Tin Halide Perovskite-Based Solar Cells

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The excellent optoelectronic properties of tin halide perovskites (Sn-PVKs) have made them a promising candidate for replacing toxic Pb counterparts. Concurrently, their enormous potential in photon harvesting and thermoelectricity applications has attracted increasing attention. The optoelectronic properties of Sn-PVKs are governed by the flexible nature of SnI_6 octahedra, and they exhibit extremely low thermal conductivity. Due to these two contradictory properties, they are equally explored in solar cell as well as thermoelectric devices.

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1. Introduction

Lead halide perovskite (Pb-PVK) has emerged as a versatile semiconducting material with extensive applications in photovoltaics [1], thermoelectricity [2], light-emitting diodes [3], laser detectors [4], piezoelectricity [5], radiation detectors [6], and optical fibers [7]. Pb-PVKs have a 3D crystal structure with ABX_3 formulae, where A is an organic or inorganic monoatomic element (methyl ammonium (CH_3NH_3/MA), formamidine (CH_3NH_2NH/FA), or Cesium/Cs), B is a bivalent metal ion (Pb^{2+}, Sn^{2+}, or Ge^{2+}), and X is a halide ion (I^-, Br^-, or Cl^-). The B metal ion of the PVK crystal is surrounded by corner-dividing BX_6 octahedra, which are flexible enough to allow such diverse electronic applications. PVK semiconductor thin films can be coated at low temperatures using a solution printing method, which makes the entire fabrication process economical and promising for mass production. The excellent optical and electronic properties of Pb-PVK have made possible a jump in the photoconversion efficiency of solar cells from an initial 3.1% to a record-breaking 25.7% within a short time of research [8][9], surpassing crystalline indium phosphide crystalline solar cells (24.20%), cadmium-free copper indium gallium selenide solar cells (23.35%), and close to crystalline Si solar cells (26.70%) [10], because of the exceptional optical-electrical properties of ambipolar, high defect tolerance, small exciton binding energy, long carrier diffusion length, and high absorption coefficient [11][12][13]. However, despite bearing such promising optoelectronic properties, the notorious toxicity of Pb has raised concerns about commercial implementations. Consequently, various studies have attempted to substitute Pb with other abundant, robust, and biocompatible metals, such as Germanium (Ge), Antimony (Sb), Bismuth (Bi), Titanium (Ti), Copper (Cu), and Tin (Sn) [14][15][16]. Replacing Pb with other metals has obvious implications for the desirable optoelectrical properties relevant to highly efficient solar cells or stable materials. The use of Bi^{3+} or Sb^{3+} results in limitations in the charge transport due to the formed layered vacant structure. Ge^{2+}-based halide perovskite materials exhibit poor chemical stability and poor solubility in polar solvents [17]. Cu^{2+}-based halide perovskite has shown maximum efficiency of 0.99% to date due to the limitations of low absorption coefficient and high effective mass of the holes [18]. Solar cells based on Ti-halide perovskites have so far shown an efficiency of 3.3% [16]. Sn-halide perovskites (Sn-PVK) have similar or superior optical and electronic properties to Pb perovskites, such as a narrow band gap, high mobility, and long-lived hot charge carriers [19]. More importantly, Sn has low toxicity and the degradation product after exposure to air is SnO_2. To date, Sn-PVK-based solar cells have attained around 14.81% efficiency, which is the best among the non-Pb-PVK-based solar cells [20]. Therefore, Sn halide perovskites are a promising material for developing efficient Pb-free halide perovskite solar cells.

Despite bearing such superior electronic properties, the achieved 14.81% efficiency of Sn-PVK solar cells is far inferior to the 25.7% of Pb-PVK solar cells [20]. Such a limitation arises mainly due to the different kinetics of fast crystallization and poor stability. The hindrance to the stability of Sn-PVK arises due to the facile oxidation of Sn^{2+} into Sn^{4+}. This formed Sn^{4+} is responsible for unwanted high doping, leading to a high background carrier density and greatly hindering the performance of the Sn-PVK solar cells. However, Sn-PVK films with high background carrier density and high conductivity can be used as thermoelectric films. Sn-PVK films have extremely low thermal conductivity and a reasonable Seebeck coefficient [21]. Therefore, Sn-PVK films can be used as energy harvesting films, either as a photon harvesting layer in solar cells or as a waste-heat-to-energy harvesting layer in thermoelectric devices. The physical and electronic properties of Sn-PVK are discussed here from the viewpoint of benign, forward-looking alternatives to Pb-PVK.

