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A review on perovskite solar cells (PSCs), materials and applications

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ABSTRACT

In recent years, the perovskite solar cells have gained much attention because of their ever-increasing power conversion efficiency (PCE), simple solution fabrication process, flyable, light-weight wearable and deployable for ultra-lightweight space and low-cost materials constituents etc. Over the last few years, the efficiency of perovskite solar cells has surpassed 25% due to high-quality perovskite-film accomplished through low-temperature synthesis techniques along with developing suitable interface and electrode-materials. Besides, the stability of perovskite solar cells has attracted much well-deserved attention. In this article we have focused on recent progress of the perovskite solar cells regarding their crystallinity, morphology and synthesis techniques. Also, demonstrated different layers such as electron transport-layers (ETLs), hole transport-layers (HTLs) and buffer-layers utilized in perovskite solar-cells, considering their band gap, carrier mobility, transmittance etc. Outlook of various tin (Sn), carbon and polymer-based perovskite solar cells and their potential of commercialization feasibility has also been discussed.

1. Introduction

Day by day, the consumption of energy gradually increased globally with population. The reduction of non-renewable energy sources like fossil fuels causes the environmental issues, which lead to the advancement of usage of renewable energy. In this connection, the solar energy is considered as significant renewable-energy source because it is abundant, clean and inexhaustible. Also, the usage of solar energy will not damage the environment [1]. Last few years, many methodologies such as solar architecture [2], solar heating [3], artificial-photosynthesis [4], photovoltaics [5], photocatalytic water-splitting [6,7], etc. have been established to harness solar-energy. Amongst, photovoltaics gained much attention which uses solar-energy and converts sun light into electricity via photovoltaic effects.

Solar-cell is a photovoltaic device that can produce electricity by using solar energy. Usually, the solar-cells are categorized into three-generations. The first-generation solar-cells are based on wafer, second-generation solar-cells are thin film based, whereas third-generation solar-cells employ organic structures. Many years, first and second-generation solar-cells have been used, however high cost, complicated preparation technology, non-eco-friendliest nature limits their usage. Hence, scientists are looking for new solar cell materials which are cost effective and pollution free. So far, different types of solar cells like polycrystalline-silicon (mc-Si cells) & single-crystalline silicon solar-cells (c-Si cells) [8–11], CIGS solar-cells [12,13], CdTe-based solar-cells [14,15], quantum dot sensitized solar cells [16], organic-photovoltaics [17] and perovskite solar-cells [18] have been reported. Specifically, the power conversion efficiency and cost of the materials play a critical role in commercialization of solar-cells. Until now, third generation silicon based solar cells are dominating with large power conversion-efficiency (PCE) of 25–26%. Nevertheless, new classes of third-generation solar-cells termed as perovskite solar-cells are an alternative for silicon solar-cells which can exhibit the PCE of 22.1% [19–22]. This article will summarize recent advances in perovskite solar cells; especially we have focused on carbon-based, tin-based and polymer-based perovskite solar cells and their applications.

2. Perovskite solar-cells

In general, photovoltaic performance of the perovskite solar cells is ascribed from their intrinsic properties like high absorption coefficient [23], tunable band gap [24], large carrier diffusion-length [25], ambipolar carrier-transport ability [26] and carrier

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mobility [27]. Especially, organic-inorganic hybrid-perovskite (OHIP) materials are the favorable candidates for establishing efficient and cost-effective solar-cells. In 1990s, Mitzi and coworkers first demonstrated the OHIP material for light emitting diode and transistor applications with chemical-formula ABX_3, where A belongs to organic-cation group like Cs^+, methylammonium (MA^+, $CH_3NH_3^+$), formamidine (FA^+, $CH_3CH_2NH_3^+$), B belongs to inorganic cation group like Sn^{2+}, Pb^{2+} and X stands for halide ions such as I, Cl, Br etc [28–35]. Further, in comparison with ordinary organic and inorganic semiconductors, the organic-inorganic hybrid perovskites exhibit unique optical and electrical properties. In addition, these OHIP materials consists large Bohr radius [36], weak binding energy [37], high dielectric constant [38], high carrier diffusion velocity and diffusion length [39–43] along with outstanding light absorbing capacity. Owing to all these advantages, the OHIP materials became the prominent candidates for fabricating highly efficient solar cells with low cost. Normally, the perovskite solar cells consist an absorber layer (for example: $CH_3NH_3PbX_3$), which is inserted between electron-transport layer (ETL) and hole-transport layer (HTL). When the perovskite-absorber is subjected to light, it inserts an electron and hole into n-type and p-type carrier-transporting materials in order to generate free charge carriers. The generated electrons reached the cathode by travelling thorough the mesoporous film and external circuit. Also, the condensed section of hole-transporting material (HTM) restores oxidized perovskite and helps to reach ground state. Accordingly, the hole extorted into HTM diffuses in counter-electrodes direction. Finally, recombines with electron and provides the current. The current generation depends on the thickness of the perovskite material [44–46]. The energy-levels and charge-transfer process of perovskite solar cells are shown in Fig. 1 (Fig. 1B of Ref. [47]).

In 2009, Miyasaka and coworkers first demonstrated the perovskite materials in solar cell applications [48]. They used $CH_3NH_3PbX_3$ as sensitizer in dye-sensitized solar cell (DSSC) which exhibit the PCE of 3.81%. Subsequent investigations disclosed that the OHIP materials are extremely interesting candidates for solar cell applications. Besides, due to abundant availability of the precursor elements in OHIPs along with the usage of simple preparation techniques like vacuum-deposition [49,50], the atmospheric solution-processing [51,52], etc., were established. A list of surprising breakthroughs is accomplished within short period of time, with the improvement of the PCE to as high as 23.3% [53]. Furthermore, in the advancement of perovskite solar-cells ETL plays a crucial role [54]. In traditional perovskite solar cells ETL is a colloidal thin-film of SnO_2 [55,56], TiO_2 [57,58], ZnO or their mesoporous systems [59,60], which consists large grain-boundaries and weak recombination at interface [61,62]. Specifically, semiconductor ETL possess natural defects arose due to oxygen vacancies and trap assisted recombination [63,64]. To overcome these shortcomings various researchers proposed several materials (single crystalline) for ETL in perovskite solar cells. In this view, nano sheet or atom thick of transition metal dichalcogenides such as MoS_2, WS_2, TiS_2 etc. [65,66], can have great potential to serve as ETL material. Because atom-thick and single-crystal nanosheet is virtually defect free [67,68]. Besides, the thin structure offers rapid transportation of charge carriers to the electrode [69,70]. Owing to low trap density and high carrier transportation ability, MoS_2 is widely used as ETL [71]. Sometimes it can also be served as HTL due to its ambipolar property [72]. For instance, in 2016, Kim et al. [73] fabricated a perovskite solar cell by using MoS_2 as HTL which exhibit the PCE of 9.53%. Followed by, Das et al. [74] used the MoS_2 thin film as HTL in an inverted p-i-n heterojunction planar perovskite solar cell which shows the PCE of 6.01%. In continuing this, first time Malek et al. [75] demonstrated the preparation of atom-thick MoS_2-nanosheets directly on indium tin oxide(ITO)-substrate. They reported that homogeneity of the nanosheet enhances with the substrate temperature and optimum at 200 °C. In addition, they observed that the usage of the prepared materials as ETL enhances the performance, stability and interfacial charge transfer capability of the perovskite solar-cell. Further, the PCE of the solar-cell is increased with decrease of the size of the MoS_2 layer. Especially, the 5-atom thick of MoS_2 nanosheet ETL exhibit short-circuit current-density (J_{sc}) of 16.24 $mAcm^{-2}$, open circuit-voltage (V_{oc}) of 0.56 V, fill-factor (FF) of 0.37 and PCE of 3.36%. Moreover, the MoS_2-based solar cell can retain 90% of its initial PCE after continuous operation for 80 s under the irradiation of sunlight at maximum power point.

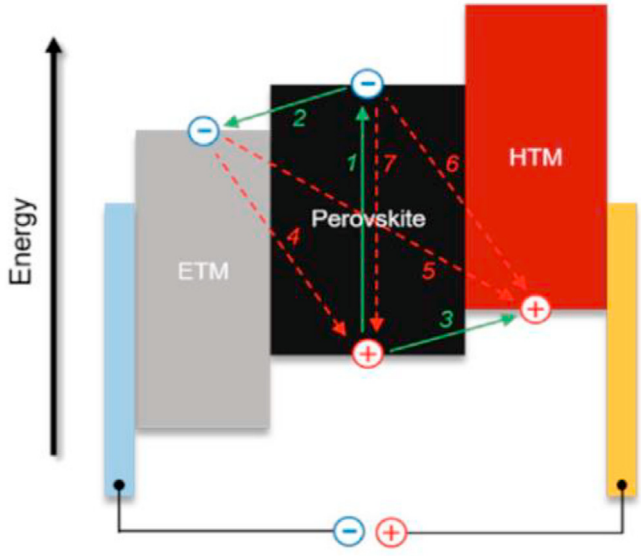

Fig. 1. The energy-levels and charge-transfer process of perovskite solar cells (Fig. 1B of Ref. [47]).

