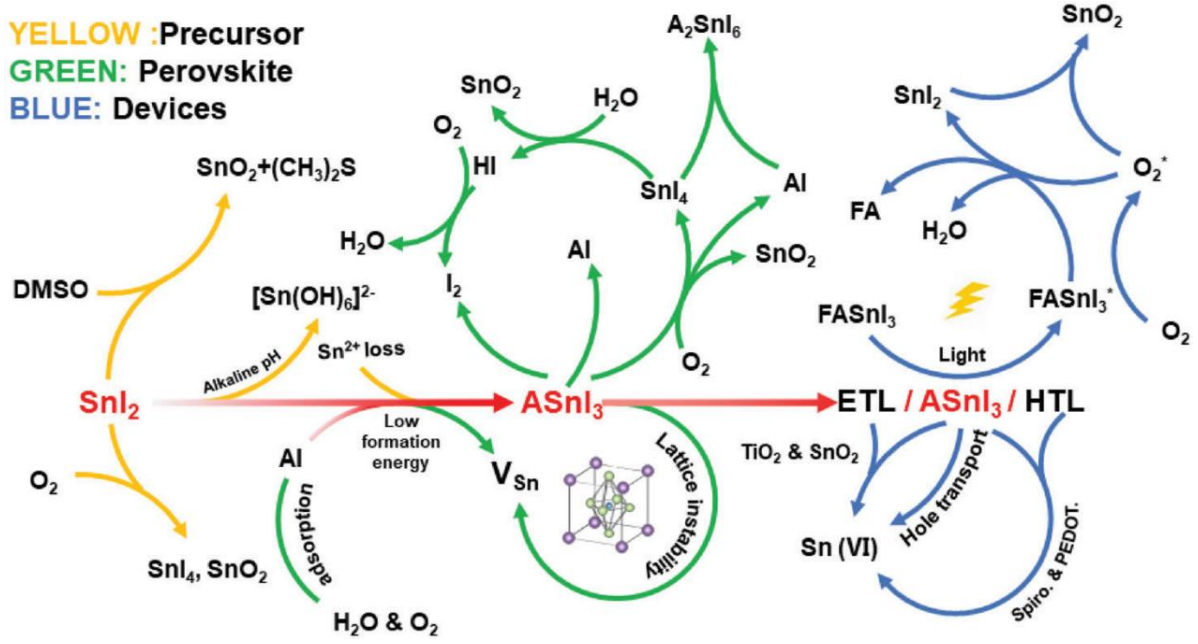

Figure 1. Schematic illustration of oxidation processes during the fabrication and operation of Sn-Pb perovskite solar cells. Copyright 2023, Wiley-VCH.

2.2. Detrimental Impacts on Material and Device Performances

The oxidation of Sn^{2+} to Sn^{4+} creates catastrophic defect structures which adversely affect device performance. This process creates Sn vacancies (VSn) which are materials having a very high non-radiative recombination rate, resulting in a drastic decrease in VOC [12]. This occurs especially in mixed Sn-Pb perovskites of approximately 20% Sn content which have an almost optimal bandgap of 1.33 eV, where large numbers of defects are created, leading to very large decreases in VOC [13]. This process results in the liberation of two holes into the lattice, which gives rise to large, p-type self-doping and high background carrier density [14]. This results in very large decreases in charge carrier lifetimes and diffusion lengths, and thus again resulting in decreases in VOC and in the fill factor (FF) [15].

The oxidation of Sn gives rise to a highly deleterious structural change in the film morphologies. It causes the generation of optically inactive vacancy organized double peroskite phases such as FA_2SnI_6, which result in poor light absorption [16]. In addition, the high reactivity of Sn gives rise to rapid uncontrolled CNDA crystallisation resulting in films of poor quality and low crystallinity resulting in further increases in defect densities [17]. Oxidation products such as molecular iodine (I_2) being extremely corrosive will attack metal electrodes, resulting in the acceleration of device failures [18]. In practice Sn-based films, if exposed resulting during fabrication in becoming rapidly yellow insulating phases. Collectively all the above-described degradation pathways reveal that Sn^{2+} oxidation is the sole ultimate means of causing high defect density, poor quality films, and short lives of devices in Sn-Pb PSC's.

