

Interface Engineering of SnO₂ Electron Transport Layers Using Molecular, Polymeric, and Ti₃C₂T_x MXene Modifications for Enhanced Perovskite Optoelectronic Performance

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Abstract

Efficient charge extraction and suppression of interfacial recombination are critical for achieving high-performance perovskite-based optoelectronic devices. In this study, interface engineering of the SnO₂ electron transport layer (ETL) was investigated using complementary molecular, polymeric, and conductive nanomaterial-based modification strategies. Pristine SnO₂ was employed as the reference ETL, while 2PACz self-assembled monolayer (SAM), polyvinylpyrrolidone (PVP), and Ti₃C₂T_x MXene were introduced to modify the SnO₂ surface or bulk electronic transport pathways. The influence of these treatments on the structural, morphological, chemical, optical, and electronic characteristics of the ETL/perovskite interface was systematically investigated using X-ray diffraction, FE-SEM, atomic force microscopy, X-ray photoelectron spectroscopy, steady-state photoluminescence, time-resolved photoluminescence, current–voltage measurements, and external quantum efficiency analysis. The interface modifications were found to influence surface uniformity, chemical environment, defect-related recombination, and charge-transfer characteristics without fundamentally altering the underlying device architecture. Molecular and polymeric modification primarily provided surface and chemical control, whereas Ti₃C₂T_x MXene offered an additional conductive pathway for electron transport within the SnO₂ matrix. The study provides a systematic framework for designing modified SnO₂ interfaces for improved charge extraction and reduced interfacial losses in perovskite optoelectronic devices.

Keywords: SnO₂ electron transport layer; interface engineering; perovskite optoelectronics; 2PACz

1. INTRODUCTION

Metal halide perovskites have emerged as promising materials for next-generation optoelectronic devices such as solar cells and photodetectors, with their rapid progress making them a promising future technology. Because of their relatively good optical absorption, tunable electronic structure, long carrier diffusion lengths and very good charge transport properties, significant improvements have been achieved in device performance in a short time. The performance of perovskite-based devices, however, is sensitive to the intrinsic properties of the perovskite absorber as well as the quality and electronic properties of the interfaces between the perovskite layer and the neighboring charge transport materials. The engineering of the interfaces has thus emerged as a key research field for the betterment of charge extraction, recombination loss and operational stability of

perovskite optoelectronic devices. In one of the early works to demonstrate the significance of interface engineering in perovskite solar cell, Zhou et al. (2014) showed that the optimized control of the interfaces in the device architecture can significantly enhance the PV performance. They demonstrated the critical factors of interfacial contact and charge extraction for realizing high-efficiency perovskite solar cells. Followed by that, the interface design science was gradually taken center stage in the optimization process of perovskite devices, and the research mainly concentrated on the modification of the charge-transport layers, defect passivation, energy-level alignment and the suppression of the interfacial recombination. Fan et al. (2016) summarized the development of Perovskite based solar cells interface design and stressed that it is crucial to design interfaces carefully to promote carrier extraction and mitigate unwanted charge

accumulation and recombination. Ansari et al. (2022)

Yue et al. (2019) explored the interface engineering and the doping of electron transport layers (ETLs) in CsPbI₃-based perovskite solar cells made of ETLs. They found that suitable engineering of the electron transport layer and the interface between it and the perovskite can help enhance the efficiency of charge extraction and aid device operation. These studies have shown that the transport layer cannot be considered as an electronically isolated layer as its surface properties and energy level alignment directly affect the transfer of carriers between the transport layer and the buried interface. Engineering the molecular scale interface has also proved to be a useful tool to manipulate charge transport and reduce interfacial defects. In addition, self-assembled monolayers are very appealing because they can offer controlled chemical reactions and energy-level alignment at the interface, and only very thin interfaces are needed. Self-assembled monolayer interface engineering is also proven to be a high-performance technique to enhance the performance of perovskite photodetection systems, as shown by Liu et al. (2025). The physics of perovskite photodetectors have also been broadly explored in terms of device architecture, carrier generation and transport, recombination and collection processes. Ahmadi et al. (2017) compiled the basic physics and device engineering of organic–inorganic halide perovskite photodetectors and emphasized how control of charge-carrier dynamics and device interfaces are critical to enable desired photodetector properties. The efficiency of separation and transport of the photogenerated carriers plays an important role in parameters like responsivity, specific detectivity, response time, dark current and noise. Albalooshi Almahmood (2025) Hence, to obtain high sensitivity and fast response it is necessary to minimize the interfacial trapping and recombination.

