RESEARCH PAPER

SnO$_2$ Layer Treatment with Tthioacetamide in Perovskite Solar Cell

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ABSTRACT

The electron transport layer plays a pivotal role in shaping the photovoltaic attributes of perovskite solar cells. SnO$_2$ stands out as an exemplary electron transport layer for perovskite solar cells due to its exceptional carrier mobility, deep conduction band, suitable band gap, and compatibility with low-temperature processing. Although surface modification of SnO$_2$ has yielded noteworthy enhancements in device performance over recent years, there remains considerable untapped potential to further refine its efficiency and long-term stability. In this study, thioacetamide was employed to modify the SnO$_2$ surface, aiming to elevate the quality of the electron transport layer and establish a robust interface with perovskite. The findings underscored that the thioacetamide-modified SnO$_2$ layer exhibited augmented perovskite absorption in the visible spectrum compared to the control sample. Additionally, the attenuation in photoluminescence intensity within the modified sample alludes to improved electron extraction and enhanced charge transport from the perovskite layer to the electron transport layer. Assessment of solar cell performance unveiled superior and more consistent photovoltaic parameters in the modified sample. Ultimately, the best efficiency was achieved with the perovskite solar cell using SnO$_2$ modified with thioacetamide, boasting an efficiency of 15.15%.

INTRODUCTION

Perovskite solar cells (PSCs) have emerged as a promising photovoltaic (PV) technology, thanks to their enhanced efficiencies and remarkably low production costs. Recent advancements in perovskite solar cell development underscore its strong potential for commercialization. The performance of perovskite solar cells is profoundly influenced by factors such as device architecture, perovskite active materials, charge transport layers, and interfaces—areas that have garnered extensive research attention. Typical perovskite solar cell configurations follow an n-i-p structure, encompassing a transparent conductive oxide electrode, an electron transport layer (ETL), an absorber layer, a hole transport layer, and a metal-based cathode [1–3]. Among these layers, ETLs play a pivotal role in enhancing perovskite solar cell performance.

The electron transport layer within perovskite solar cell structures assumes a critical function in facilitating electron extraction and collection, significantly impacting overall device efficiency. This layer promotes charge transfer and minimizes carrier recombination at the interface between ETL and perovskite. It can directly influence key photovoltaic parameters like short-circuit current density, open-circuit voltage, and fill factor [4,5]. A variety of materials, such as TiO$_2$, SnO$_2$, ZnO,

and organic semiconductors have been widely employed as electron transport layers in perovskite solar cells. However, when using TiO_2 or ZnO-based perovskite solar cells, device performance typically deteriorates to 10-30% of the initial value after just a few hours of storage, irrespective of the storage conditions. This instability is attributed to TiO_2's optical instability and the perovskite layer decomposition caused by residual -OH groups on ZnO surfaces. In contrast, SnO_2, an n-type semiconductor with a wide band gap, stands out as an excellent electron transport material for perovskite solar cells due to its high carrier mobility, low optical loss, chemical stability, insensitivity to UV light, and suitability for low-temperature processing [6–9].

Efforts to enhance electron transport layers frequently involve modifications that aim to improve electron injection and hinder electron recombination. Approaches include elemental doping, integration with other electron transport materials, and surface modifications. Modifying the SnO_2 layer holds the potential to considerably enhance perovskite solar cell performance. This includes altering energy levels to enhance electron injection and hole blocking, passivating surface defects to suppress recombination, enhancing the absorber layer's morphology and interface, and bolstering device long-term stability. Notably, during the solution-based processing of SnO_2 nanoparticles, defects, such as oxygen vacancies, often arise. The abundance of these defects, which varies depending on the manufacturing method, adversely affects the electronic properties of the electron-transporting layer. This results in suboptimal charge transfer and significant charge accumulation at the interface between the electron transport layer and perovskite. Furthermore, due to the lattice mismatch between the electron transport layer and perovskite, dangling bonds may form at the interface. These mismatched sites can introduce deep-level defect states within the energy band-gap, exacerbating excess charge recombination. This unfavorable interface not only degrades perovskite solar cell performance but also contributes to hysteresis behavior [9–11].