3. Strategies to Suppress Sn^{2+} Oxidation

To address the instability caused by Sn^{2+} oxidation, researchers have developed various strategies focusing on precursor engineering, additives, and interfacial design [19].

3.1. Solvent and Precursor Environment Engineering

Precursor engineering seeks to limit the oxidation of Sn^{2+} prior to or during film formation. Key to this discussion is the form of solvent that is used with the precursors, as solvents such as dimethyl sulfoxide (DMSO) can enhance the oxidation of Sn^{2+} species [20]. In fact, even under glovebox conditions, the effect of unfavourable residual O_2 or the oxidative capabilities of DMSO can readily attack Sn^{2+} species in wet films, and hence the necessity for greater coordination of the solvent species is necessary. Adducts are usually formed from $SnI_2 \cdot$ xDMS0 species, where $x \leq 2$, which have been a source of under-coordinated cluster formations perhaps exposing Sn $5s^2$ lone pair electrons which can lead to oxidation. DFT calculations can show that coodinated $SnI_2 \cdot 3DMSO$ species are by far more stable towards oxidation since an increased oxidation barrier exist [21]. This result would require the use of a one to one (OOS) synthesis of a state-of-the-art comproportionation reaction between SnI_4 and metallic Sn by Zhu et al. which can easily adjust the stochiometry and thus effect exclusively the formation of adduct $SnI_2 \cdot 3DMSO$ species (Figure 2) [22]. The precursor species prepared in this fashion was found to yield more homogeneous precursor solutions with smaller narrow particle size distributions and films with greater crystallinity and reduced trap densities leading to PSC's with a PCE of 22.64% and good operational stability retaining about 90% of its initial efficiency after 530 hours of continued operation.