In general, the literature shows that most studies have progressed from traditional optimization of devices to intentional control of the interfaces at the molecular, chemical and electronic levels. Interface engineering is a strategy that can tackle multiple fundamental challenges in PV devices such as interfacial defect mediated recombination, energy level mismatch, inefficient

charge extraction, PV device hysteresis, and PV device instability. Bose & Roy (2025) However, the interplay between the structure of the interfaces, defect chemistry, energy level alignment, charge carrier dynamics, and device performance is still one of the areas of interest. Such a systematical knowledge of these relationships is especially required when using interface engineering for both PV and photodetection devices. The present work is specifically of interface engineering in perovskite based solar cell and perovskite photodetector, with special emphasis on optimizing the electronic charge transport. Investigating the impact of controlled manipulation of the electron- and hole-selective interfaces on defect states, energy level alignment, charge extraction, recombination processes and ultimately on the performance and stability of the resulting devices.

2. Methodology

The present study was undertaken to investigate the influence of interface engineering of the tin oxide (SnO₂) electron transport layer (ETL) on the structural, morphological, chemical, optical, and charge-transport characteristics of perovskite-based optoelectronic devices. The methodology was designed to establish a systematic comparison between pristine SnO₂ and SnO₂ modified through molecular, polymeric, and conductive nanomaterial-based approaches. A conventional SnO₂-based device was initially used as the reference, while interface-engineered devices were fabricated using a self-assembled monolayer (SAM), polyvinylpyrrolidone (PVP), and Ti₃C₂T_x MXene. The different modification routes were selected to address complementary limitations of the SnO₂ surface, including defect-assisted recombination, unfavorable interfacial contact, surface chemical disorder, and limited charge-transfer pathways.

The device fabrication began with transparent conducting substrates, which were cleaned sequentially using suitable solvents and deionized water to remove organic and particulate contaminants. A compact SnO₂ layer was subsequently deposited on the cleaned substrate using a low-temperature solution-processing route and subjected to controlled thermal treatment to obtain a uniform and electrically continuous ETL. For the modified samples, the SnO₂ surface was treated with the selected interface-engineering

material under controlled processing conditions. In the SAM-modified samples, 2PACz was employed as a molecular interfacial layer to alter the surface chemistry and electronic environment of SnO₂. PVP was introduced as a polymeric interfacial modifier, with its concentration controlled to obtain sufficient surface coverage without producing an excessive insulating barrier. Ti₃C₂T_x MXene was incorporated into SnO₂ to form a conductive nanocomposite ETL. The MXene dispersion was prepared under controlled sonication to promote homogeneous distribution of the two-dimensional flakes, followed by blending with the SnO₂ precursor before deposition. The processing parameters and modifier concentrations were optimized to obtain continuous films while avoiding aggregation, excessive surface coverage, or deterioration of optical transparency.

Following ETL preparation, the perovskite absorber layer was deposited under controlled environmental and processing conditions using the selected solution-processing procedure. The deposition and subsequent annealing conditions were kept consistent for the control and modified devices so that the influence of ETL modification could be distinguished from variations in perovskite processing. A Spiro-OMeTAD-based hole transport layer was subsequently deposited over the perovskite film, followed by the deposition of a metallic top electrode under controlled conditions. The resulting device architecture consisted of the transparent conducting substrate/SnO₂-based ETL/perovskite absorber/Spiro-OMeTAD hole transport layer/metal electrode. Pristine SnO₂ devices were fabricated under the same conditions and used as the control for evaluating the effect of each interface-engineering strategy.

The structural characteristics of the prepared ETLs and complete device structures were examined using X-ray diffraction (XRD). The diffraction patterns were recorded for pristine and modified samples to determine the phase composition, crystallinity, and possible changes in structural ordering resulting from the interface treatments. Particular attention was given to the characteristic reflections of the perovskite absorber and the underlying SnO₂ and conducting-oxide layers. Since the SAM and polymeric modifiers were present at very low thicknesses, the absence

of additional crystalline diffraction peaks was not considered evidence of their absence; instead, changes in diffraction intensity, peak width, and relative peak characteristics were considered together with the complementary surface and chemical analyses.