In addition, HTL also plays a key role in enhancing the performance of the photovoltaic devices. Besides, huge efforts have been devoted to improve the conductivity of HTL also to suppress the recombination of charge carriers at HTL and perovskite interface by modifying the HTL through dopants [76–78]. Amongst various HTMs, 2, 2′, 7, 7′-Terakis [*N*,*N*-di(4-methoxyphenyl)amino]-9,9′-spirobifluorene(spiro-OMeTAD) exhibit some special features such as high solubility, glass transition temperature, etc. Nevertheless, due to insufficient oxidation states raw spiro-OMeTAD shows poor PCE. Usually, high oxidation time is required for attaining good photovoltaic performance in perovskite solar cells [79]. In this regard, in order to step up the oxidation time of spiro-OMeTAD, Kim et al. employed oxygen plasma [80]. But, under plasma the perovskite phase is decomposed to PbI_2., this can be resolved by doping of trivalent (p-dopants) materials which can also enhance the hole-transport properties of spiro-OMeTAD. So far, metal-organic complexes, metal-oxides, organic molecules etc., are effectively active p-dopants [81–83]. However, low solubility and complex decomposition methods are limiting their applications in perovskite solar cells. Further, cobalt complexes and $FeCl_3$ can act as efficient p-type dopants which can oxidize spiro-OMeTAD and also generate new holes to enhance the conductivity [84–86]. Also, acid additives can enhance the oxidation process and shortens the aging time of perovskite solar cells [87]. Recent investigations are focused on designing the HTL by using acid additives and modifying the morphology of spiro-OMeTAD. In continuing this, Guan et al. [88] recently investigated the effect of benzoic-acid on oxidation process of spiro-OMeTAD. They reported that the doping of benzoic acid fastens the oxidation-process and enhances the hole

transportation capability of HTL. Moreover, optimization of doping concentration effectively reduces the hysteresis in HTL-based perovskite solar cell. Also exhibits improved PCE of 16.26% under standard AM 1.5G illumination.

Furthermore, Yang et al. [89] synthesized a low cost and dopant free fluorinated spiro-OMeTAD-2,7-diamine (2 mF-X59) for solar cell applications. They reported that, the prepared material is sensitive and dopant free which can be applied as HTL into $CsPbI_2Br$ perovskite solar cells. Up next, they modified the surfaces of $CsPbI_2Br$ perovskite and 2 mF-X59 using 2,3,5,6-tetrafluoro-7,7,8,8-tetracyanoquino-dimethane (F4-TCNQ). The modified and dopant free $CsPbI_2Br$ perovskite solar cells shows the PCE of 14.42% with an impressive V_{OC} of 1.23, which is very high compare to doped ones. Besides, the solar cells based on proposed HTL materials exhibit the extraordinary stability i.e., it shows 94% retention of initial PCE even after aging in air for 30days without encapsulation. So, the prepared dopant free HTL is a prominent candidate for preparing high performance $CsPbI_2Br$ perovskite solar cells. For the first time Zhao et al. [90] demonstrated the preparation of CuSeCN-films for HTL application in p-i-n perovskite solar-cells utilizing the organic amines solvent (H_2O/ETA/EDA/DTA) mixture with 2:6:1:1 vol ratio for solution-processing. They reported that the prepared HTM-based perovskite solar-cells obtained the PCE of 15.61% at forward-scan whereas at reverse-scan it shows the PCE of 15.97%. Also, CuSeCN-based perovskite solar-cell displays almost negligible hysteresis and excellent long-term stability. Hence, CuSeCN-films have the great potential to serve as an HTM for photovoltaic applications.

Due to high electron mobility [91], variety of nanostructured morphology [92,93] and multiple growth methods, ZnO can also be a promising candidate for solar cell applications [94,95]. As we know that the function of perovskite solar cell is depends on crystalline quality and surface-morphology of the perovskite capping-layer. Besides, the selection of suitable solvent for preparation of ZnO solution can significantly regulate the morphology (i.e., grain size and surface rough ness) of the active layer of the perovskite, accordingly the solar cells performance. Recently, with help of cost-effective ultrasonic bath technique, Ahmadi et al. [96] synthesized ZnO nanoparticles by using three different solvents 2-methoxyethanol (2 ME), isopropyl alcohol (IPA) and ethanol to fabricate ETL for perovskite solar-cells. The outcomes of structural, morphological and device performance investigations are evinced that the ZnO layer prepared by using 2 ME as solvent exhibit good characteristics among all the prepared ZnO layers. Moreover, the perovskite solar cell fabricated using methyl ammonium lead iodide ($MAPbI_3$) as capping perovskite layer and ZnO (2 ME) as ETL shows excellent PCE of 22%. This is due to lowest defect density at ZnO (2 ME)/$MAPbI_3$ interface, larger grain sizes and good surface coverage by $MAPbI_3$. Hence, the ZnO based solar cells are suitable candidates for solar cell applications. Owing to high transmittance nature, ZnSnO (ZTO) can also be used as ETL in solar-cells. Usually, the oxygen-vacancies present in ZTO plays a vital role in the transportation of charge carriers. Further, the existence of large number of oxygen-vacancies is the major defect in ZTO. To overcome this, Miao et al. [97] demonstrated the effect of oxygen-vacancies in ZTO through fabricating ZTO-based solar-cells with doping of silicon in various doping concentrations. For this purpose, with the help of RF magnetron sputtering technique silicon-ZnSnO (SZTO) amorphous metal oxide films are prepared by varying the silicon content. Besides, they reported that the x-ray photo-luminescence spectra (XPS) analysis confirms the reduction of oxygen-vacancies with respect to silicon concentration. Moreover, the reduction of oxygen vacancies in SZTO causes the improvement in electronic extraction and transfer ability. By using the prepared SZTO as an ETL, they fabricated a perovskite solar cell which exhibits the maximum PCE of 13.4%, J_{OC} of 21.6 $mAcm^{-2}$, V_{OC} of 1.04 V along with FF of 0.67.

Further, TiO_2-based solar cells exhibit good PCE over 20%. Nevertheless, the TiO_2-ETL in n-i-p structured perovskite solar-cell causes instability and fast decay of short-circuit current-density (J_{SC}) under the irradiation of ultraviolet radiation [98]. Many researchers put their efforts to overcome these shortcomings or to protect the perovskite solar cells from UV-light induced degradation through employing an interface layer at TiO_2-ETL and perovskite [99–103]. Still, the performance of the device is affected owing to TiO_2 in all of the above reported techniques. So, there is a need to fabricate the stable perovskite solar cell with improved performance. In this view, many ETL materials which are stable under ultraviolet (UV) radiation gained much attention [104–106]. Amongst, owing to excellent electron-mobility and deeper conduction-band, $Mg_xZn_{1-x}O$ (MZO) can serve as an effective ETL material in perovskite solar cells which is also stable under ultraviolet irradiation [107,108]. In continuing this, recently Han et al. [109] demonstrated the MZO-based perovskite solar-cell with outstanding stability under UV-radiation. They reported that, compare to TiO_2, MZO shows superior carrier mobility and conduction mechanism. This property causes the reduction in charge growth at the interface of the MZO and perovskite also enhances the charge transfer between MZO-layer and perovskite. Further, they fabricated MZO-based device, at optimized state the device exhibits large open circuit voltage of 1.11 V along with high efficiency of 19.57%. In addition, even after one year of aging at room temperature, 40–80% relative humidity and 8 h ultraviolet irradiation, the proposed device exhibits 76% of retention of initial J_{SC} whereas under same conditions the TiO_2-based device shows only 12% of initial J_{SC}. Owing to excellent stability under UV-radiation, the fabricated device shows reduced electron trap state density in MZO-ETL. Indeed, zinc-interstitial sites and oxygen-vacancy sites of MZO-ETL strongly opposes the degradation of perovskite-layer under the exposure of UV-radiation. Hence, the proposed MZO-ETL can serve as potential candidate for fabricating stable UV-resistive perovskite solar-cells.

In order to enhance the conductivity of ETL layer, Teimouri et al. [110] demonstrated the effect of lithium (Li) doping on TiO_2. In this connection they prepared Li-doped TiO_2 films by using ultrasonication technique. They reported that, in solution form Li-doped TiO_2 shows improved conductivity and decreased solar power loss. In addition to increased conductivity Li-doped TiO_2 films provides faster electron transport. Besides, to study the effect of various Li-concentrations on efficiency of perovskite solar cells, they modeled the structures by using solar cell capacitance simulator (SCAPS). The perovskite solar cell consisting of an ETL with 0.3 M Li-doped TiO_2 exhibited the PCE of 24.23% which is almost 1.97% larger than the undoped composition. Furthermore, in comparison with the pure TiO_2, doped TiO_2 shows lower trap-density at the interface of the absorber and ETL. All these outcomes are evinced that the Li-doped TiO_2 can also be a prominent candidate to serve as ETL-layer in perovskite solar-cells. With the help of sol-gel synthesis technique, Yang et al. [111] synthesized different compact anatase TiO_2-ETLs (c-TiO_2) by varying the concentration of TiO_2. It is observed that among all the prepared ETLs, the ETL with 2.0 M concentration of TiO_2 shows dominant properties. Besides, the fabricated c-TiO_2 based perovskite solar-cells accomplished a high PCE of 16.11% with FF of 0.656, V_{OC} of 1.1 V and J_{SC} of 22.32 mA/cm^2. So, c-TiO_2 based perovskite solar-cells show a new direction for preparing low temperature perovskite sola-cells with better efficiency. Fig. 2 shows the schematic representation of c-TiO_2 based perovskite solar cells (Fig. 1b of Ref. [111]).