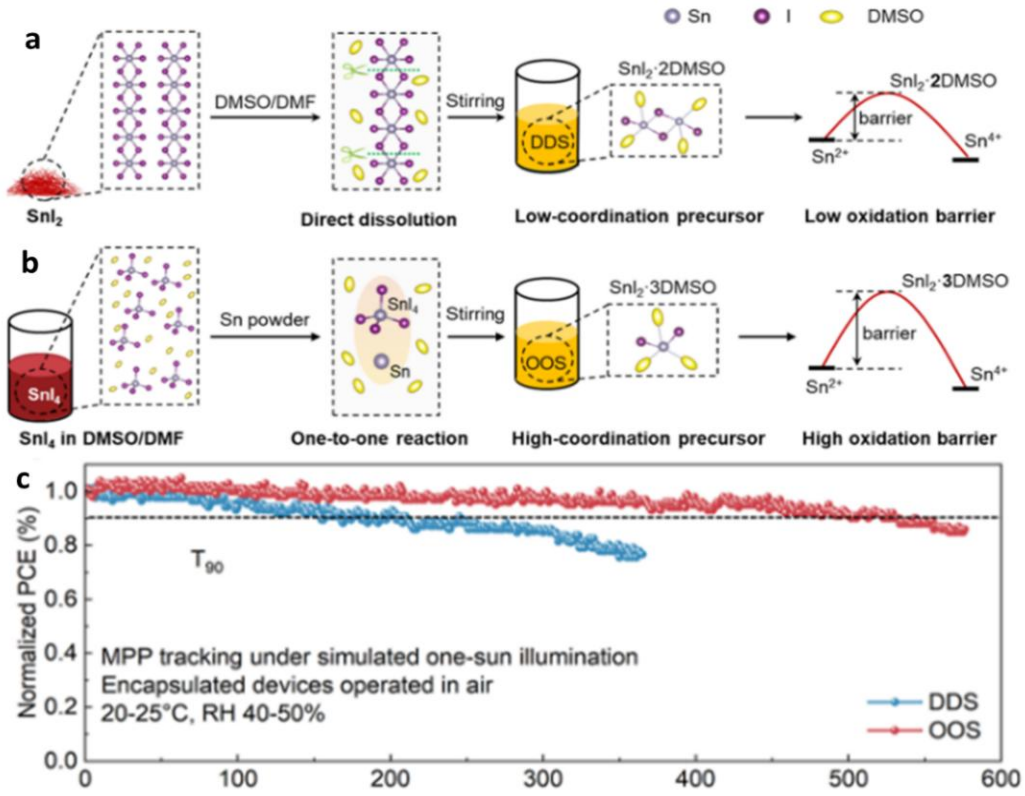

Figure 2. Schematic illustration and performance comparison of the one-to-one synthesis (OOS) strategy for stabilized Sn–Pb perovskite solar cells (PSCs) [22]. Synthetic illustration of (a) DDS and (b) OOS. (c) MPP tracking of the encapsulated devices operated in air (20−25 °C, RH 40−50%), under simulated one-sun illumination. Copyright 2025, American Chemical Society.

Another efficient method is the use of less oxidizing solvents instead of DMSO. Ge et al. [23] have investigated some of the most common solvents systematically and concluded that it was the ability of DMSO to oxidize due to the weak S=O bond of DMSO being weak and unstable which leads to breakage of the bond and the formation of acids. N,N'-Dimethylpropyleneurea (DMPU) has been shown to be an excellent substituent solvent due to its weak oxidizing properties and the ability to take the precursors into co-ordination as well as its high dielectric constant and the fact that the thermal properties are favorable to the forming of homogenous films. By use of a DMF:DMPU solvent mixture they were able to suppress the formation of Sn^{4+} greatly (from 41.78% Sn^{4+} of the films made with DMSO down to 14.76% Sn^{4+}), they were able to increase the carrier lifetime to 139.40 ns and to achieve a device efficiency of 21.04%. Similarly, Zhang et al. [24] showed that another regimented system with DMF+DMPU without DMSO would produce films with less trap density and increased lifetime of carriers and would therefore permit PSC with > 20% PCE. These studies show that rational procedures of solvation can be a great weapon for decreasing Sn^{2+} oxidation. However, it might be wise in future work to extend to multi-functional systems of solvents or synergistic systems of solvent and additive which can maintain the good effects of solubilization but neutralize the oxidative elements.

3.2. Additive Engineering

Apart from the solvent design, the employment of functional additives is critical for the stabilization of Sn^{2+} [25]. The functional additives can be employed as oxygen scavengers, reducing agents, or redox mediators to prevent oxidation events and passivate defects.

One approach involves the usage of functional additives that can competitively react with oxygen. Yan et al. employed benzaldoxime (BZHO), which is known to go through Alder–ene reaction with singlet oxygen and prevents it from oxidizing SnI_2 (Figure 3) [26]. This mechanism also enabled regulation of crystallization of perovskites and modifications in its electronic structure. Furthermore,

the rational employment of radical scavengers is another useful tool. Jung et al employed 4-hydroxy-2,2,6,6-tetramethylpiperidine-1-oxyl (TEMPOL), which has a stable nitroxyl free radical (N–O·), which is very good at quenching reactive oxygen species [27]. The reaction mechanism of a radical scavenger is such that the radical scavenger donates either an electron or hydrogen atom in order to avert the highly reactive radical species without causing chemical damage which ends the chain reaction leading to degradation. At the same time the –OH group forms hydrogen bonds which has the ability to interact with the precursor materials to form ordered crystallization and also imparts greater conductivity and stability to the HTL, since the radical scavenger interacts with PEDOT:PSS. Thus, the functional approach was able to control oxidation of Sn^{2+} and enhanced the PCE from 11.08% to 13.42% besides showing outstanding operational stability under environmental stress such as excessive light, oxygen, and temperature.