Surface defects stemming from the problematic interface between the electron transport layer and perovskite can also constrain the long-term performance of perovskite solar cells. As a countermeasure, surface defects must be suppressed through effective surface passivation. Doping the electron transport layer with metal cations presents an effective strategy to enhance its properties and the interface with perovskite. By introducing metal oxide impurities, thin films can be covered adequately, and energy levels can be shifted to achieve proper alignment with adjacent layers. This enhances thin film conductivity and overall device performance. In the case of doping the SnO_2 electron transport layer, elements such as Al^{3+}, Y^{3+}, Li^+, Nb^{5+}, Mg^{2+}, Ga^{3+}, and Sb^{5+} have been employed [12]. Among these, Li^+ has proven highly effective in enhancing the electronic characteristics of the SnO_2 ETL [13,14]. For instance, Park et al. achieved Li-doping by mixing $SnCl_2 \cdot 2H_2O$ with lithium bis(trifluoromethane)sulfonimide (Li-TFSI) salt. The resultant Li-doped SnO_2 exhibited increased conductivity and a downward shift in the conduction band, facilitating electron injection and transfer from perovskite to SnO_2 [14]. Furthermore, researchers like Guo, Xing, et al. harnessed SnO_2:$InCl_3$ ETL in planar perovskite solar cells. The simultaneous doping of ETL and passivation of defects at the ETL/perovskite interface led to substantial performance enhancements attributed to SnO_2 modification with $InCl_3$. This modification included lower trap density, larger grain size, and enhanced ETL electron mobility [15].

In recent times, a novel approach involves the synergistic combination of the electron transport layer with surface modification to enhance the performance of perovskite solar cells. This integration of the electron transport layer and surface modification serves multiple purposes, such as controlling energy level deviations at interfaces, mitigating surface losses arising from recombination, refining the morphology of the active absorbent layer, optimizing the work function of contact layers, refining interface contact quality, and bolstering the long-term stability of the devices. Presently, a range of techniques is employed for incorporating or modifying the SnO_2 surface in perovskite solar cells. These include the utilization of inorganic metal oxides (such as TiO_2, MgO, Sb_2O_3, ZnO) as the secondary layer in conjunction with the SnO_2 ETL, along with the incorporation of organic materials like self-assembled monolayers, polymers, carbon-based materials, and ionic salts (e.g., KCl, KF, RbF, NH_4Cl, and NH_4F) [6,12].

In recent years, substantial attention has been directed toward the modification of the SnO_2 electron transport layer through the incorporation of carbon-based materials. These materials are primarily utilized to enhance the crystallinity and morphology of perovskite films, thus facilitating

an efficient pathway for charge collection [9]. An example of this approach is demonstrated by Pang et al., who combined graphene quantum dots (GQDs) with SnO_2 to enhance the performance of perovskite solar cells. Their findings indicated that the incorporation of GQDs led to improvements in electron extraction and transfer, attributed to a better alignment of energy levels within the devices. This enhancement subsequently resulted in improved short-circuit current density (JSC) and open-circuit voltage (VOC), leading to better stability and performance of the perovskite solar cell [16].

In a pioneering study, Cao et al. introduced a fullerene derivative known as pyrrolidinofullerene C60-substituted phenol (NPC60-OH) for SnO_2 ETL modification. This resulted in the enlargement of grain size, thereby promoting electron transfer and reducing recombination velocity. Additionally, it contributed to the reduction in the density of trap states within the perovskite film [17].

Ionic salts have proven effective in passivating charged surface defects, including hydroxyl groups, oxygen vacancies, exposed Sn atoms, and dangling bonds. This is achieved through hydrogen bonding and electrostatic interactions [12]. Jung et al. harnessed ammonium fluoride (NH_4F) as a bifunctional surface passivator, which effectively lowered defect density and adjusted the Fermi level of SnO_2. The inclusion of ammonium ions (NH_4^+) served to suppress surface hydroxyl groups, while fluorine ions (F^-) substitution at defect sites resulted in diminished energy losses and improved efficiency of perovskite solar cells [18].

In the scope of this study, thioacetamide emerged as a key modifier for the SnO_2 layer and its interface with perovskite. Thioacetamide (CH_3CSNH_2), abbreviated as TAA, represents a low molecular weight compound containing hydrogen (H) atoms and strongly electronegative atoms (N and S) with lone electron pairs. Notably, thioacetamide releases hydrogen sulfide during hydrolysis reactions, thus proving to be a useful substitute for hydrogen sulfide (H_2S) in both quantitative and qualitative analyses [19].