2. Normal Structure

In solar cells with normal structure, the perovskite ink is deposited on electron-selective metal oxide layers consisting of TiO_2 or SnO_2. It is worth noting that these metal-oxide layers are porous with a common phenomenon of oxygen vacancies. These vacancies may serve as a trap center to adversely affect the photoelectric performance. Ogomi et al. showed that the $MASnI_3$ semiconducting layer has no photoelectric properties in solar cells with an n-i-p structure and TiO_2 electrodes [22]. The first report on Sn-PVK solar cells was published by Snaith et al. in 2014. The photo-harvesting layer of $MASnI_3$ was used to fabricate the cells in an n-i-p structure on TiO_2 electrodes. A maximum efficiency of 6.4% at 0.88 V Voc

was achieved. However, looking closely at the distribution bar, the average efficiency was less than 2% and most of the solar cells had an efficiency close to 0% [23]. Zhao et al. reported solar cells based on $MASnI_3$ and $FASnI_3$ in a normal n-i-p structure. The cells were fabricated as FTO glass/cp-TiO_2/mp-TiO_2/Sn-PVK/spiro-MeOTAD/Au. However, poor diode rectification performance was obtained with efficiencies of 0.16% and 0.65%, respectively [24].

The reason for this unusual behavior of poor diode characteristics or short-circuit in Sn-PVK solar cells with normal structure was investigated by Hamada et al. [25]. Using, a thermally stimulated current, they showed that when the inorganic TiO_2 layer was passivated by Sn-PVK, the defect states were largely enhanced. In contrast, TiO_2 passivated with Pb-PVK showed suppressed defect states. This poor photoelectric performance of Sn-PVK in n-i-p-based solar cells due to the enhanced defect density with metal oxide films led to this structure attracting less interest from researchers.

Jen et al. fabricated Sn-PVK solar cells with normal and inverted structures [26]. They showed that the addition of trimethyl iodide in $FASnI_3$ succeeded in achieving a high efficiency of 7.09% in an inverted structure and only

4.43% efficiency was obtained in a normal structure. However, the point is that a thin C60-coated electron-selective layer SnO_2 layer was used in the normal-structure solar cells. With this modified electron transport layer, they succeeded in fabricating functional Sn–PVK solar cells. Xu et al. developed {en} $FASnI_3$-based solar cells in normal n-i-p structure [27]. They used an inter-layer of C60-pyrrolidine over inorganic SnO_2. The double-layer SnO_2-C60 pyrrolidine tris-acid was used as an electron transport layer. The performance of the solar cell was improved to 7.40% with a Voc of 0.72 V and a Jsc of 16.45 $cm^2/V{\cdot}s$. However, the solar cells fabricated on bare SnO_2 ETL showed poor performance with a Voc of ~0.2V and Jsc of ~2 mA/cm^2. Such a C60-pyrrolidine over inorganic SnO_2 was able to suppress the charge recombination. Hamada et al. showed that the use of C60-COOH as an interlayer between the photoactive Sn-Pb PVK layer and the TiO_2 electrode is essential for functional n–i–p-based Sn-Pb solar cells [25]. This C60-COOH interlayer strategy resulted in an enhanced photoelectric performance of 7.91% compared to 5.14% of bare SnO_2 electrodes. They concluded that without the C60-COOH interlayer, Sn-Pb PVK was itself oxidized to form appreciable amounts of Sn^{4+}. The reason for this Sn^{4+} formation with metal oxide and Sn–PVK contact was recently investigated by Baranwal et al. [28][29].

Baranwal et al. reported that Sn-PVK coated on an inorganic Y_2O_3 layer exhibited enhanced electrical conductivity due to the formation of Sn^{4+} by the oxidation of Sn^{2+} [29]. This was also confirmed by the lowering of the Seebeck coefficient values. This behavior is similar to that of the normal Sn-PVK solar cells with an n-i-p structure, where poor rectification or short-circuiting was observed due to Sn^{4+} formation without an interlayer at the metal oxide/PVK interface. This unusual Sn-PVK oxidation on the inorganic metal oxide layer was explained by an ultraviolet photo spectroscopy study that showed that the inorganic metal-oxide layers have sufficient defect energy states in the bandgap, above the VBM, and below the work function energy level. These electron acceptor defect states easily attract and trap the electrons from shallow energetic Sn-PVKs. Sn-PVKs tend to lose electrons easily, and, therefore, Sn-PVK becomes positively charged and Sn^{4+} is formed immediately even if the fabrication process is done in an inert or N_2 atmosphere [28]. It was concluded that the reason for the poor efficiency and limited reports of n-i-p-based Sn-PVK solar cells with normal structure was the oxidation of Sn-PVK due to the defect states of the metal oxide layers.

3. Inverted Structure

To date, Sn-PVK solar cells with high efficiency have been described in inverted structure with PEDOT:PSS as the HTL. SnF_2 is the most common component of the precursor to compensate for the Sn defects. Zillner et al. found an interesting result [30]; they investigated the effect of SnF_2 addition on $FASnI_3$ perovskite solar cells. It was found that SnF_2 accumulates at the top of PEDOT:PSS. Sn of SnF_2 and S of PEDOT:PSS preferentially form a 1.2 nm thin interlayer of SnS at the interface of PEDOT:PSS/$FASnI_3$. However, this intermittent layer of SnS was not formed when the mixed Pb-Sn perovskite $FA_{0.75}MA_{0.25}Sn_{0.50}Pb_{0.50}I_3$ was used.