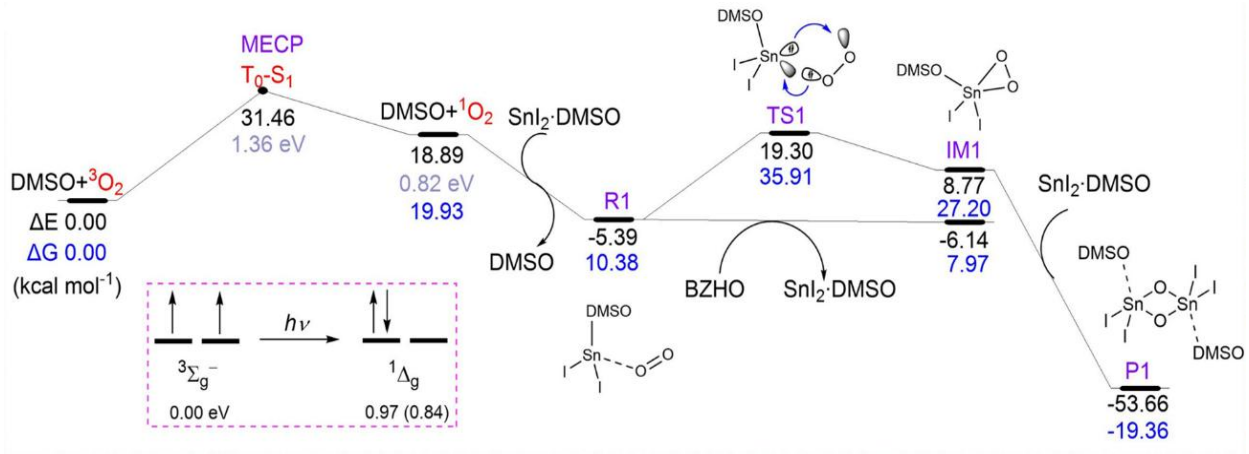

Figure 3. DFT calculations for the pathways of tin (II) oxidation and the inhibition process by BZHO: Excitation of $^3O_2$ to $^1O_2$ and subsequent reaction with SnI_2·DMSO, ΔE and ΔG were presented [26]. Copyright 2024, Wiley-VCH.

Beyond O_2^- driven oxidation, defects arising from iodide and photo-induced I_2 formation also hasten the $Sn^{2+} \rightarrow Sn^{4+}$ transformation. Bai et al. dealt with this issue using phenylhydrazinium (PEH+) additives that strongly coordinate to Sn so that Sn-I bonding is enhanced, but also affecting the negatively charged defects (VSn, VFA, ISn, Ii, etc) positioned for photo-induced I2 formation [28]. In parallel, this required the addition of the reductant 2-mercaptobenzimidazole (MBI) to protect against oxygen attack (presumably via oxidation of the organic material) as well as reduce the density of deep defects. Using this dual method, single junction Sn-Pb PSCs yielded a champion PCE of 23.0% (89.4% after 400 h MPP tracking), while two terminal tandem devices reached 27.9% (certified 27.2%), retaining an efficiency of 90.3% after 300 h.

Utilization of antioxidants and reductants provides a straightforward method for combating Sn^{2+} oxidation. Liu et al. utilized ascorbic acid (AA) in a two-step fabrication process that deters oxidation while creating pores in the film of halide that results in complete conversion and retained photodetector performance [29]. The effect of such additives act to decrease over time, however, as they are consumed in processes. Thus, regenerative systems have been employed. He et al. reported that Sn–Pb perovskites have a sluggish intrinsic self healing redox cycle ($Sn^{4+} + Pb^0 \rightarrow Sn^{2+} + Pb^{2+}$) and accelerated by using a V^{3+}/V^{2+} redox shuttle [30]. A redox shuttle is a catalytically active species which cycles between oxidized and reduced forms in order to effect another redox reaction, in this example resulting in continual conversion of detrimental Sn^{4+} back to active Sn^{2+}. This essentially allowed retention of 90% of initial performance of devices of which the original 21.22% PCE had been retained after 1000 hours of operation. Similarly, Yuan et al. synthesized an organic redox shuttle with tris(2-carboxyethyl)phosphine (TCEP), which functioned by complexing with Sn^{2+} and regulating crystalline growth while in the role of a redox mediator. The oxidation product TCEPO would then be subsequently redoxed back to TCEP such that a dynamic cycle could occur allowing sustained $Sn^{4+} \rightarrow Sn^{2+}$conversion [10].