MATERİALS AND METHODS
Materials
N,N-dimethylformamide (DMF,99.8%), dimethylsulfoxide (DMSO,99.9%), chlorobenzene (99.9%), ethanol (99.99%), and Thioacetamide (99.9%) were supplied by the Merck Co. Tin chloride pentahydrate ($SnCl_4 \cdot 5H_2O$,98%), CsI

(99%) were provided by Sigma Aldrich Co. In addition, lead (II) iodide (PbI_2, IRASOL), lead (II) bromide ($PbBr_2$, IRASOL), formamidinium iodide (FAI, Great Cell), and methyiammonium bromide (MABr, IRASOL) were used. Also, $CuInS_2$(CIS) ink and carbon paste (IRASOL) were utilized.

Device fabrication
The pre-patterned ITO (14mm × 14mm) were cleaned with detergent, deionized water, ethanol, 2-propanol and deionized water in sequence. Then the substrates were treated 30 min with UV-Ozone (UVO) before depositing SnO_2 ETLs. An SnO_2 precursor solution with concentration 0.1 M was prepared by dissolving 0.035 g of $SnCl_4 \cdot 5H_2O$ into 1 mL ethanol. The SnO_2 precursor solution was spin-coated on ITO substrate at 4000 rpm for 30 s and then annealed at 180 °C for 30 min at ambient condition. The Thioacetamide precursor solution was obtained by dissolving 0.018 g in 1 ml deionized water (0.08 M). Thioacetamide solution was spin-coated on UVO-treated SnO_2 films at 6000 rpm for 30 s and then heated at 180 °C for 30 min. The triple cation perovskite with a formula of $Cs_{0.05}$ $(MA_{0.17}FA_{0.83})_{0.95}Pb$ $(I_{0.83}$ $Br_{0.17})_3$ were deposited on ETL layers as follows. Precursor solution containing FAI (1 mmol), PbI_2 (1.5 mmol), $PbBr_2$ (1.5 mmol) and MABr (1 mmol) in anhydrous N,N-Dimethylformamide (DMF) and dimethyl sulfoxide (DMSO) mixed solvents with a volume ratio of 4:1 was prepared. Then, 50.6 µL of 1.5 mmol CsI solution dissolved in DMSO was added to the mixed perovskite precursor solution. The precursor solution was spin-coated at 1000 rpm (10 s) and then 6000 rpm (20 s). 270 µL anti-solvent of chlorobenzene was injected on the spinning substrates in the last 8 s of the procedure. The samples were annealed at 100 °C for 60 min. As the hole transport layer (HTL), $CuInS_2$ ink was deposited by spin coating at 3000 rpm for 30 s and heated at 100 °C for 10 min on the PSK layer. This procedure was repeated again for increasing the thickness of the $CuInS_2$ layer. Finally, for the back electrode, the carbon paste was deposited on the HTL layer using the doctor blade method. The prepared samples were heated in the oven at 100 °C for 30 min.

Characterization
FE-SEM images were taken using MIRA3 TESCAN field emission scanning electron microscope. The crystallinity of perovskite films was obtained by grazing incidence X-ray diffraction
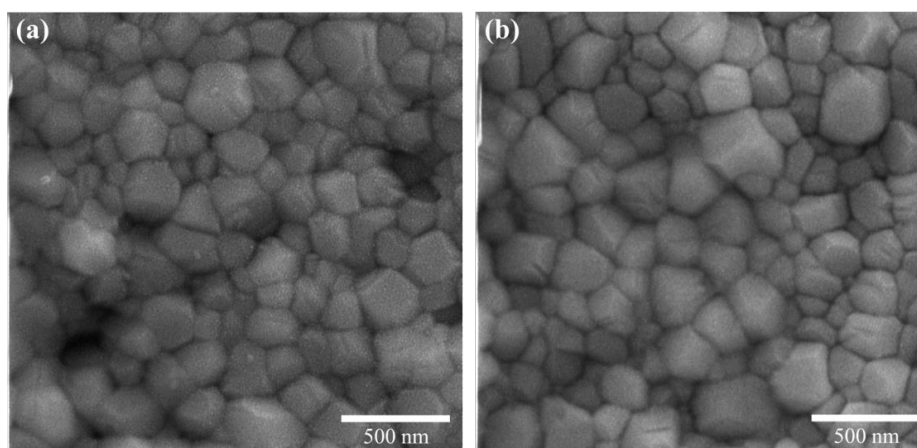
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Fig. 1. Top-view SEM images of the perovskite films spin-coated on (a) SnO$_2$ and (b) the SnO$_2$/TAA layer.