With the help of simple sintering technique, Zhang et al. [112] designed $MgTiO_3$-coated TiO_2 mesoporous scaffold layers with

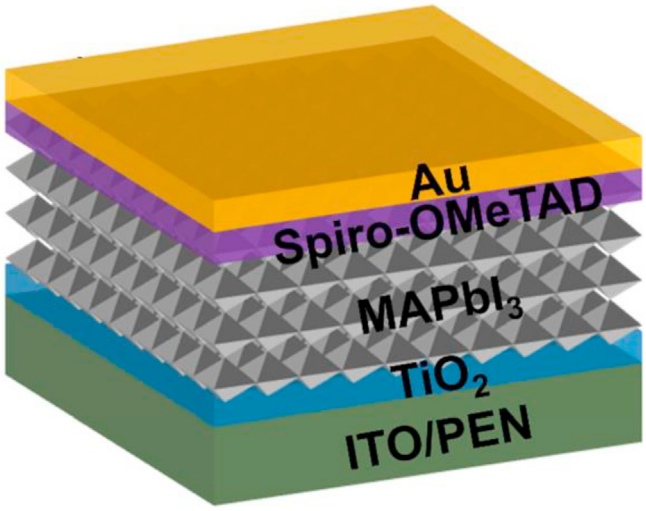

Fig. 2. Schematic representation of c-TiO_2 based perovskite solar cells (Fig. 1b of Ref. [111]).

different treating concentrations for solar cell applications. They observed significant improvement in the photovoltaic performance after employing the prepared scaffolds as shell layers. Besides, as a blocking layer the $MgTiO_3$ shell effectively retard the recombination of the charges at $MAPbI_3$ and TiO_2 interface. Also, the $MgTiO_3$ introduction improves the crystallinity of $MAPbI_3$ crystals which play a critical role in the formation of high-quality perovskite-film. The perovskite solar-cell based on optimum treating concentration of 0.10 M exhibit the maximum PCE of 10.39%. Moreover, the perovskite device retains 88.35% of its initial PCE even after 1008 h stored at air with normal humidity. So, the long-term stability, high PCE and simple preparation process suggest that the $MgTiO_3$-coated TiO_2 mesoporous scaffold layers are feasible materials for next generation photovoltaic devices. In addition, due to high chemical stability, high electron-mobility, wide-bandgap and antireflective nature, the indium oxide (In_2O_3) thin-films have great potential to serve as ETL-materials in perovskite solar cells [113,114]. From previous literature it is clear that the hygroscopic nature of In^{3+} causes the production of pinholes, cracks and imperfect morphology. Because, In^{3+} cations react with water molecules at the time of synthesis [115–117] of samples. This could bind the advancement of In_2O_3 as a prominent ETL-candidate in solar cells. To improve the performance of photovoltaic device it is desirable to prepare defect free In_2O_3. In this connection, for the first time Zhang et al. [118] demonstrated the production of compact In_2O_3-films processed at low-temperatures from highly stable indium precursor solution to serve as stable ETL in perovskite solar cells. Actually, indium precursor does highly-stable in ethanol comprise 0.2% of water and addition of acetylacetone (acacH) as a chelation ligand to the solution effectively prevent the hydrolysis reactions by indium [119]. All these contribute the establishment of In_2O_3-film at low temperatures about 200 °C and at 40–50% relative humidity. Fig. 3 shows the schematic representation of stability of indium-precursor solutions with and without acetylacetone as chelation ligand (Fig. 1a of Ref. [118]). The prepared In_2O_3-film as ETL significantly improves the transportation of charges and electron extraction at the ETL/perovskite interface. Accordingly, the air processed perovskite solar cells based on compact-In_2O_3 film exhibits high PCE of 13.97% whereas pristine In_2O_3-film shows the PCE of 9.81%. Along with high PCE the proposed indium-based perovskite solar cell shows long term stability and retains 94% of its PCE even after 31 days of storage. All outcomes disclosed that, In_2O_3 films as ETL significantly improves the stability and PCE of air-processed perovskite solar-cells.

Tseng et al. [120] introduced a new technique for enhancing the PCE of perovskite solar-cells by optimizing the morphology, interface and charge concentration of $Cu_2O/MAPbI_3/SiO_2$ heterostructure. With the help of RF-magentron sputtering, they fabricated Cu_2O-HTL and ultrathin SiO_2-ETL. Besides, the device consisting Cu_2O as HTL and SiO_2 as ETL exhibit the improved PCE as well as reduced recollection and recombination of the charge-carriers. Moreover, these Cu_2O-HTL/SiO_2-ETL layers significantly inhibit the pinholes and defects which attain high V_{OC} of 1.12 V and FF of 78.60%. As result of enhanced V_{OC}, the device shows highest PCE of 18.4%. Hence, some of the inorganic compounds such as Cu_2O, SiO_2 etc., are also prominent candidates for the applications in perovskite solar-cells.

Recently, Zhu et al. [121] demonstrated the fabrication of high-performance perovskite solar-cells. For this purpose of achieving high PCE, they inserted ultrathin-mesoporous-TiO_2 (thin-*m*-TiO_2) layer between the interface of compact-TiO_2 and perovskite-film in a planar perovskite solar cell, thin-*m*-TiO_2 layer serves as interfacial modifying layer. They observed that, introduction of thin-*m*-TiO_2 significantly enhances firmness and grain-size of the perovskite-films. In addition, compare to traditional planar and mesoporous perovskite solar cells, the proposed solar cells containing the interfacial modifying layer display enhanced performance with PCE of 18.5% and low hysteresis-coefficient of 4.5%. Moreover, the interfacial modifying-layer not only improves the efficiency of carrier-separation but also maintain high transportation capacity, which may play a vital role in the improvement of the efficacy of next generation perovskite solar-cells. Further enhancement of the PCE, Hu et al. [122] introduced a new strategy called multi-functional interface layer (MFIL) method. In MFIL technique the roles of different parts such as electron transport layer, near infrared photocurrent enhancement, moisture barrier, ion migration suppression and trap passivation in inverted perovskite solar cells are combined. Owing to this, MFIL based device shows improved device efficiency and long-term stability. Fig. 4 represents the device based on MFIL with all integrated roles of all layers (Graphical abstract of Ref. [122]). Besides, the device fabricated with MFIL presents the large PCE of 21% along with outstanding stability upto 1700 h without encapsulation under different conditions such as thermal, moisture and light. Finally, the studies on molecular orientation at MFIL and perovskite interface disclosed that the design of advanced interlayers enhances the interface molecular bonding, reduces the trap density accordingly improves the device performance. Hence, the outcomes suggested that the proposed MFIL approach opens a new window for enhancing the performance of next generation perovskite based photovoltaic devices. Yun et al. [123] demonstrated the preparation of ZnO nanorods via low temperature water bath for perovskite solar cell applications. They reported that, on fluorine doped tin oxide (FTO) substrate, well ordered ZnO nanorods are grown whose length is varies with respect to reaction time. Fig. 5 shows the ZnO-nanorods with different lengths on FTO substrate (Graphical abstract of Ref. [123]). Further, the prepared materials have some of the advantages such as shortened route for electron transportation, improved contact area, high visible transmittance and compact interface. Considering all these advantages they fabricated solar cells by using the prepared ZnO nanorods as an ETL-layer. The solar-cell fabricated with optimum sized ZnO nanorods exhibits the highest PCE of 14.22% under AM of 1.5G illumination. Therefore, the ZnO-based materials can also serve as efficient ETLs in high performance perovskite solar-cells.

In the advancement of perovskite solar cells, researchers are combined the photovoltaic-systems with tandem solar-cells to

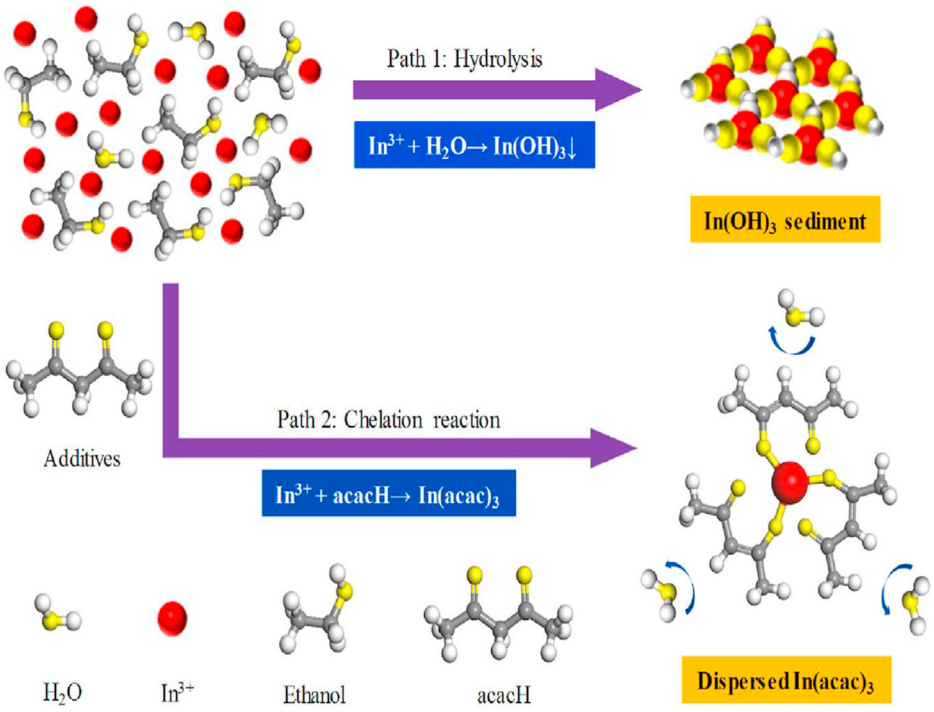

Fig. 3. Schematic representation of stability of indium-precursor solutions with and without the addition of acetylacetone (Fig. 1a of Ref. [118]).