Research into multifunctional additives that perform Sn^{2+} compensation, Sn^{4+} reduction and provide defect passivation has also been pursued. Wang et al. have described the additive being tin(II) oxalate (SnC_2O_4), where additionally Sn^{2+} is present to compensate for vacancies and the reductive oxalate groups ($C_2O_4^{2-}$) renders Sn^{4+} back to Sn^{2+} and coordinates to Sn^{2+} and Pb^{2+} giving defect passivation [31]. The synergistic effect of this gave rise to films of improved crystallinity, less non-radiative recombination and a PCE of 21.43% with enhanced stability. Further evolved methodology to quest for the Sn(II) species is via a regenerative redox cycle stimulus. Jin et al. have used the additive 4-mercaptobenzonic acid (4-MBA), whereby the thiol group reduces Sn^{4+} thereby producing a disulfide [32]. This disulfide can be cleaved upon UV irradiation leaving regeneration of the active thiol site providing a self-healing cycle whereby the regenerating species can be healed. The –SH group presents passivation to sites taken up by Sn^{2+} group and the –COOH group acts with FA^+ cations leading to dual site defect passivation. The treatment allows for devices to retain 100% PCE over 1100 hours of day/night simulated testing. These examples show a clear evolution of the organic additive chemistry employed from simplistic decertifies to dynamic regenerative systems allowing long-term stability.

3.3. Interface Engineering

The buried perovskite/charge transport interface is one of the most prominent bottlenecks in Sn–Pb-based PSCs [33]. The great liability of Sn^{2+} for oxidation, the easy generation of halide vacancies and the accumulation of carriers at energetically incongruous interfaces combine to induce nonradiative recombination and to accelerate cell degradation. There are two classes of stressors which affect the interface generally considered: chemical, including the acidity and hygroscopicity of hole-transporting materials (HTM's) such as PEDOT:PSS and defect-rich NiO_x, and photophysical, including field-induced accumulation of holes at poorly extracting contacts.

The hole-transport layer (HTL) of PEDOT:PSS is a common choice, however its acididity accelerates the oxidation of Sn^{2+}. One of the principal advancements in this field was made by Chen et al. who used potassium citrate in PEDOT:PSS to improve the acidity of the transport layer, chelate interfacial Sn^{2+} catalysts, and provide K^+ ions which diffuse into the lattice passivating halide defects (Figure 4 a-b) [34]. Using this unique approach provided for efficiencies in single junction devices of 22.7% and tandem devices of 26.1% with T_{80} times up to ~700 h. Li et al. [35] have also rejuvenated PEDOT:PSS contacts by doping acetylcholine chloride (AChCl) to neutralize the acidity of PEDOT:PSS and passivate the defects. The weakly basicity and polar nature of the AChCl headgroup removes the sulfonic acid, passivating interfacial defects which are evident in PEDOT:PSS contacts, while the chloride and carbonyl moieties induce growth of large columnar grains. This substantially decreases the Sn^{4+}/Sn^{2+} ratio in devices simutaneously improving the efficiency of tandem cells to over 27% (certified 26.41%). For those employing NiO_x contacts, the source of intrinsic Ni^{3+} defects which acts as active redox centers, which necessitated the work of Zhou et al. [36] who introduced the use of hydroxyphenethyl ammonium halides (OH-PEAX), which passivated the Ni^{3+} states and guided the low strain growth of favorable FA-rich Pb–Sn perovskites which achieve PSC efficiencies of ∼20 x%. In a complementary manner Zhang et al. introduced the use of 4-aminobenzenesulfonic acid (4-ABSA) interlayer (Figure 4 c-d) [37]. The 4-ABSA molecule developed an orientated dipole moment which increased the energy alignment (decreasing the hole transfer barrier from 0.3 eV to 0.01 eV), passivating Ni^{3+} defects by concentration of electrons, while decreasing the severity of the parasitic reaction, Ni^{3+}/Sn^{2+}. In these competently optimized layers, the all-inorganic Pb–Sn PSCs achieved an efficiency of 17.4%, with over 80% of the performance retained after 240 hr of UV exposure.