(GIXRD) patterns with a Cu Kα radiation source (λ = 1.54056 Å) at a grazing incidence angle (θ) of 2⁰. The UV-vis spectra of ETL/perovskite films were measured by a Perkin Elmer Lambda 25 UV/VIS spectrophotometer. Photoluminescence (PL) spectroscopy was carried out using Avantes TEC-2048 spectrometer. Incident photon-to-current efficiency (IPCE) was taken in a wavelength range from 370 to 940 nm by IRASOL IPCE-020 equipment. Photovoltaic parameters measurement was conducted with Palmsens potentiostat and a solar simulator (IRASOL, SIM-1000) at 100 mW.cm^{-2} illumination AM 1.5 G, calibrated with a standard silicon solar cell. Electrochemical impedance spectroscopy was acquired using a potentiostat galvanostat-PGE18 (IRASOL).

RESULTS AND DİSCUSSİON

To study the impact of SnO$_2$/TAA on the surface morphology of perovskite films, the top-view field-emission scanning electron microscopy (FE-SEM) measurement was performed. Fig. 1 a,b shows the top-view SEM images of the perovskite films spin-coated on the pristine SnO$_2$ and SnO$_2$/TAA layers, respectively. In Fig. 1a, the SnO$_2$/perovskite film showed non-uniform film with pinholes. Nevertheless, the perovskite deposited on SnO$_2$/TAA (Fig. 1b) exhibited a continuous film without obvious pinholes and slightly larger grain size than that of the SnO$_2$ ETL, which indicated an enhanced quality of the perovskite film and agreed with the enhanced device performance.

X-ray diffraction (XRD) analysis was performed to investigate the crystal structure of perovskite layer (Fig. 2). For the pristine perovskite film, characteristic peaks can be observed that are

associated with the defined crystal planes in triple cation perovskite [20]. The position of characteristic peaks of perovskite did not change significantly in the presence of SnO$_2$ layer, which indicates that it will not affect the structure of perovskite. However, the intensity of the peak at 14.2 degrees increased in both the SnO$_2$ and SnO$_2$/TAA, which was possibly caused by the formation of perovskite with better crystallinity or increased thickness that is consistent with SEM results.

In order to better investigate the improvement of light absorption and transmission of the perovskite layer in the presence of the SnO$_2$ layer, the Ultraviolet–visible (UV–Vis) measurements of the perovskite layer without the SnO$_2$ layer and with the unmodified and modified SnO$_2$ layer were compared with each other (Fig. 3).

The perovskite layer in the presence of the modified SnO$_2$ layer with thioacetamide had a higher absorption and less light transmission in the visible region, which shows that the interface of SnO$_2$ layer with perovskite has improved; however, all the films demonstrated identical absorption edges, which indicates that the bulk structure and energy bandgap of perovskite has not changed. Thioacetamide can modify the surface of the layer and its interface with perovskite by forming a new bond with surface dangling bonds that are related to surface defects and act as recombination centers, and cause more light absorption.

Fig. 4 represents the photoluminescence (PL) spectra of the perovskite film with and without ETL. The quenching in the PL intensity of the perovskite layer in the presence of the SnO$_2$ layer indicates the improvement of electron charge extraction from the perovskite, and the surface modification of the
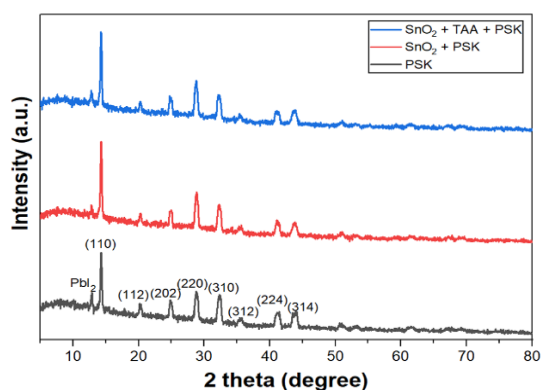
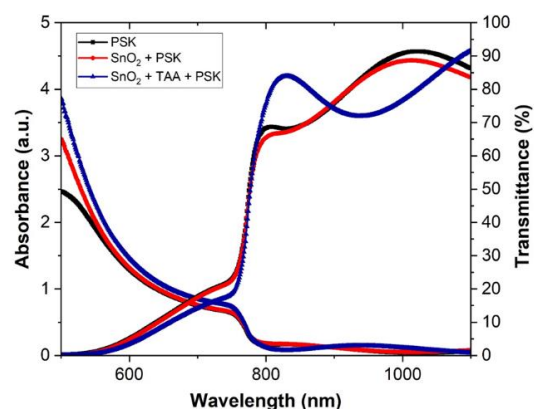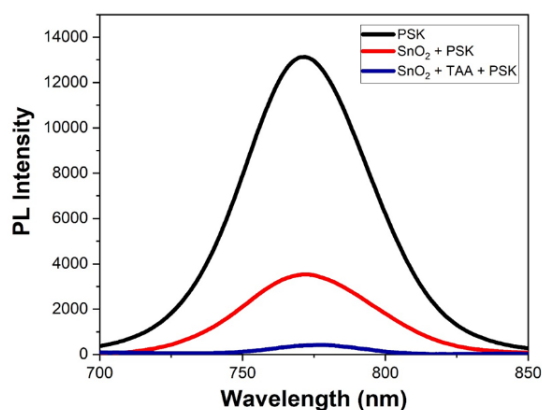Fig. 2. X-ray diffraction pattern.