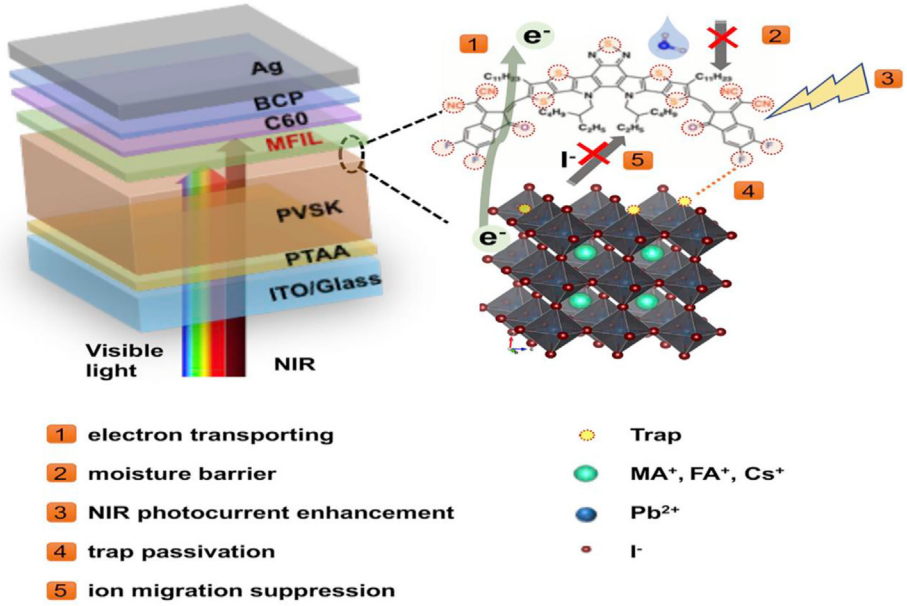

Fig. 4. Multifunctional interface layer with integrated roles of all layers (Graphical abstract of Ref. [122]).

generate high PCE. However, this concept yet to be implemented successfully for the solar-cells based on perovskites. Usually, a tandem solar cell can be designed by using two terminals called monolithic device and four terminal devices are named as mechanical stack. The theoretical limit of PCE for two terminal devices is of 45.7% and four terminal devices is of 46.1% [124]. Owing to simple architecture design and higher PCE the two terminal tandem solar cells gained much attention than four terminal tandem solar cells [125,126]. So far, the devices with two terminal tandem solar-cells based on perovskite/perovskite, perovskite/crystalline-silicon etc., has been reported [127–132]. All the above said tandem perovskite solar cells exhibit unique advantages such as compatibility with technologies, low temperature processing and low fabrication costs [133–135]. Further, based on opto-electronic analysis the single junction silicon counterpart exhibits the PCE of 26% whereas crystalline-silicon tandem-perovskite solar cell can achieve the PCE of 30% [136,137]. Nevertheless, perovskite materials using in solar cell facing stability problems, due to this the usage of tandem converter in perovskite solar cells received little attention. In this connection, the researchers are focused on to

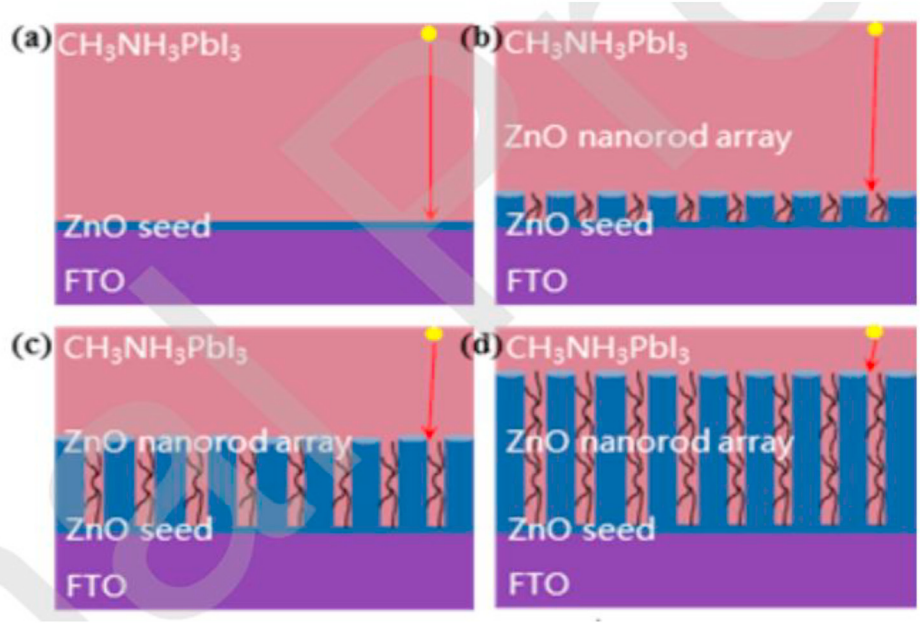

Fig. 5. Illustration of growth of ZnO nanorods on FTO substrate (Graphical abstract of Ref. [123]).

overcome these shortcomings of tandem perovskite solar cells. Recent reports disclosed that FA-based perovskite exhibit more stability than others owing to improved photostability and thermal stability [138–140]. Recently, Cherif and Habib [141] reported the possibility of using perovskite semiconductor with varied bandgap as tandem concentrator combined with crystalline silicon. Besides, they developed an optoelectronic model to optimize and also to achieve the highest PCE of proposed tandem-perovskite concentrator solar-cell. Interestingly, under the exposure of high solar concentration (70 suns), the proposed device exhibits large peak efficiency of 34.62%, same exhibit 30.52% under 1 sun. Therefore, the outcomes are evinced that, the proposed model shows a new pathway in the advancement of tandem perovskite/c-Si solar-cells to attain higher efficiencies under different conditions.

As we know that perovskites are ionic-semiconductors which consists huge number of mobile-ions. These mobile ions are responsible for overall conductivity. Few studies are available for movement of ions at steady state and dynamic conditions [142–144]. Further, in the reported techniques various admittance spectroscopy is utilized to disclose the role of mobile ions in dispersion of dielectric constants with respect to compositions. However, in perovskite materials the conductivity related ionic species can be extracted with the help of deep level transient spectroscopy (DLTS) [145,146]. In addition, the DLTS and current-transient DLTS(CDLTS) can differentiate the common effects instigated by capturing of charge carriers, also emission by profound traps in bandgap of perovskite-materials. Besides, the peaks observed in DLTS/CDLTS are formed due to the processes of electron trapping and de-trapping. Therefore, one can apply the similar procedure to investigate the defects in multi cation perovskite solar cells with two-dimensional (2D) perovskites and three-dimensional (3D) perovskite layers. In general, the 2D-perovskites with general formula $R_2(A)_{n-1}B_nX_{3n+1}$ plays a pivotal role in stability improvement of the perovskite solar-cells [147]. Recently, few research groups reported the fabrication of 2D/3D bi-layered perovskites for generating highly-stable photovoltaic device [148]. Until now, the PCE of 20.75% can be achieved by the reported techniques [149] in which bulk cation is deposited on preformed 3D-perovskite surface to produce *in situ* evolution of 2D-layer [150]. The 2D/3D perovskite solar cells developed through these methodologies can exhibit outstanding charge transport capacity, decreased current voltage hysteresis and charge recombination also exhibit 85% retention of its initial PCE even after 800 h illumination at the temperature of 50 °C. Recent year's 2D-perovskite layer is applied as passivating layer in perovskite solar-cells. One can prepare 2D perovskite layer through introducing large size hydrophobic-cation into the 2D perovskite crystal lattice. The inserted cation suppresses the intrusion of moisture also improves the stability of perovskite solar cell. In continuing this, Shikoh et al. [151] demonstrated the DLTS and reverse-DLTS (RDLTS) to categorize n-i-p structured 2D/3D perovskite solar-cells. Fig. 6 illustrates the schematic representation of prepared 2D/3D-perovskite device (Fig. 1b of Ref. [151]). Furthermore, they reported that the electron/hole traps and existence of deep traps of electrons and holes in bandgap of perovskite can be measured by using DLTS and RDLTS. Besides, the photoinduced voltage transient spectroscopy (PIVTS) analysis revealed that 2D/3D perovskite structures are robust than 3D structures, in which the mobile ions play a dominant role also provides higher PCE around 20%.

3. Carbon-based PSCs

In modern perovskite solar-cells, organic-inorganic halide perovskites play a pivotal role. However, noble metals and organic HTL materials are expensive, which is unfavorable to the commercialization of perovskite solar cells [152,153]. To overcome this, by means of reducing the fabrication cost and to enhance the stability of the solar-cell, many researchers proposed HTL-free carbon-based perovskite solar-cells (C–PSCs) [154,155]. Nowadays, the HTM free C–PSCs gained much attention and those devices attained the maximum PCE of 16.37%. Also, the manufacturing cost of C–PSCs is very low and they exhibit outstanding stability against moisture [156]. Despite of the advantages, the C–PSCs have some limitations. In comparison with the conventional perovskite solar cells the C–PSCs exhibit low PCE which is considered as one of the major limitations of C–PSCs. Moreover, The ETL and perovskite interface is the only interface in C–PSCs which efficiently split up the electron-hole pair. Further, the ETL between the electrode and

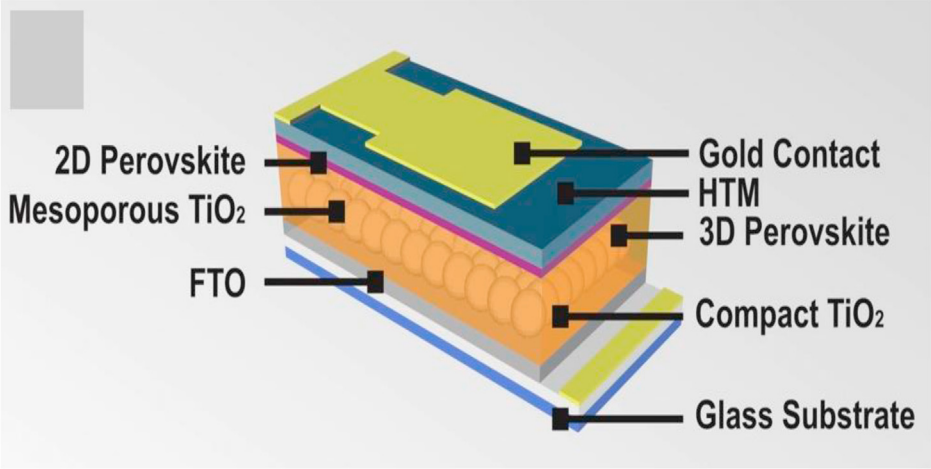

Fig. 6. Schematic representation of 2D/3D perovskite device (Fig. 1b of Ref. [151]).