3.1. Device Engineering

Wang et al. investigated a new device structure using inorganic SnO_x as an ambipolar layer in Sn-PVK solar cells [31]. The SnO_x was used in situ as the bottom surface and on the top surface of Sn-PVK. Thus, SnO_x served two different purposes as HTL and PVK protective layers. The bottom SnO_x can efficiently transfer the charge carriers, and the upper SnO_x served as a protective layer reducing Sn^{4+} to Sn^{2+} and supporting facile electron transfer but blocking hole transfer. The SnO_x layer was deposited in a two-step simple process. First, Sn powder was vapor-deposited in a thermal evaporation method. Sn powder has a moderately low melting temperature of 231.9 °C. Second, this thermally evaporated tin was exposed to plasma under ambient conditions to convert to SnO_x. The charge-conducting properties of SnO_x were adjusted by controlling the plasma exposure on the evaporated Sn film. This plasma exposure time adjustment varied the ratio of Sn and O of SnO_x to work as an ambipolar layer. The bottom and top layer SnO_x thicknesses were maintained at 10 and 2 nm, with plasma exposure times of 5 min and 2 s, respectively. This simple in-situ SnO_x strategy resulted in an efficiency of 14.09% for Sn-PVK solar cells.

3.2. A 2D/3D Photoactive Layer

Yu et. al. introduced 4-fluoro-phenethylammonium bromide (FPEABr) as an A-site substitution in $FASnI_3$ [20]. The FPEABr substitution resulted in a 2D/3D perovskite layer where, FPEABr was sitting simultaneously at the grain boundary, surface, and throughout the bulk. Thus, the bulk and interfacial defect density were suppressed. This leads to a lifetime twice that of the original $FASnI_3$ film. The crystal growth was highly oriented and grain boundaries were fused with optimized 10% FPEABr. The resulting energetics showed a better energy matching, as 2D/3D perovskite had shallow VBM and CBM, approaching better alignment with PEDOT:PSS and ICBA. Due to the increased crystallinity, suppressed Sn^{4+} defects, and better energy alignment, a record efficiency of 14.81% efficiency was achieved, which is the best-reported efficiency in Sn-PVK to date. Diau et al. used a sequential deposition method to deposit various bulky ammonium cations on top of the 3D layer to form a 3D/quasi-2D layer [32]. Firstly, the Sn-PVK layer was fabricated. Next, various 2D layers of butyl ammonium, hexylammonium, allylammonium, cyclohexylammonium, phenylethylammonium, anilinium, 2-fluoro phenylethylammonium, and 2-fluroannilium were coated over the PVK layer. Isopropanol (IPA) solvent was used to dissolve these 2D molecules. However, IPA damaged the Sn-PVK surface. Therefore, hexafluoro-2- propanol (HFP) was used as the solvent to avoid the dissolution of the Sn-PVK. Fluorine of HFP interacts with these 2D molecules to slow down the reactivity with the Sn-PVK surface, thus forming a 2D or quasi-2D layer. Therefore, an ultrathin layer of 2D was capped on the Sn-PVK surface covering the grain surface and grain boundary. The anilinium-based Sn-PVK bilayer photoactive device exhibited an enhanced efficiency of 10.6%. The cells were tested under a broad range of possible conditions of dark storage under N_2 and ambient, light soaking test at the maximum power point (N_2), and ambient thermal stress test under dark and illumination. Excellent stability was obtained under ambient thermal stress tests (50 °C to 55 °C (10 cycles)) and cooling at 20 °C illumination tests for anilinium-based devices. The solar cells were encapsulated for these severe tests. This was the first report to discuss thermal stress tests of Sn-PVK, which were successful due to the self-healing phenomenon of the capping layer of anilinium. It was also reported that the temperature of the solar cells following 1-h illumination exposure led to the oxidation of the silver electrodes.