Fig. 3. UV-Visible spectra of perovskite, SnO$_2$/perovskite and SnO$_2$/TAA/perovskite.

Fig. 4. PL spectra of the perovskite film with and without ETL.

SnO$_2$ layer by thioacetamide improved this process.

Fig. 5 illustrates the current-voltage curve and external quantum efficiency of the best perovskite solar cell based on SnO$_2$ and SnO$_2$/TAA ETLs. Table 1 summarizes the photovoltaic parameters of the best cell based on control and modified sample by thioacetamide. The best efficiency obtained in the modified sample is 15.15%. The surface modification of the SnO$_2$ layer has improved the photovoltaic performance of the cell. Additionally, the current values obtained from the current-voltage curve are consistent with the integral of the current density obtained from the external quantum efficiency curve. Enhancement of external quantum efficiency in the modified sample indicates the reduction of recombination at the SnO$_2$/perovskite interface, which also improved the current. The sulfur and nitrogen atoms in thioacetamide can react and bond with the dangling bonds of the SnO$_2$ layer and modify the surface of the SnO$_2$ layer. Moreover, sulfur

atoms can bond with uncoordinated lead atoms of perovskite and improve the interface of SnO$_2$ layer with perovskite. Liu et al. investigated the effect of thioacetamide doped in TiO$_2$ and spin-coated on TiO$_2$ layer on perovskite properties. The nitrogen atom (N) can coordinate with the titanium atom (Ti) in the electron transfer layer and the sulfur atom (S) can coordinate with the lead atom in the perovskite layer, which effectively reduces the defects and passivates the film surface [21].

Electrochemical impedance spectroscopy (EIS) measurements were performed to obtain more information about charge transfer properties and recombination phenomena. Electrochemical impedance measurements were conducted with alternating current voltage of 20 mV in the frequency range between 1 MHz and 1 Hz, under AM1.5G illumination and at forward bias voltages of 0.8, 0.6, 0.4, 0.2 and 1.1V. Fig. 6 shows the Nyquist plot of perovskite solar cells based on SnO$_2$ and SnO$_2$/TAA electron transport layer along with the

Table 1. Photovoltaic parameters of the best cell based on control and modified sample by thioacetamide.