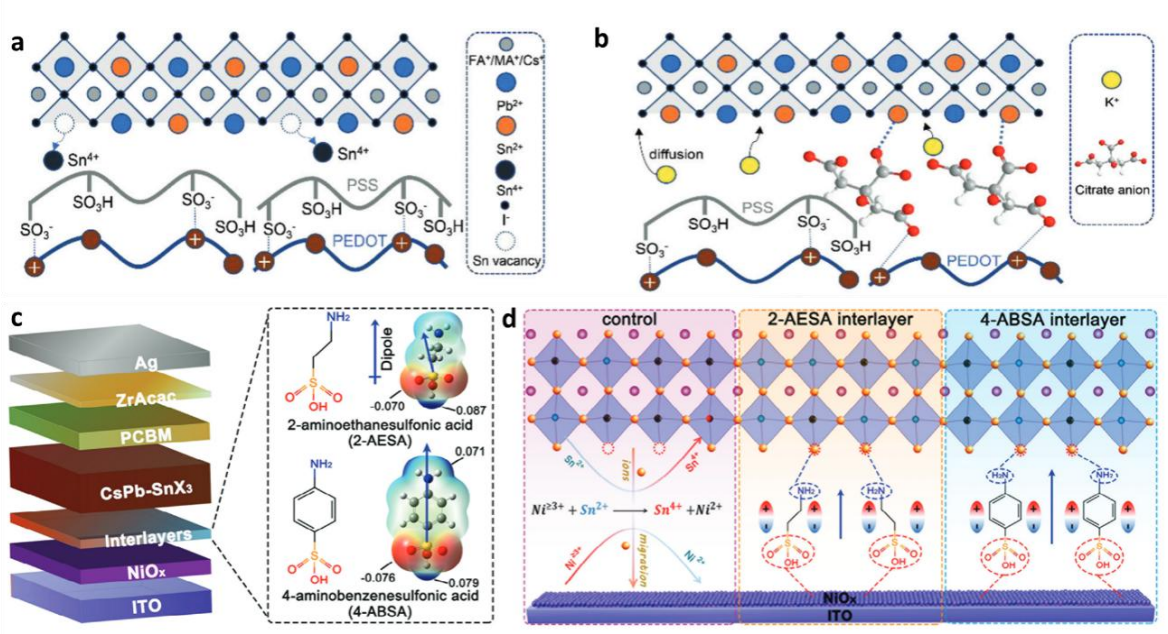

Figure 4. (a-b) Schematic illustration of the lattice structure of Sn-Pb perovskite a) without and b) with potassium citrate passivation [34]. Copyright 2023, Wiley-VCH (c-d) Schematic structure of the device, and chemical structure and electrostatic potential profile of the 2-AESA and 4-ABSA (c); interaction mechanism between 2-AESA, 4-ABSA, and perovskite (d) [37]. Copyright 2025, Wiley-VCH.

Aside from chemical passivation, charge dynamics must also be addressed. Yuan et al. have shown that hole accumulation can result in the oxidation of Sn^{2+} (Fig. 5a-b) [38]. By utilizing a three-dimensional HTL (P3CT/Me-4PACz) which was imbedded into the perovskite, and photoinjected holes aligned to its Fermi energy with the valence band, hole extraction was accelerated, so hole driven oxidation was lessened and devices efficiencies up to 24.03 % with a VOC of > 0.92 V were achieved with ~82 % efficiency retained after 1000 hrs at 55 °C. To complement this Jiang et al. [39] showed an interface energetics reversal approach, using surfactant (i.e. trichloro[3-(pentafluorophenyl)propyl]silane) to change the perovskite surface polarity from n-type to p-type so that it correlated with the valence band HTC. This reduced the non-radiative recombination to record a non-radiative recombination induced qVOC losses (~57 meV). This work also led to a certified device efficiency of 25.9 %. On the electron-transport side, He et al. [40] crosslinked SnO_2 with bisphenol linkers, where phenolic groups served to passivate oxygen vacancies and sulfone groups to coordinate with perovskite precursors. The result was the formation of a uniform SnO_2 network aiding in oriented growth and reduced non-radiative losses of energy. Lai et al. [41] design n-dodecylammonium acetate (DOAAc) to serve for the top-side passivation (Fig. 5c). Here, the acetate anion passivated Sn^{2+}/Pb^{2+} coordination and Coulombically passivated halide vacancies, while the long-chain ammonium passivated surface adhesion through H-bonding. This type of passivator reduced the Sn^{4+} content at the surface from ~24 % to ~9 % and achieved a decrease in mobile ion density of a factor of 4.5 in all cases resulting in improved thermal stability and 23.3 PCE. Wang et al. [42] used a mercapto-functionalized mesoporous silica scaffold at the buried interface, the result was that the resulting templatioanal growth yielded dense film a network which overcame ray Sn oxidation, through strong -SH coordination. Ultimately such devices of narrow band-gap achieved a device efficiency of 23.7 % with a VOC of ~0.89 V equaling work on all perovskite tandems to give enhanced efficiency of 29.5 % PCE certified.