3.3. Surface Passivation of Photoactive Layer

Akmal et al. used the Lewis base ethane-1,-2diamine (edamine) as a post-treatment agent to passivate the surface defects of Sn-PVK [33]. Sn-PVK surfaces have dangling and under-coordinated bonds, and high electronegative amine of edamine was coordinated with these unsaturated bonds to control the Sn^{2+} oxidation and Sn vacancies. An optimum 0.05mM edamine passivation converts unreacted SnI_2 into a perovskite phase and improved the surface morphology. Edamine donates electrons to reconstruct the Sn-PVK surface properties from p- to n-type. The better energetics and easy charge transport assisted in an improved efficiency of 9.37%. This post-treatment strategy of Sn–PVK using edamine was successfully replicated by various authors [34][35][36][37]. The use of edamine as post-treatment was explored in Sn-Pb perovskite solar cells also. It was revealed that the p-type properties of Sn-Pb perovskite were transformed into n-type and the built-in potential was enhanced from 0.56 V to 0.76 V. This contributed to an enhancement in Voc of 100 mV [38]. Recently, a similar post-treatment strategy was followed using 6-maleimidohexanehydrazide trifluoroacetate as a surface cationic and anionic passivator [39]. The reductive nature of the hydrazine and carboxyl group neutralize the charged defects states and changed the properties of the Sn-PVK surface from p to n-type. The n-type surface leads to a better band-bending between the surface and bulk of Sn-PVK, assisting better electron transfer at the C60/$FASnI_3$ interface and prohibiting hole recombination. This strategy resulted in a 13.64% efficiency in $FASnI_3$-based solar cells. Lia et al. used a low-dimensional phenylammonium bromide (PEABr) coating (dissolved in IPA) on the $FASnI_3$ surface. Such thin PEABr-coated Sn-PVK films achieved improved crystallization, better energetic alignment, reduced defect density, and suppressed Sn^{2+} oxidation. A 7.86% efficiency was reported, which retained 80% of its original values during a 350 h light test under ambient conditions [40]. Liu et al. performed pretreatment of $FASnI_3$ with n-propyl ammonium iodide (PAI) during the spin coating step after dripping the antisolvent and before annealing. The Sn-PVK crystallization process is very fast. To slow it down, DMSO is used as a co-solvent as it makes the SnI_2-3DMSO complex. The solvent for PAI was a mixture of DMSO and chloroform. Therefore, the intermediate phase $FASnI_3$.3DMSO was reconstituted with a better morphology. Intermediate film morphology had bright grain boundaries, which were grown in a template such as in the (100) plane. The PA cation did not form a 2D layer over the surface but was located at the grain boundary instead of the lattice. Overall, a highly crystallized $FASnI_3$ film with a reduced trap density photoactive layer resulted in an efficiency of over 11% [41]. Zhang et al. performed the surface passivation of $FASnI_3$ with trimethylsilyl halide (TMS-X). The halide vacancies on the perovskite were passivated by the halide of TMS-X, as X was changed to Br, Cl, and I. TMS-X has ionic characteristics with TMS^+ and X^- as ionic moieties, and they passivated anion and cation trap states. The VBM of TMS-X passivated Sn-PVK showed deeper VBM in the order of (untreated < TMS-Cl < TMS-Br < TMS-I). The fermi level was also suppressed and became shallower, such that (untreated > TMS-Cl > TMS-Br > TMS-I). It was postulated that the X^- ions passivated the I^- vacancies and TMS^+ remains on the surface, such that TMS-X formed a hydrophobic protective surface over Sn-PVK. This strategy resulted in enhanced photoelectric performances of 12.22% (TMS-Br passivation), 11.68% (TMS-Cl passivation), and 11.58% (TMS-I passivation) [42].

Surface passivation of perovskites using a liquid solvent is a very common phenomenon. However, it has the interesting disadvantage that the film thickness decreases because the passivating solvents can dissolve the perovskite to some extent. Zhang et al. reported that the liquid passivation strategy reduces the thickness of the Sn-PVK active layer, thus affecting the Jsc values [35]. However, the use of vapor-assisted passivation can avoid such a problem. With such a strategy, better efficiency was achieved with edamine vapor passivation compared with liquid passivation. An improved diode factor of 1.43 with vapor edamine treatment was obtained compared to 1.57 for a liquid edamine-treated film. In the vapor passivation, a high Jsc of 24.05 mA/cm^2 was revealed; conversely, the liquid-treated film showed a suppressed Jsc of 22.38 mA/cm^2. This method showed an improved efficiency of 11.29%. A sequential strategy to passivate the defects and improve the crystallinity was explored recently using acetylacetone (ACAC) and edamine as post-treatment agents [43]. First, Sn-PVK films were prepared. In the post-treatment, these films were passivated with ACAC molecules. ACAC treatment fuses the grain boundaries and enhances the grain size under the Ostwald ripening effect. The films were placed on a hot plate to remove the solvents. In the next step, edamine vapor passivation was performed. Such edamine passivation suppressed the Sn^{4+} defects by coordinating unsaturated bonds. This dual passivation strategy resulted in a promising 13% efficiency with 0.79 V Voc. Chowdhury et al. reported large area (1.02 cm^2) Sn-PVK solar cells with an efficiency of 6.33% [44]. After the deposition of $FASnI_3$, the perovskite surface was treated with methylammonium chloride (MACl) vapor. To generate the MACl vapor, MACl powder was placed on a hot plate. This strategy resulted in homogenous and crystal defect-free $FASnI_3$ film.