perovskite absorbing layer helps to extract the photogenerated electrons from absorbing layer and to transport them to electrode. Hence, ETL-material has significant role in enhancing the stability and performance of C–PSCs. Also, ETL act as hole-blocking layer to suppress the recombination of electron-hole pair [157,158]. In this regard, modern researchers are focused on to develop optimized ETL materials with advanced features. Owing to superior opto-electronic properties, nontoxicity, high chemical stability etc., TiO_2 is the material which is extensively employed as an ETL in perovskite solar-cells [159,160]. In perovskite solar cells the photo generated charge carriers move across the perovskite and ETL interface. Especially, in case of HTL free C–PSCs the leakage of photocurrent takes place due to the exposure of perovskite layer by ETL, which in turn affects the performance of the device. To avoid this insulating layer of SiO_2 and ZrO_2 is introduced between them to avoid direct contact also to improve the efficiency of the device [161,162]. Further, the usage of methylammonium (MA), amino groups also help to increase the performance of TiO_2-based C–PSCs. Nevertheless, traps and oxygen-vacancies at the surface, interaction of TiO_2 with amino groups causes to the reduction in PCE of C–PSCs [163–166]. Recent reports are evinced that the incorporation of ferroelectric oxide (e.g. $PbTiO_3$) between the perovskite and TiO_2 significantly improves the PCE of C–PSCs [167]. For further enhancement of the electron transport capacity, Tao et al. [168] prepared TiO_2@$PbTiO_3$ core-shell via in-situ reaction and used as mesoporous ETL in C–PSCs. They observed that the coating of $PbTiO_3$ significantly reduces the defect density on the TiO_2 surface and also the interfacial charge recombination. Due to reduction of trap states, the device based on TiO_2@$PbTiO_3$ ETL shows good PCE. They reported that TiO_2@$PbTiO_3$ ETL based C–PSCs exhibit the improved PCE of 7.97% which is larger than $PbTiO_3$-free C–PSCs PCE (6.55%).

Further, Du et al. [169] demonstrated the preparation of methyl ammonium lead bromide ($MAPbBr_3$) by an optimized two-step sequential deposition technique for carbon-based perovskite solar cell applications. In first they introduced small amounts of MABr in to the precursor solution of $PbBr_2$ to prepare $MAPbBr_3$ perovskite film with low trap density, improved crystallinity and longer carrier life time followed by fabrication. They reported that, the prepared perovskite films are utilized in the fabrication of HTL free C–PSCs. Those perovskite solar cells exhibit the high PCE of 7.64% under optimized condition. Besides, the prepared C–PSCs exhibit high thermal stability i.e., the designed carbon based $MAPbBr_3$ perovskite solar cells show 95% retention of its initial PCE even after 120 h of storage at the temperature of 353 K and relative humidity of 40–70%. These are highly stable at room temperature i.e., in dry air the PCE of as prepared C–PSCs remains unchanged for almost a year. So, this two-step sequential deposition technique shows a new way for fabricating highly stable carbon-based perovskite solar-cells. In addition, owing to excellent luminescent properties inorganic $CsPbBr_3$ quantum dots are recognized as promising light emitting materials for photovoltaic applications. First, Zeng et al. [170] presented that the inorganic $CsPbBr_3$ perovskite-based LEDs, which exhibit the luminescence intensity of 946 $cdcm^{-2}$ with external quantum efficiency of 0.12%. There are many reports available on luminescent properties of inorganic $CsPbBr_3$ perovskite-based LEDs [171–173]. Besides, the optical band gap of $CsPbBr_3$ based perovskite is 2.3 V and it exhibit outstanding stability in limited cell efficiency. The ambient stability, balanced band gap and simple preparation process make that $CsPbBr_3$ based perovskites are the promising candidates for designing new carbon-based inorganic perovskite solar-cells. With the help of dual source evaporation technique Ma et al. [174] demonstrated the carbon-based $CsPbIBr_2$ perovskite solar-cells which show the PCE of 4.7%. Next, Zhu et al. [175] reported the properties of carbon based $CsPbIBr_2$ perovskite solar-cells fabricated by utilizing precursor solution of $CsPbIBr_2$. The fabricated C–PSCs display the maximum PCE of 5.7%. Recently, with the help of one step spin coating technique, Wang et al. [176] prepared high quality $CsPbIBr_2$ perovskite film through optimizing preheating and post annealing temperatures of substrate. They reported that the optimized temperatures greatly improve the crystallinity and light absorbing capacity of as prepared perovskite films. Also, reduces the recombination of the charge carriers. Besides, the fabricated C–PSC by using $CsPbIBr_2$ perovskite film exhibits the maximum PCE of 8.10% along with high V_{OC} of 1.27 V.

For further enhancement of the stability and efficiency of C–PSCs, Pei et al. [177] prepared $CsPbBr_3$ perovskite film via multistep solution spin coating technique. In the preparation process first $PbBr_2$ film is modified by $BiBr_3$ layer followed by CsBr coating, as a result $CsPbBr_3$ film is formed. The attained perovskite film exhibits an excellent crystallinity and improved carrier extraction property. Owing to these carbon-based CsPbBr3 perovskite solar-cells exhibits the improved PCE of 8.73%. Fig. 7 shows the pictorial representation of the carbon-based $CsPbBr_3$ perovskite solar-cell (Fig. 1c of Ref. [177]). Hence, the proposed multistep solution spin coating technique shows a new route for preparing the high-quality perovskite films.

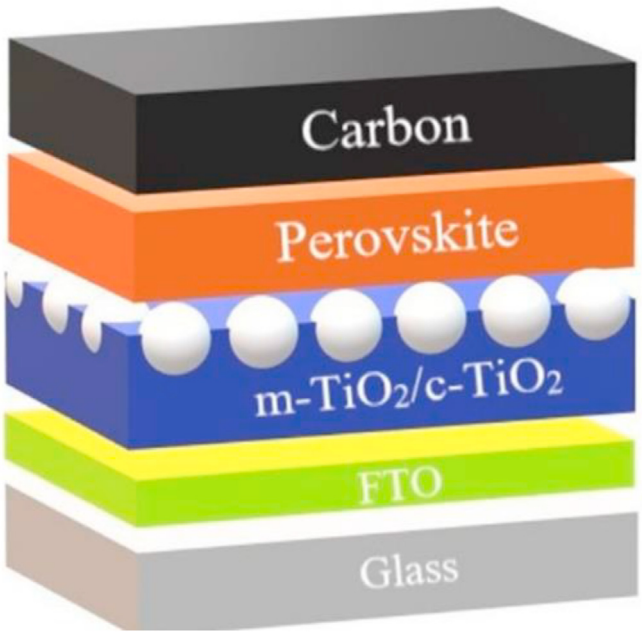

Fig. 7. Pictorial representation of the carbon based $CsPbBr_3$ perovskite solar cell (Fig. 1c of Ref. [177]).

Likewise, improvement of local contacts at the interfaces of HTM/Perovskite & HTM/back-electrode lead to effective photo-generated charge-extraction for carbon materials incorporated with HTMs, which also be a factor of increasing the performance of the photovoltaic device [178]. In this regard, Zong et al. [179] demonstrated a facile one-step pyrolysis technique to prepare MoO_2/N-doped carbon nanospheres by employing inorganic-organic-molybdate/polyaniline-pyrrole hybrids as a precursor. Actually, the precursors in liquid phase causes to MoO_2 dispersion uniformly in MoO_2/N-doped carbon nanospheres. Besides, nitrogen (N) doping enhances the conductivity of the nanospheres and also improves the contact between HTM and perovskite surface [180]. They reported that the usage of as-prepared MoO_2/N-doped carbon nanospheres as HTM in C–PSCs reduces the surface defects, energy offset at perovskite/carbon interface also promotes hole extraction, which results reduced charge recombination and energy loss in perovskite solar-cells. Hence, the MoO_2/N-doped carbon-nanospheres based $CsPbBr_3$ perovskite solar cells display superior PCE. Zong et al. [179] fabricated $CsPbBr_3$ perovskite solar cell by employing the prepared nanospheres as HTMs, which attained the maximum PCE of 9.4%. Besides, without HTM the same device shows the PCE of 6.86%, which is very small compare to the device with HTM. In addition, the fabricated solar cell is highly stable (it is stable over 800 h at 80% relative humidity). All these outcomes are evinced that the C–PSCs with inorganic HTMs exhibit superior performance than the C–PSCs without HTMs.

In continuing this, Yang et al. [181] proposed a facile technique to prepare carbon-electrode for C–PSCs. The proposed method includes hot pressing of a free-standing carbon-film on HTL of perovskite solar cells. Owing to mono-plasticity of prepared carbon-films, conductivity of the electrode enhances over 10 times and hot-pressing process strengthens the adjacent layer of carbon electrode. They reported that, at optimized applied temperature of 80 °C, significant improvement in the mechanical and electrical properties takes place, which also lead to improvement of device performance. Fig. 8 illustrates the statistical photovoltaic parameters of prepared samples at various temperatures (Fig. 4 of Ref. [181]). Besides, the C–PSC fabricated by using the prepared hot-pressed carbon electrode exhibits the PCE of 15.3%, which is 70% greater than the device at normal temperatures. In addition, C–PSC retains 93% of its initial PCE even after using 80 days without encapsulation at the relative humidity of 55–70%. All these parameters suggest that the proposed facile technique provides a new way to fabricate highly efficient next generation perovskite solar cells.