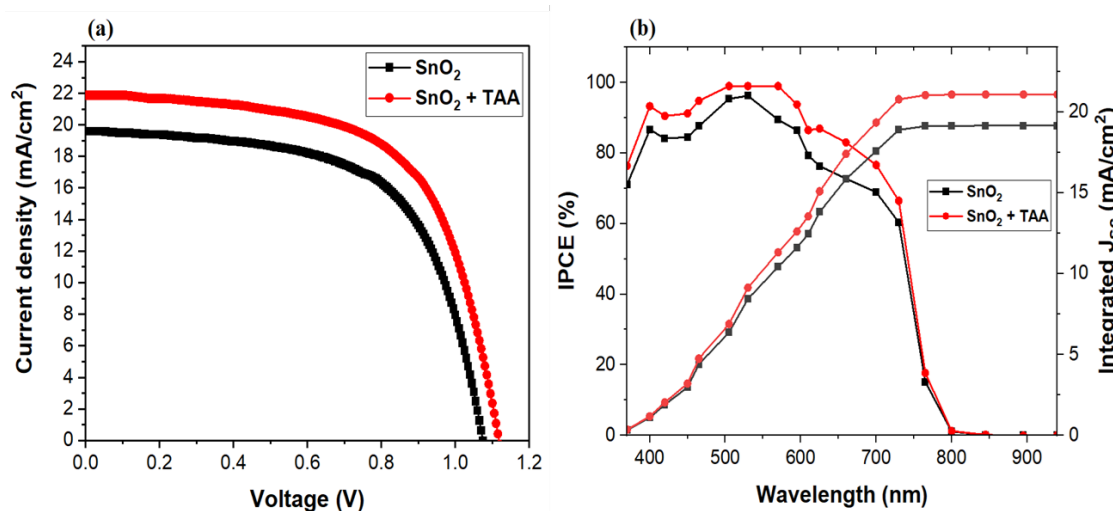| Groups	J_{SC} (mA/cm²)	V_{OC} (V)	PCE (%)	FF
SnO₂	19.58	1.07	13.06	0.62
SnO₂/TAA	21.86	1.11	15.15	0.62

Fig. 5. (a) Current-Voltage curve and (b) External quantum efficiency of the best perovskite solar cell based on SnO₂ and SnO₂/TAA ETLs.
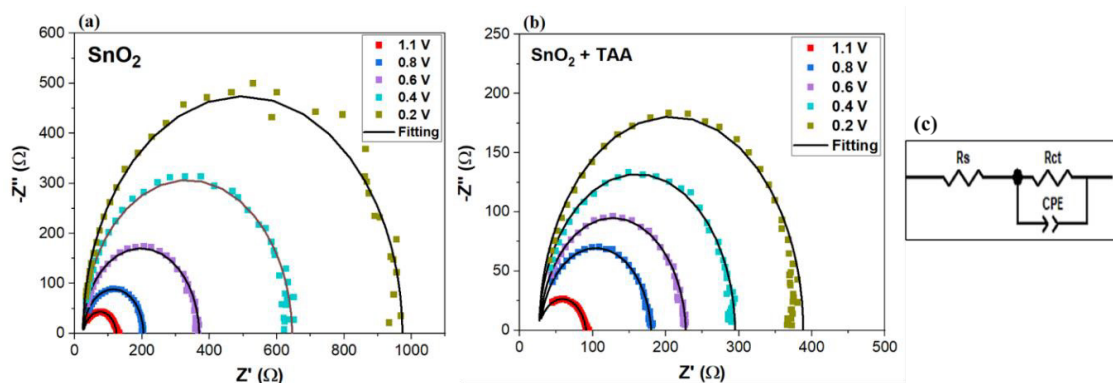

Fig. 6. Nyquist plot of perovskite solar cells based on electron transfer layer (a) SnO₂, (b) SnO₂/TAA and (c) equivalent circuit model.

equivalent circuit model. Since the interface of the hole-transporting layer with perovskite is the same in both cases, the difference in resistances is related to the interface of the electron-transporting layer with perovskite. As shown in the Fig. 6 a,b, with the increase in voltage, due to the increase in carrier density, the charge transfer resistance decreases, which means an increase in diode conduction. The decrease in charge transfer resistance in SnO₂/TAA sample compared to SnO₂ sample shows the improvement of charge transfer at the interface of the electron transport layer with the perovskite.

CONCLUSİON

Many research works have been reported to improve the electronic properties of SnO₂ to increase the performance of perovskite solar cells, including surface passivation. Due to having strong electronegative atoms, thioacetamide can improve the interface of SnO₂ layer with perovskite through

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the interaction of these atoms with the surface of SnO$_2$ layer and deactivation of surface defects. As the results showed, the modified SnO$_2$ layer has more perovskite absorption in the visible region compared to the control sample. Furthermore, the decrease in the intensity of the photoluminescence spectrum in the modified sample indicates the improvement of electron extraction and charge transfer from the perovskite layer to the electron transport layer. The decrease in charge transfer resistance in the impedance spectrum of the sample modified with thioacetamide compared to the reference sample shows the improvement of charge transfer at the interface of ETL with perovskite. Finally, the study of the performance of the solar cell indicated that modification of the surface SnO$_2$ improved the photovoltaic parameters of the cell compared to the pristine SnO$_2$.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

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