3.4. A-Site Substitution

Bulky organic cations butylammonium iodide (BAI) and ethylene diammonium iodide ($EDAI_2$) were first used as an additive by Diau et al [45]. Two-dimensional BAI improved the connectivity of grains and altered the orientation of the growth of the crystal in a preferential direction. However, fast crystallization was not suppressed, and a large number of pin holes were visible on the surface. To sustain the defects and vacancies of Sn-PVK and to slow down the crystallization rate, $EDAI_2$ as an additive was proposed. In addition to this, $EDAI_2$ served the purposes of retaining smooth surfaces by controlling nucleation and growth and suppressing the surface defects, and improving stability, as EDA^{2+} may occupy the vacant position of two FA^+ ions. However, various efficient Sn-PVK solar cells are reported with the substitution of FA^+ by EDA^{2+} [34][43][46]. Chen et al. used $EDABr_2$ as a substitute at the A-site in the fabrication of $FASnI_2Br$-based wide-bandgap solar cells [47]. This results in smooth and low-defect perovskite film with enhanced efficiency of 4.48%. Diau et al. replaced FA with azetidinium (AZ) [48]. Both cations have a similar radius of 253 pm and 250 pm, however, AZ has a high dipole moment. The crystallization speed of Sn-PVK was quick because of the strong interaction of AZ and SnI_6 octahedra. Therefore, a new antisolvent trifluorotoluene (TFT) with a high dipole moment (D) of 2.86 was used in place of traditional chlorobenzene antisolvent (1.69 D). TFT produced uniform nucleation seeds and provided sufficient nucleation growth. This strategy resulted in a uniform and crystalline Sn-PVK surface. A better photovoltaic performance of 9.6% was obtained due to the increased band gap energy, band alignment, and improved crystal growth. The crystallized Sn-PVK had a much smoother surface and lower defect density resulting in better charge transport. It was shown for the first time that the shelf life of Sn-PVKs could maintain 90% of their original efficiency at 55% humidity for 15 days. Nishimura et al. used an ethyl ammonium cation to replace the A-site FA [34]. The resulting film exhibited an aligned energy cascade as the VBM and CBM become deeper, and achieved an efficiency of over 13% due to suppressed defect states. They also show that the Sn-PVK has halogen defects on the surface. Zhang et al. used diethyl ammonium iodide (DEAI) to replace the A cation [36]. The DEA-substituted Sn-PVK films exhibited larger grain sizes and deeper energetics. Higher efficiency was observed due to the suppression of Sn^{4+} and better energy alignment.

Chen et al. optimized the A-site of $FASnI_2Br$ by replacing FA with guanidinium bromide [49]. Such an A-site substitution promotes a pin-hole-free surface, better energy alignment, and charge extraction at the interfaces. An improved efficiency of 7% at 0.64 Voc was reported for the wide band gap $FASnI_2Br$ PVK solar cell.

3.5. Vacuum-Assisted Strategy

Zhao et al. fabricated Sn-PVK using an antisolvent-assisted strategy [50]. After spin coating, the films were not annealed. In place of annealing, the authors stored the films under a vacuum for different time intervals. It was determined that the boiling point of Dimethylformamide (DMF), Dimethylsulfoxide (DMSO), and chlorobenzene were much higher than the film baking temperature of 70 °C. Therefore, these solvents must remain as a residue in the film. The authors show that, after prolonged storage in a vacuum, these solvent residues were evaporated completely. By using the strategy of storing the films for 12 h under a vacuum, before depositing C60 and BCP, a solar cell with a performance of over 10% was fabricated. Such a strategy was able to suppress the defect states of Sn-PVK. However, such a long-time vacuum treatment strategy to evaporate solvents is not practical despite the promise of suppressed bulk and surface defect densities. Yin et al. fabricated $CsSnI_3$-based solar cells using a thermal vapor passivation strategy [51]. First, SnI_2 was thermally deposited on a PEDOT:PSS film. Then, various passivation layers of thiourea, thioacetamide, thiosemicarbazide (TSC), and guanidine hydroiodide were deposited by a thermal evaporation method. Finally, CsI was thermally deposited and annealing was carried out. Among the passivation layers, TSC was anchored as a Lewis acid-base reaction between the S=C-N functional group and Sn ion. This strong coordination anchoring leads to an electron cloud on defects and improved the defect formation energy. During the annealing process, the perovskite precursor overcame the energy barrier, and CsI and SnI_2 reacted to form the $CsSnI_3$ film. During the annealing, TSC was linked to the $CsSnI_3$ surface and allowed the lower nucleation site to achieve a bigger grain surface. The passivated $CsSnI_3$ film with TSC interlayer exhibited a higher efficiency of 8.20%. Such high efficiency with low tolerant $CsSnI_3$ was possible due to the suppressed deep defect densities of undercoordinated Sn^{2+} and vacant Sn inhibiting the nonradiative recombination.