4. Tin (Sn)-Based PSCs

As we know that, the perovskite solar cells contain lead (Pb). In fact, the lead-based perovskite solar cells exhibit outstanding performance. However, toxicity of Pb increase the environmental damage also it is a barrier for mass production and commercialization of perovskite solar cells. Hence, it is important to introduce Pb-reduced or Pb-free perovskite to enhance the affordability of perovskite solar cells [182–186]. In order to replace the toxic lead in perovskite unit cells one should consider the stability and ionic radius of the perovskite structure. As per Goldschmidt's tolerance & octahedral-factor, the divalent metal-ions like calcium (Ca) [187,188], tin (Sn) [189–191], barium (Ba) [192], strontium (Sr) [193] etc., are the promising candidates for substituting Pb in perovskite unit cell. Amongst, Sn gained much attention owing to similar coordination geometry and electron configuration with Pb. Generally, Sn-based perovskites consisting of the formula $ASnX_3$ are the best alternative to replace Pb. In comparison with Pb-based devices, the Sn-based perovskite solar cells exhibit superior carrier mobility, bandgap, low excitation binding energies, short circuit current density and theoretical PCE of 33%. Nevertheless, the efficiency of Sn-based perovskite solar-cells is much smaller (10%) than that of Pb-based perovskite solar-cells. Also, the stability of perovskite solar-cell is strongly affected due to oxidization of Sn^{2+} to Sn^{4+} [194]. In this respect, the partial replacement of lead by divalent metal-ion can improve the performance of perovskite solar cell without causing any damage to environment. Continuing this Ji et al. [195] demonstrated the perovskite solar cell with Pb–Sn mixed triple cation, which exhibit the PCE of 16.10%. Furthermore, the incorporation of bulky organic-ligands modulates the orientation and growth of the grains, which inturn causes to enhance the out of plane photoinduced bulk-polarization as well as the spin-orbit coupling (SOC). These are responsible for photovoltaic-performance in 2D/3D-perovskite solar-cells. Especially, photovoltaic action is brightened by SOC i.e., enhanced SOC significantly improves the spin-conversion from optically induced states. The bright-states are formed due to spin allowed recombination whereas the spin forbidden recombination is responsible for dark states. In contrast, the grains which are oriented in out of plane in 2D/3D-perovskites with condensed traps are auspicious to align optical-transition dipoles which improve photovoltaic actions. Recently, with the help of anti-solvent engineering technique Zhang et al. [196] introduced the bulky organic cations of PEA^+ into 2D/3D Pb–Sn alloys to fabricate perovskite solar-cells with high efficiency. After incorporation, organic cations modulate the growth-oriented alignments and SOC in Pb–Sn perovskites, which induces the photoinduced bulk-polarization. The enhanced SOC and bulk polarization improves the photovoltaic actions. They reported that the 2D/3D-Pb–Sn alloy-based perovskite solar-cells exhibit the high PCE of 15.93%.

We know that the ETL plays a key role in facilitating high device performance in perovskite solar-cells [197,198]. Owing to their intrinsic properties such as high thermal and chemical stability, high dielectric constant, high conductivity etc., metal oxides gained much attention in the advancement of ETL materials for solar cell applications [199]. To date, TiO_2 is the material which is commonly utilized in making highly efficient perovskite solar cells [200]. Still,

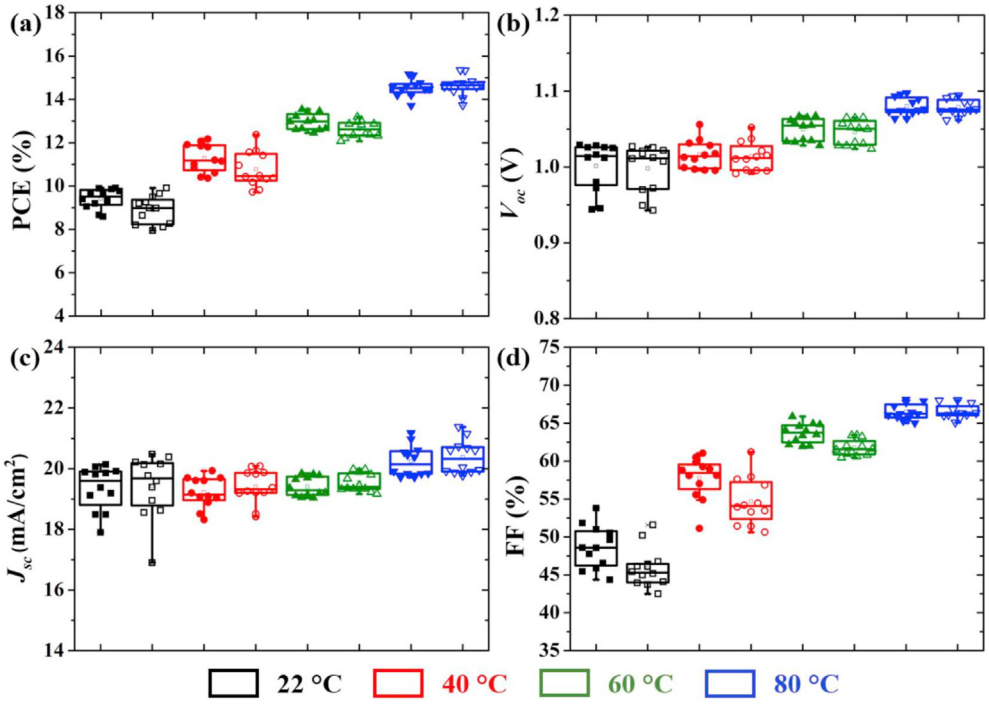

Fig. 8. Statistical photovoltaic parameters of prepared samples at various temperatures (Fig. 4 of Ref. [181]).

TiO_2 has some shortcomings such as low electron-mobility (0.1–1.0 $cm^2V^{-1}s^{-1}$), requirement of high sintering temperature (>450 °C), degradation of perovskites under the illumination of light etc. [201]. Hence, to overcome these shortcomings the researchers are focused on to introduce an alternative ETL material. Due to superior electron mobility and low temperature fabrication, the binary metal oxides like SnO_2 [202], ZnO [203], etc., are considered as appropriate replacements to TiO_2. But, ZnO-based photovoltaic devices shows poor stability, owing to OH residue on ZnO which lead to decompose the perovskites [204, 205] So, in recent years SnO_2 has become a star material to serve as ETL in perovskite solar-cells. It is proved that, high quality SnO_2-based device has shown good efficiency which is similar to that of TiO_2-based device. Besides, SnO_2-based devices exhibit superior stability than ZnO and TiO_2-based devices [206,207]. Nowadays, so many researchers are focused on improving the efficacy and stability of the perovskite solar-cells through modifying the structure of the ETL materials.

Generally, the oxidation property of Sn strongly affects the device performance by creating vacancies in cells. To overcome this, Mohammadian-Sarcheshmeh et al. [208] demonstrated the usage of uric acid (UA) as a natural antioxidant in the fabrication of economical and eco-friendly Sn-based perovskite solar cells without HTL. They reported that the addition of UA causes the reduction in oxidation and carrier recombination inturn improves the device performance. This means uric acid prevents the oxidation of Sn and expands the device performance. In addition, for the first time Ghahremani et al. [209] used intense pulsed light for the fabrication of efficient perovskite solar-cells by swift annealing of SnO_2-ETL and triple-cation perovskites. At the time of intense pulsed light annealing, the addition of di-iodomethane alkyl-halide (CH_2I_2) obstruct the regular crystallization and improves the surface-morphology of the perovskite-film by supplying iodine slices through ultraviolet energy. They reported that the SnO_2 based perovskite solar cell fabricated through intense pulsed light annealing exhibit the maximum efficiency of 12.56%. Further, Vijayaraghavan et al. [210] designed highly efficient perovskite solar-cells by introducing SnO_2 quantum-dots as ETL. With the help of low-temperature solution processing technique, they prepared SnO_2-quantum dots. In comparison with the high temperature processed ETLs, the SnO_2-quantum dots exhibit superior electron-extraction and ability of hole-blocking. Besides, the device fabricated using the SnO_2-quantum dots as ETL shows the large efficiency of 13.64%.

In continuing this, Deng et al. [211] demonstrated a new synergetic technique to prepare Sn-doped TiO_2 ETL material for perovskite solar cell applications. For the first time they used hydriodic acid (HI) to passivate TiO_2 film and surface while doping of Sn to TiO_2. Initially, HI controls the hydrolysis of TiO_2 and removes the trap states of related oxygen vacancies. At that moment, incorporation of Sn into HI-passivated TiO_2 forms the TiO_2/SnO_2 films. The addition of SnO_2 suppress the defects throughout the film also enhances the mobility of electrons. Besides they reported that among all the prepared samples 0.05 M SnO_2 doped TiO_2-based perovskite device exhibits low-hysteresis and excellent stability with the efficiency of 17.77%. In addition, even after continuous heating at 100 °C for 21 h, the TiO_2/SnO_2 (0.05 M) device retains 86% of its initial efficiency. Hence, the addition of SnO_2 enhances the stability and efficiency of perovskite solar cells compare to pure TiO_2 based devices. To optimize the energy level alignment, passivate the trapping defects and improve the electron coupling, Zhang et al. [212] introduced the compact and ultrathin SnO_x layers derived from $SnCl_4$ at the interface of ETL and perovskite layer. Moreover, the perovskite solar-cell based on SnO_2 and Cl–SnO_2 as ETL exhibit the PCE of 18.6% whereas without Cl the same device shows the PCE of 16.3%. Furthermore, Huang et al. [213] added the LiCl to SnO_2-ETL by using a simple low temperature process. The addition of LiCl significantly improves the conductivity of SnO_2 which inturn improves the charge transfer process and hinders the recombination of charges. They reported that the perovskite solar cell based on Li:SnO_2-ETL achieves a maximum PCE of 19% whereas under steady state the same device shows the PCE of 18.35%.