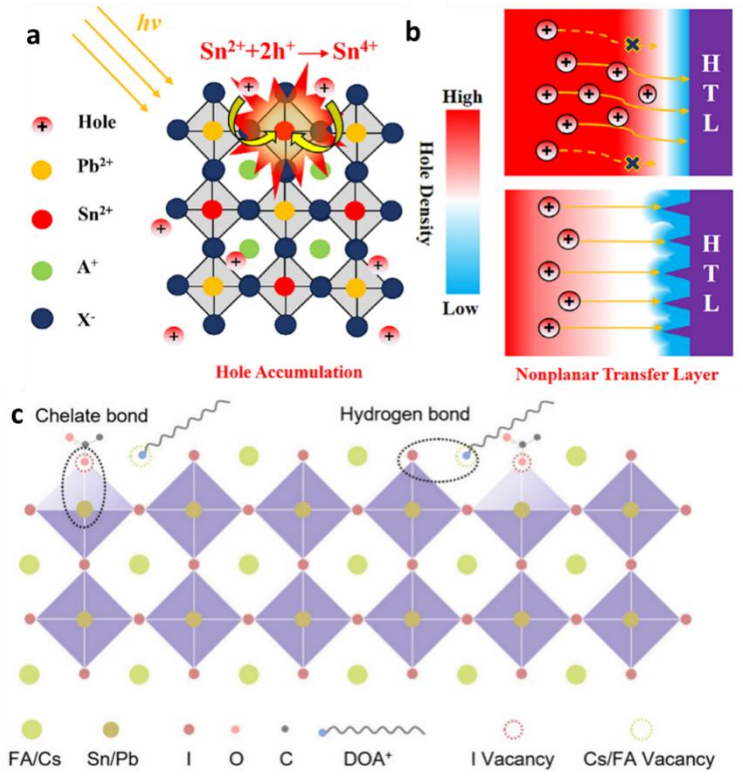

Figure 5. (a-b), Holes-induced Sn^{2+}oxidization (a) and mechanism of the nonplanar hole transport layer (b) [38]. Copyright 2025, Wiley-VCH (c) Schematic illustration of the likely defect passivation mechanisms of DOAAc [41]. Copyright 2025, American Chemical Society.

Availing of emerging concepts to promote interfacial stability through a regenerative chemistry. Jin and colleagues report redox-active interlayers based on reversibly exchanged thiol/disulfide which reduce Sn^{4+} ions to Sn^{2+} during operation [32]. This is thus a transition from static to dynamic interfacial passivation. These advances clearly show that there are chemical buffering and defect passivation strategies and the energetic position of the substituted species that will result in a highly stable buried interface due to minimized carrier residence time and suppressed oxidation processes culminating in highly efficient and durable devices.

4. Conclusion and Outlooks

To inhibit the Sn^{2+} oxidation in Sn–Pb-based PSCs, great progress has been made using precursor, additive and interface manipulation strategies. These strategies afford better film quality, lower defect density, better device performance, and higher stability, but barriers remain to the commercialization of these materials, requiring more insight into the mechanistic understanding of oxidation, ion migration and organic cation decomposition processes using advanced in-operando characterization and multi-scale modeling. The development of green and scalable fabrication practices to replace toxic solvents and improved processing in tandem for rapid production of these coatings must also be addressed. Future advances will be made utilizing innovative and synergistic stabilization strategies involving the incorporation of chemical and interfacial modifications to achieve the necessary extended stability needed to address real-life applications. With constant innovation Sn based-Pb perovskites hold great promise for efficient and stable low-toxic materials for next generation photovoltaics.

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