3.6. Solution Engineering

Sn-PVK precursor solutions are commonly dissolved in DMF:DMSO solvents. In 2019, Baranwal at al first reported that stirring the precursor solution of Sn-PVK in DMSO as cosolvent oxidizes Sn^{2+} into Sn^{4+} [52]. This was confirmed by electrical conductivity and Seebeck coefficient measurements. Later in 2020, Abate et al. found a similar phenomenon using NMR spectroscopy that Sn^{2+} is oxidized under the DMSO effect [53]. Thereafter, different solvents were explored as substituents for DMSO. Using DEF:DMPU as a solvent to dissolve Sn-PVK, an efficiency of 6.2% was reported (Voc = 0.533 V, Jsc = 21.9 mA cm^{-2}, FF = 0.53) without using SnF_2, Sn(0), and reducing agent. The solar cells were fabricated in the inverted structure as PEDOT:PSS/Sn-PVK/C60/BCP/Ag [54]. Subsequently, various efforts were made to limit the use of DMSO as a co-solvent. However, DMSO can slow down the rapid crystallization of Sn-PVK by forming the SnI_2-3DMSO complex. Thus, DMSO remains an essential co-solvent. Baranwal et. al. investigated the aging effect of the Sn-PVK precursor solution on the photoelectric performance of Sn-PVK solar cells [46]. It was found that DMSO as a co-solvent can increase the oxidation rate of the Sn-PVK solution. Conversely, DMF as a stirring solvent can suppress such Sn-PVK oxidation inside the solution. In addition, GeI_2 as an additive was shown to control the effect of oxidative solvents. A judicious strategy was followed to fabricate the solar cells. The Sn-PVK precursor solution was continuously stirred in DMF only and a controlled amount of DMSO was added 5 min before the deposition of the Sn-PVK film. In this process, PVK solution stoichiometry was maintained. An efficiency of 10.26% was obtained when DMF was used as the stirring solvent and an efficiency of 7.12% was obtained when DMSO was used as the stirring solvent. DMF was used as a solvent to dissolve Sn-PVK, and a controlled amount of DMSO was added 5 min before the film fabrication. Recently, Abate et al. used 4-tert-butyl pyridine as a co-solvent with DMF and 1,3-dimethyl-2-imidazolidinone [55]. DMSO and SnF_2 were not used in the fabrication process. Inverted structure solar cells with a photoactive layer of $FA_{0.78}MA_{0.2}EDAI_{0.02}SnI_3$ achieved an efficiency of 7.2%. Wu et al. showed that the color of SnI_2 powder dissolved in DMF changed rapidly in air compared to SnI_2 dissolved in DMSO [56]. This is because SnI_2 forms a complex with DMSO, which reduces the dissociation of SnI_2. However, DMSO itself is oxidative, so various co-solvents were used as a co-additive to DMF. The tin perovskite precursor was dissolved in DMF and N-methylformanilide (NMF) or DMF and 4-acetaminophen (AP). Inverted Sn-PVK solar cells with DMF-AP showed an efficiency of 10.03% and DMF-NMF-based solar cells showed an efficiency of 8.68%. Diau et al. investigated the two-step fabrication of Sn-PVK [57]. In the first step, SnI_2 and SnF_2 were coated with DMSO as the solvent. In the second step, FAI and $EDAI_2$ were coated with a solvent system hexafluoro-2-propanol (HFP), isopropanol (IPA), and chlorobenzene (CB). The strong interaction of SnI_2 with IPA inhibited perovskite formation, so, HFP was used to suppress the reactivity of IPA by establishing hydrogen bonding with IPA. The CB used in the second step served as an antisolvent.

3.7. Additive Technology

GeI_2 was used as an additive in the fabrication of Sn-PVK solar cells to reduce defects and control the background charge density. Small radius Ge can fill the voids in the crystal as well as the surface holes to minimize the defects and vacancies. The sacrificial oxidation of Ge forms a GeO_x layer at the top and bottom surface of Sn-PVK, which serves as a protective layer against moisture and humidity ingress. Ge can also sit at the grain boundary [47][58][59]. $EDAI_2$ and BAI were used as additives in Sn-PVK solar cells [45]. BAI changed the surface morphology to fuse the crystal boundary which significantly improved the crystallinity and controlled the crystal growth direction. $EDAI_2$ curbed the fast crystallization and passivated the defects of Sn-PVK film by controlling the Sn^{2+} vacancy defects and Sn defects. These two additives act together to improve the photoelectric performance of Sn-PVK solar cells. Their role is discussed in detail in the additive engineering section. Phenylhydrazine chloride (PHCl) was used as an additive in $FASnI_3$. The phenyl group is hydrophobic, and has 2D nature and hydrazine is a reducing agent. Bulky 2D cations are insulating and impede the photoelectric properties, therefore 3D/2D or 3D/quasi2D photoactive layers are fabricated. PHCl as an additive suppressed Sn^{4+} defects and repair Sn vacancies making the bigger crystal size. The bulky phenyl moiety was incorporated into the crystal lattice, blocking oxygen and H_2O. The addition of chloride salt makes the VBM and CBM deeper, which allows a better energetic alignment with PEDOT:PSS and C60. An efficiency of 11.4% was obtained showing stable behavior when stored in the glove box for 110 days. However, the solar cells showed decreasing efficiency when exposed to air. When these cells were stored in a glove box, the initial efficiency was recovered under the hydrophobic and reductive effect of PHCl. An enhanced 24.2 ns lifetime was reported compared to 7.6 ns of the pristine film [60]. It is worth noting that, hydrazinium chloride, hydrazinium iodide, and hydrazine vapor have been already utilized as additives and reducing agents to boost the photoelectric performance in Sn-PVK solar cells [61][62][63]. Zou et al. explored the combined beneficial effect of benzylamine and the F atom by using 4-fluorobenzylammonium iodide (FBZAI) as an additive to $FASnI_3$ [64]. The benzylamine and the F atom served a dual purpose; benzylamine is hydrophobic, has π