In order to upsurge the performance of the SnO_2-based perovskite solar-cells, Du et al. [214] introduced an amino-acid or glycine self-assembled layer onto the SnO_2-ETL at low temperature as a buffer layer. In fact, buffer layer modulates lattice mismatch between SnO_2 and perovskite layer. Besides, electrostatic interactions between the perovskite framework and amino group enhance interaction of SnO_2 and perovskite at the interface. Fig. 9 illustrates the schematic representation of device structure of SnO_2 based perovskite solar cell (Fig. 1A of Ref. [214]). This leads to the reduction in recombination of charge carriers and enhancement in transportation efficiency of charge carriers. They reported that, glycine modified SnO_2-based perovskite solar-cell attained the maximum efficacy of 20.68% along with J_{sc} of 24.15 mA/cm^2, V_{oc} of 1.10 V and FF of 0.78. The better efficiency is evinced that SnO_2/glycine can serve as excellent electron buffer-layer for highly efficient perovskite solar-cells. In addition, compare to binary metal oxides, ternary metal oxides show better properties. Because the relative ratios of the cations can be modified in ternary metal oxide materials, accordingly the optoelectronic properties such as bandgap, electric resistivity can also be regulated. Therefore, the ternary metal oxides such as $BaSnO_3$ [215], $SrTiO_3$ [216], Zn_2SnO_4 (ZSO) [217] etc., are promising ETL-materials for establishing the perovskite solar-cells with high efficiency. Among all the ternary metal oxides, ZSO exhibit superior properties of such as high electron-mobility (10–30 $cm^2V^{-1}s^{-1}$), wide optical-bandgap (3.8 eV), suitable conduction band edge etc. Owing to this ZSO appears to be an attractive electrode candidate in perovskite solar-cells [217]. For the first time, Oh et al. [218] reported the ZSO ETL based perovskite solar cells which exhibit the PCE of 7%. Later on, Shin et al. [219] demonstrated a new method to prepared ZSO nanoparticles for photovoltaic applications. The perovskite solar cells based on prepared ZSO nanoparticles display the PCE of 15.3%. Up next, Jung et al. [220] used the solution-processed ZSO-film as an ETL in perovskite solar cell which shows a champion efficiency of 20.02%. Recently, with the help of simple cost-effective hydrothermal synthesis technique Zheng et al. [221] synthesized Zn_2SnO_4 single crystal. In the proposed technique hydrothermal reaction time regulates the size of the particles and morphology of the ZSO single crystal. Also, the perovskite solar-cell based on ZSO-single crystal exhibits high PCE of 18.32% along with high J_{SC} of 24.79 mAcm^{-2}. Further, the device is stable after 15 days of placing in air with 20% humidity. Hence, all the outcomes are evinced that the ZSO is a promising ETL candidate for the fabrication highly efficient photovoltaic devices.

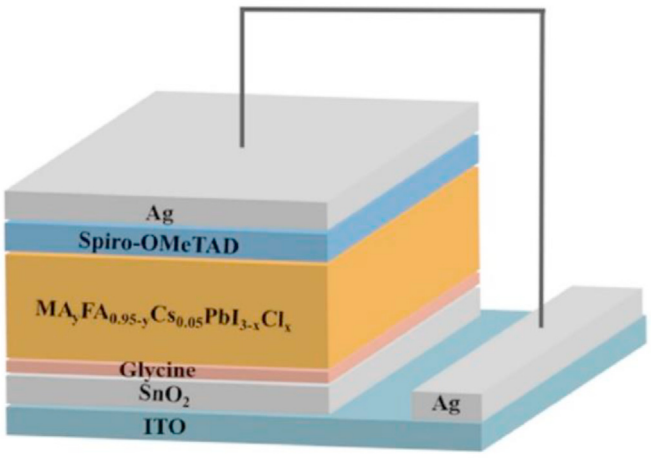

Fig. 9. Schematic illustration of device structure of SnO_2 based perovskite solar-cell (Fig. 1A of Ref. [214]).

Recently, Sadegh et al. [222] employed the ZSO as an ETL in perovskite solar cells. They reported that, the surface of the ZSO-layer is modified with the help of chemical bath deposition (CBD) technique. CBD alters the surface morphology and density of the perovskite-film. Hence, the CBD results a perovskite-film with good surface-coverage & elaborated grains. The recombination of charge carriers causes the reduction in losses. These improvements effectively suppress the trap-assisted recombination and enhance charge-extraction at ETL/perovskite interface. Accordingly, the same time the photovoltaic performance is also increases. Finally, the CBD-modified ZSO-ETL-based perovskite solar-cells exhibit the maximum PCE of 21.3%. Specifically, the prepared device shows the excellent stability i.e., it exhibits 90% retention of its initial efficiency even after 1000 h continuous illumination. Fig. 10 illustrates the J-V characteristics and stability of the prepared CBD-modified ZSO ETL-based perovskite solar cell (Graphical abstract of Ref. [222]). The research is still going on to introduce new kinds of Sn-based materials to improve the performance of the perovskite solar cells.

5. Polymer-based PSCs

Recent years, the researchers are focused on the enhancement of the stability of perovskite solar-cells through device encapsulation [223], interfacial modification [224], and inverted configuration [225], so on. It is reported that, stability of perovskite can also be enhanced by intrinsic modification, especially through mitigation of moisture-corrosion [226,227]. Generally, the interfacial-modification is directed among the functional layer such as [228,229] ETL or HTL or photovoltaic-layer and buffering layer. Nevertheless, the density of defects is more at perovskite-perovskite interface also called intergranular interface [230], owing to which the moisture can easily spreads into the perovskite-film [231–233]. Hence, it is necessary to improve the crystallinity at intergranular interface, which in turn improve the morphology along with charge transportation and separation of the perovskite film [234,235]. Synergistically, the improved charge separation and transportation of the perovskite film lead to accomplish excellent photovoltaic performance. Therefore, the perovskite solar cells with moisture resistant perovskite intergranular interface exhibit the improved stability [233]. In order to get moisture resistant intergranular interface researchers are focused on the enhancement of interactions among the perovskite grains such as π-π stacking, hydrogen bonding, acid base, electrostatic interactions etc [230]. For instance, the π-π stacking and hydrophobic interactions can be increased by replacing the methylamine (CH_3NH_2) with phenethylamine ($PhCH_2CH_2NH_2$). Besides, substitution of methylamine (CH_3NH_2) with formamidine causes the improvement in the hydrogen bonding interaction. As a result, the perovskite solar-cells exhibit better-quality PCE and stability. Owing to existence of multitudinous active sites [236,237], polymers such as polymethylmethacrylate (PMMA) [238], polyethylene oxide (PEO) [239], polyvinylpyrrolidone (PVP) [240], polyethylenimine (PEI) [241] etc., exhibit the strong intergranular interactions. Due to existence of strong intergranular interactions, the polymer based photovoltaic devices exhibit excellent stability and better PCE [242,243]. Till now, all reported polymers modulate the intergranular-interactions of the perovskite through adjusting their curly macromolecular-configuration. Or else, they will play a detrimental role such as decreasing the crystallinity of perovskite, weakening the photoelectric-property etc. To overcome these difficulties, the researchers are focused on to introduce a new kind of polymers with improved configuration. In this view, dendrimers or dendritic polymers with 3D spherical structure gained much attention. Because, while interacting with the perovskite-grain

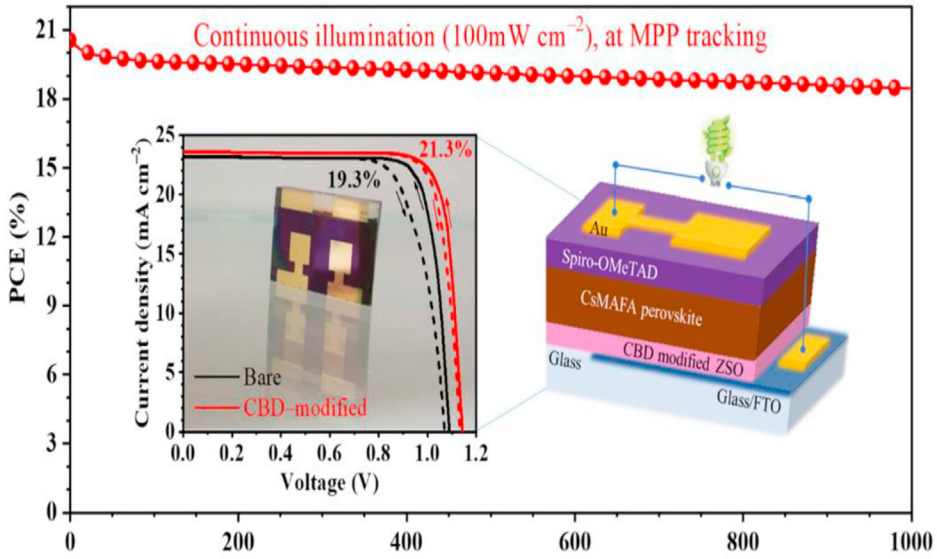

Fig. 10. J-V characteristics of the prepared CBD-modified ZSO ETL based perovskite solar cell (Graphical abstract of Ref. [222]).

surface, molecular structure affords slight adjustment in configuration, which also leads to prevent local accumulation inside the distinct linear-macromolecular configuration [244,245]. This improves the crystallinity of the dendrimers. So, the usage of dendritic polymers can effectively improve the stability and efficacy of perovskite devices.

In continuing this, Du et al. [246] designed a new molecular guideline for enlightening the effectiveness of perovskite solar-cells. The proposed model effectively regulates the intergranular perovskite interface which in turn improves the PCE. Du et al. [246] used polyamidoamine (PAMAM) dendrimers as dendritic crystallization framework to template perovskite crystallizing procedure. Actually, at molecular periphery the PAMAM consists methyl esters, whose amino & carbonyl-groups have great potential to interact with grain surface of the perovskites. Owing to this the perovskite grains cross linked with dendritic-polymer backbone which exhibit strong intergranular interfacial interactions.