conjugated system, and shows conducting behavior; conversely, fluorine has a high electronegativity to tune the electron density. Enhanced spectra in the region of 450–650 nm showed reduced defect density in the bulk, and improved spectra in 750–850 nm showed better contact at the PVK/C60 interface, suggesting fewer traps at the interfaces. Thus, FBZAI also improved the quality of the C60-Sn-PVK interface by reducing the interface defect states, resulting in easy charge transfer. Due to the high electronegativity of F, $FBZA^+$ attracts electrons from adjacent atoms to become more negatively charged and NH_3^+ becomes more positively charged. Such a dipole synergistically passivated the defect states by forming ionic or hydrogen bonds. An increased efficiency of 13.85% was reported. A similar mechanism of defect passivation was earlier employed using additives benzyl amine, fluoro-phenethylammonium iodide, and pentafluorophen-oxyethylammonium iodide [65][66][67][68]. Ivan et al. reported $FASnI_3$-based solar cells with dipropyl ammonium iodide and sodium borohydride as dual additives [69]. Di-propyl ammonium is a bulky 2D ion, whereas sodium borohydride is a reducing agent. The synergetic effect of these two additives achieved a smooth and defect-tolerant film. After three days of light exposure, the solar cells achieved 10.80% efficiency. The fabricated solar cells showed maximum power point stability for 1300 h measured in an N_2 atmosphere with a UV filter. The reduced Sn^{+2}/Sn^{+4} and I^-/I_2 ratios observed in the XPS measurements compared to the pure $FASnI_3$ were attributed to the ability of the additives to passivate and limit the loss of iodine species.

Zhao et al. used ethylenediammonium halide salts $EDAI_2$ and $EDABr_2$ as additives in the fabrication of Sn-PVK solar cells [70]. Although both salts were able to suppress oxidation and passivate trap states, $EDABr_2$ was more effective in passivating the grain defects, reducing the Sn vacancies, and controlling the background charge density. This was possible because Br^- with its small radius has a larger electrostatic potential and shorter bond length between Sn and Br than between Sn and I. This strategy leads to the suppression of deep SnI

defect states. A high efficiency of 14.23% was reported. Chen et al. used 3,3′-(((2,2-diphenylethene-1,1-diyl)bis(4,1-phenylene))bis(oxy))bis(N,N-dimethylpropan-1-amine)tetraphenylethene] (PTN-Br) as an additive to $FASnI_3$ [71]. PTN-Br is a π-conjugated semiconducting polymer with deeper VBM. Grain boundaries are a vulnerable site of degradation against UV exposure and moisture/oxygen, and a weaker site for charge conduction. PTN-Br filled the grain boundaries, served as a hole transport center, and improved hole collection, improving the Voc and FF. Dimethylamine of PTN-Br made a Lewis adduct with undercoordinated Sn. The reduced defect states and improved hole collection led to an improvement in efficiency from the original 5.12% (Voc = 0.435) to 7.94% (Voc = 0.544). This demonstrated that the cells maintained 66% of initial efficiency following 5 h continuous UV exposure due to the protective and coordinating ability of PTN-Br. 2,2,2-trifluoroethylamine hydrochloride (TFEACl) was used as an additive. Bulky $TFEA^+$ were present at the grain boundaries, inhibiting the oxygen ingress and Cl^- was inserted into the lattice, extending the crystallinity. The defect states were suppressed and improved efficiency was reported [72]. Mi et al. synthesize a 0.8M solution of SnI_2 by dissolving Sn granules and I_2 in a solvent mixture of DMF:DMSO (4:1 *v*/*v*) [64]. This simple method yielded a clear yellow solution of SnI_2 with overnight dissolution.

After this, FAI, PEAI, and trimethyl thiourea were dissolved in SnI_2 solution to yield Sn-PVK precursor for solar cell fabrication. Thiourea worked as a soft Lewis base additive and the functional group C=S had a greater affinity towards Sn^{2+}, the N-H group of thiourea formed a hydrogen-bond donor to I^- defects, resulting in a uniform and compact layer with fewer Sn^{4+} defects. Thiourea and the PEDOT:PSS formed hydrogen bonding. This strategy had multiple advantages of being flat, compact with great adhering, and overlapped grain boundary with greater crystal size $FASnI_3$. The cells were able to achieve a record efficiency of 14.3% with 0.92V Voc. Moreover, such a facile SnI_2 preparation process shows that extremely pure SnI_2 granules are not essential to attain high photoelectric performance. [37]

3.8. Three-Layer Mesoscopic Solar Cell

Three-layer mesoscopic PVK solar cells are known for their unparalleled stability among PVK solar cells [73]. Diau et al. fabricated Sn-PVK solar cells based on FTO glass/TiO_2/Al_2O_3/carbon-infiltrated Sn-PVK [74]. In the photoactive $FASnI_3$ layer used, I varied with tetrafluoroborate (BF_4) to form $FASnI_{3-x}(BF_4)_x$. The BF_4^- ion had stronger coupling with Sn^{2+} ion, thus, the optimized $FASnI(BF_4)_2$ precursor suppressed Sn^{4+} formation and achieved a maximum 1.3% efficiency with a Jsc of 20.2mA/cm^2, Voc of 0.194V and FF of 0.34. It should be noted that such poor efficiency may have arisen due to the electron acceptor defect states of TiO_2 and Al_2O_3 [28]. The cells showed excellent dark storage stability at 65% humidity measured over 1000 h. An additive strategy was proposed for the $CsSnI_3$-infiltrated three-layer mesoscopic solar cells c-TiO_2/m-TiO_2/Al_2O_3/NiO/carbon [75]. 2-aminopyrazine was used as an additive to SnF_2. The pyrazine ring of APZ provides electrons and acts as a Lewis donor to stabilize Sn^{2+}. An efficiency of 5.12% was achieved with 0.40 V Voc, 21.7 mA/cm^2, and 0.59 FF. The solar cells were able to maintain 90% of their original efficiency after being stored in a glove box filled with N_2 for 100 h.

3.9. ETL/HTL Modification

Chen et al. modified the PEDOT:PSS surface by treating it with tetrafluoro-tetracyanoquinodimethane (F4TCNQ) [76]. The energy mismatch of PEDOT:PSS with Sn-PVK was resolved, as the PEDOT:PSS treated with F4TCNQ showed deeper energetic alignment. C=N group and F^- of F4TCNQ were coordinated to Sn^{2+} and halide ions. This coordination prohibited the defect density and passivated the PVK film. The proper energy cascade and reduced defect states at the HTL/Sn-PVK interface led to a jump in efficiency. An electron transport layer (6,6)-phenyl-C61-butyric acid hexyl ester (PCBH) with a flexible hexyl group was used to modify the charge collection at the ETL [77]. The hexyl group was able to enhance the interconnection with Sn-PVK and fullerene resulting in an improved photovoltaic performance of 9.21%. Chen et al. applied monolayer engineering to treat the PEDOT:PSS HTL layer [78]. PEDOT:PSS was treated with a 2PACz monolayer. This resulted in an improvement in conductivity due to the coupling of the O^- of 2PACz to the S^+ of PEDOT:PSS. This anchoring resulted in the VBM of PEDOT:PSS-2PACz being −5.73 eV, which was deeper than the −5.10 eV of PEDOT:PSS. The modified PEDOT:PSS-2PACz bilayer exhibited an improved mobility of 18.45 compared to the 8.85 cm^2/V·s of the original PEDOT:PSS. $FASnI_2Br$ had a VBM of 5.82 eV. Better energetic and crystallization properties led to an improved efficiency of 8.66% for the Sn-PVK solar cells with a bandgap of 1.66 eV. Nitrogen-doped graphene oxide ($N_{0.12}GO$) as an additive in PEDOT:PSS as HTL showed deeper work function and better energetic matching leading to improved Voc in the operation of Sn-PVK solar cells. An interlayer of Al_2O_3-$N_{0.12}$GO on PEDOT:PSS- $N_{0.12}$GO as HTL could suppress the interface defects, resulting in a high Voc of 0.961 V [79].

3.10. Flexible $FASnI_3$ Solar Cell

Portable $FASnI_3$ solar cells were realized by the additive strategy of graphite phase C_3N_4 (g-C_3N_4) [80]. g-C_3N_4 follows the size-effect to retard the crystallization rates by providing a crystallographic template and passivating the defect states. The hydrogen bonding between FA and g-C_3N_4 optimized the lattice matching and passivation on a flexible substrate. The 2D nature of g-C_3N_4 helped to improve the stability of the solar cells. A respectable efficiency of 8.56% was achieved in inverted structure solar cells based on a flexible polydimethylsiloxane substrate. The flexible solar cells maintained 92% of their original efficiency for 300 cycles at a 3 mm curvature radius. Padture et al. fabricated $FASnI_3$-based flexible solar cells by Ge alloying [81]. An amorphous GeO_2 was formed on the NiO_x-HTL layer to serve as a barrier to the Sn^{2+} oxidation and allow better bonding between the Sn-PVK and NiO_x layers. The Sn-PVKs were coated with 4-(aminomethyl) piperidinium diiodide. A record efficiency of 10.43% was obtained for the flexible solar cells. The cells showed near-stable and consistent performance under 700 h of illumination in a glove box filled with N_2. The cells were also stable in the 5 mm radius banding test for 1000 cycles.

3.11. Electrode Engineering

Han et al. proposed Sn-PVK solar cells without a hole transport layer fabricated on ITO glass [82]. They showed that the addition of $EDAI_2$ was essential to achieve better band bending and charge extraction in the case of HTL-free Sn-PVK solar cells. Such solar cells achieved a remarkable efficiency of over 10%. The fabricated HTL-free solar cells showed high stability. They retained 90% of the original efficiency after a heat test at 80 °C for 300 h in a N_2 medium. Diau et al. found that continuous photo illumination increases the temperature of solar cells and corrodes the Ag electrodes [32]. Thus, one of the stumbling issues with the stability of the solar cells was the corrosion of the Ag electrodes. Hatton et al. investigated the test of Sn-PVK solar cells soaked in ambient light [83]. They found that the combination of bathocuproine and copper electrodes can prevent the corrosion effect of I_2 (which is formed during the decomposition of Sn-PVK). In addition, the copper electrodes are more resistant to the ingress of water and O_2. However, the use of copper electrodes has the practical limitation that the energetics does not match the Sn-PVK photoactive layer, which is a limitation that must be tackled.

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