In addition, by removing the pin holes and suppressing the grain boundaries of perovskite, considerably improves the morphology of the perovskite phase. Hence, the dendritic PAMAM crosslinking the perovskite grain strengthens the interactions at perovskite intergranular interface also provides a uniform, non-pinhole and compact perovskite-film. Owing to this, at ambient condition the unencapsulated perovskite solar cells utilizing the PAMAM dendritic-polymer backbone exhibits large PCE of 42.6%. In addition, after 400 h the PAMAM-modified device shows 73% retention of its initial PCE. The PAMAM modification leads to improvement of perovskite intergranular-interactions which are main reason for accomplishing high PCE. Fig. 11 illustrates the schematic representaion of regulating perovskite morphology by PAMAM dendrimers along with device configuration of PAMAM modified perovskite solar cell (Fig. 2 of Ref. [246]). Furthermore, interlayers also play a pivotal role in elevating the PCE of perovskite solar-cells. Recently, Kang et al. [247] demonstrated the effect of polyelectrolytes on the device performance when they are used as buffer layers in both p-type substrate (P–I–N) and n-type substrate (*N*–I–P) geometries. They used nonconjugated polymer electrolytes (NPEs) based on poly(ethyleneimine) (PEI) backbone with various counterions such as iodide (I^-), bromide (Br^-), and tetrakis (imidazole) borate (BIm_4^-) to prepare buffer layers. Besides, the performance of the perovskite solar-cell is varying with respect to the size of counterion. Actually, the non-conjugated polymers generate the electric dipoles at the interface of the NPE/metal electrode which can tune the work functions and the energy levels of the electrodes. Owing to this the NPE buffer layer based photovoltaic device exhibits the PCE of 14.71% and 13.79% in *N*–I–P and P–I–N geometries respectively.

In general, HTL-layer extracts the holes and prevents the recombination of charge carriers at the perovskite and electrode interface which in turn influence the performance of perovskite solar-cell. Hence, advancement of hole transport layers is also essential for designing perovskite solar cells with high performance [248]. Till now, poly(ethylenedioxythiophene) (PEDOT): poly(-styrenesulfonate) (PSS) is one of the widely used HTL in inverted perovskite solar-cells, because of their excellent film morphology, high conductivity and solution processing at low temperatures [249,250]. Despite of the advantages, acidic-nature of PEDOT:PSS is one of the major limitation of PEDOT:PSS based HTL in perovskite solar cells [251]. Many researchers are searching for a new way for enhancing the device performance by overcoming the shortcomings of PEDOT:PSS HTLs [252–254]. Recently, Xu et al. [255] proposed a novel method for fabricating new HTL by utilizing broadly available copper (I) thiocyanate (CuSCN). They reported that, the addition of CuSCN to PEDOT:PSS followed by low temperature annealing causes improvement in the efficiency of the charge extraction and reduction in energy barrier along with acidic nature. Owing to this the perovskite solar cell based on CuSCN modified PEDOT:PSS-HTL exhibits the PCE of 15.3% at V_{OC} of 1.0 V, which is 16% better than that of PEDOT:PSS-HTL based perovskite solar-cells. Besides, lower acidity results outstanding long-term stability i.e., the device retains 71% of its initial PCE after the exposure of N_2 for 175 h under full sun illumination. For further enhancement of PCE, Ma et al. [256] introduced a small triphenylamine-based molecule called *N*,*N*′-(Bis-(1-naphthaleny)-*N*,*N*′-bis-phenyl-(1,1′-biphenyl)-4,4′-diamine(NPB) as a multifunctional buffer layer into the perovskite solar cell. Fig. 12 represents the device structure of the NPB-based perovskite solar-cell (Fig. 1 of the Ref. [256]). They reported that NPB as a buffer layer reduces the defects and pinholes of perovskite film, also alter the energy level mismatch between PEDOT:PSS layer and perovskite structure. The recombination of the charge carriers are strongly restricted in NPB-modified device, owing to the reduced pinholes and defects at the interface of the

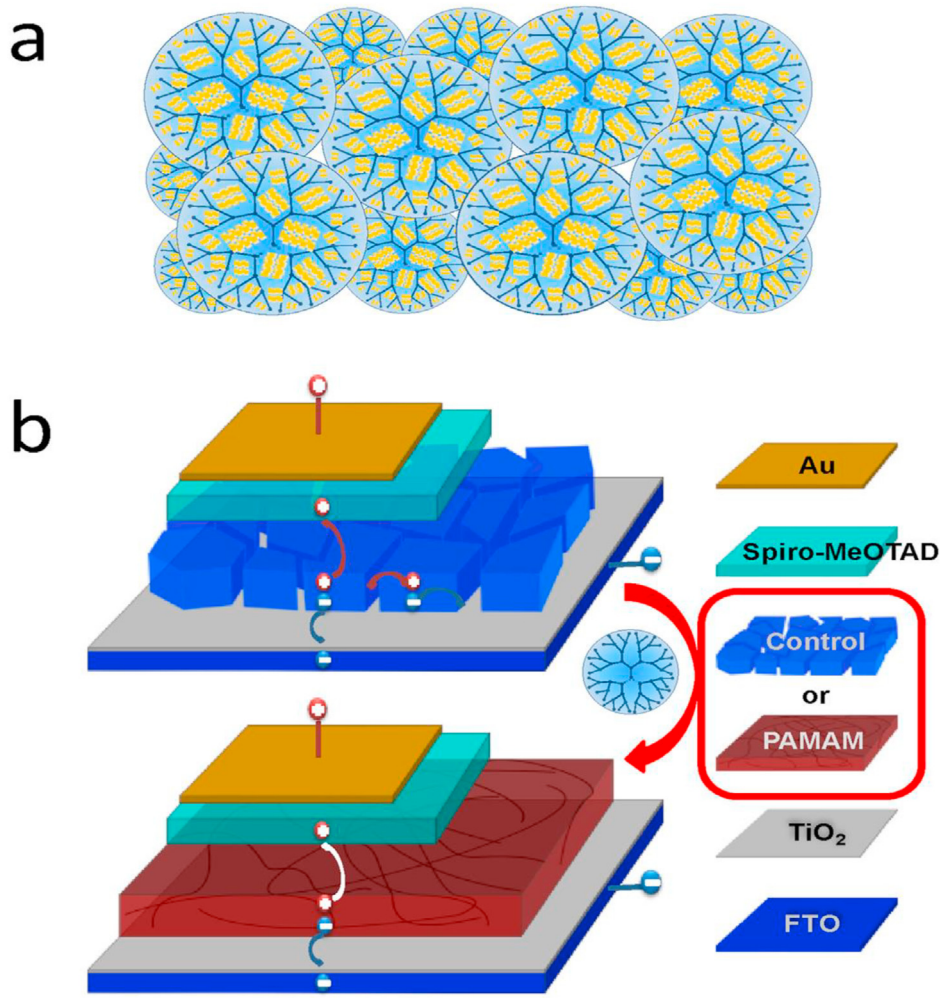

Fig. 11. a) Schematic representaion of regulating perovskite morphology by PAMAM dendrimers b) Device configuration of control and PAMAM-modified perovskite solar cells (Fig. 2 of Ref. [246]).

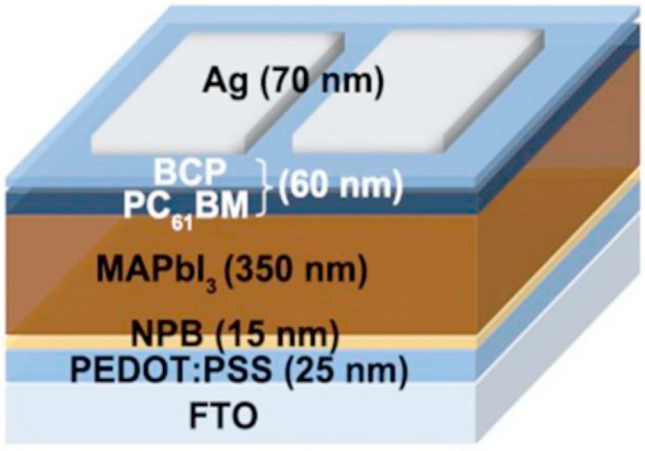

Fig. 12. Device structure of the NPB-based perovskite solar-cell (Fig. 1 of the Ref. [256]).

perovskite and PEDOT:PSS layer. As a result, the NPB modified perovskite solar cell exhibit the PCE of 18.4%. The same device shows the PCE of 14.4% without hysteresis and excellent stability under ultraviolet radiation. The proposed approach suggests that the NPB as a buffer layer may play a key role in designing next generation highly stable, efficient and flexible perovskite solar-cells.

So far, almost all the reported flexible perovskite solar cells have small areas. It is well-established that the PCE is condensed when the area of the devices is amplified to large-scale, owing to the inevitable loss of homogeneity in the films. Accordingly, the deposition procedure for the thin-films at large scale directly influences the performance of the large-area flexible perovskite solar cells. Hence, large area techniques need to be developed for the fabrication of all the layers in flexible perovskite solar cells. So, the alternative technology should be introduced to further decrease the fabrication cost, which hopefully promotes the realization of practical applications.

6. Conclusion

Nowadays, the performance of perovskite solar cells has made great progress. Many fabrication techniques and new perovskite compounds have been established to design high performance solar-cells. In recent years great number of materials such as H_2O/ETA/EDA/DTA, SZTO, MZO, $Cu_2O/MAPbI_3/SiO_2$, TiO_2@$PbTiO_3$, $CsPbBr_3$, 2D/3D Pb–Sn alloys, TiO_2/SnO_2, Li:SnO_2, CBD-modified

ZSO, PAMAM, PEDOT:PSS, etc., have been designed which are utilized as ETL, HTL and buffer layers in perovskite solar cells to accomplish high PCE and stability. Although massive research is going on in the advancement of perovskite solar-cells, significant challenges remain. However, to develop perovskite solar cells for practical applications, numerous scientific challenges and issues must be overcome, such as diminishing charge separation, transportation, and collection losses. Amongst, charge carrier injecting/collecting properties are essential for enlightening the performance, which are intensely associated to the interfacial properties between the electrodes and photoactive layers. It is well known that the properties of interface materials, such as the morphology and energy level alignment, have important effects on the charge transportation in photovoltaic devices. Researchers are putting their continuous efforts to overcome the challenges and also towards the development of next generation perovskite solar-cells with enhanced power conversion efficiency and long-term stability.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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