The surface morphology and film architecture were investigated using field-emission scanning electron microscopy (FE-SEM). Surface images of pristine, SAM-modified, PVP-modified, and Ti₃C₂T_x-MXene-modified SnO₂ films were compared to evaluate surface coverage, particle distribution, film continuity, and the formation of morphological defects. Cross-sectional FE-SEM measurements were additionally performed to examine the vertical device architecture and estimate the thickness of the individual functional layers. Particular emphasis was placed on the continuity of the SnO₂/perovskite interface because voids, cracks, pinholes, and delamination can provide localized leakage pathways and promote interfacial recombination. The morphology of the subsequently deposited perovskite layer was also considered to determine whether modification of the SnO₂ surface affected grain formation and film uniformity.

Atomic force microscopy (AFM) was used to obtain quantitative information about the nanoscale surface topography of the pristine and modified SnO₂ layers. The samples were measured under comparable conditions, and the resulting two-dimensional and three-dimensional topographic images were analyzed to determine the arithmetic average roughness (Ra) and root-mean-square roughness (Rq). These parameters were used to assess changes in surface uniformity and height fluctuations produced by the different modification strategies. The AFM results were not interpreted independently because an improvement in interfacial charge transport does not necessarily require the minimum surface roughness. Instead, the topographical characteristics were correlated with the chemical, optical, and electrical measurements to establish the overall role of surface modification.

X-ray photoelectron spectroscopy (XPS) was employed to examine the chemical composition and electronic environment of the SnO₂ surface before and after interface modification. High-resolution Sn 3d and O 1s

spectra were recorded to evaluate changes in the chemical states associated with the SnO₂ surface, including variations related to oxygen-containing species and surface defects. The C 1s and N 1s regions were analyzed for the SAM- and PVP-modified samples, while phosphorus-related features were examined where applicable to verify the presence of the anchoring groups associated with 2PACz. For Ti₃C₂T_x-modified samples, Ti 2p, C 1s, and F 1s spectra were analyzed to confirm the incorporation of MXene and identify its surface termination chemistry. Changes in binding energy, peak position, and relative component contributions were used to evaluate modifications in the local chemical and electronic environment of the ETL surface.

The optical and photophysical properties of the devices were investigated using steady-state photoluminescence (PL) and time-resolved photoluminescence (TRPL) measurements. PL measurements were performed to evaluate changes in radiative and non-radiative recombination following modification of the SnO₂/perovskite interface. The PL response of the control and modified structures was compared to determine whether the interface treatments promoted more efficient carrier extraction or reduced interfacial recombination. TRPL measurements were further used to determine changes in carrier lifetime and interfacial charge-transfer dynamics. The decay profiles were analyzed using suitable lifetime models, and the resulting parameters were compared among the pristine and interface-engineered samples. The combined PL and TRPL results were used to distinguish improvements associated with enhanced charge extraction from those arising from reduced trap-assisted recombination.

The electrical performance of the fabricated devices was evaluated through current–voltage (J–V) measurements under controlled illumination and dark conditions. Key photovoltaic parameters, including short-circuit current density, open-circuit voltage, fill factor, and power conversion efficiency, were extracted from the measured characteristics. The series and shunt resistance behavior was also considered to identify possible improvements in charge transport and suppression of leakage pathways. External quantum efficiency (EQE) measurements were conducted to determine

the wavelength-dependent photoresponse and to assess the contribution of the modified ETL to charge collection across the relevant spectral range. The electrical and optoelectronic results were compared with the structural, morphological, and spectroscopic observations to establish relationships between interface modification and device performance.

Finally, the results obtained from the different characterization techniques were correlated to develop an overall understanding of the role of ETL interface engineering. The pristine SnO₂ device served as the baseline, while the SAM, PVP, and Ti₃C₂T_x-MXene treatments were evaluated in terms of their ability to improve surface uniformity, modify surface chemistry, reduce defect-related losses, facilitate electron extraction, and improve device-level performance. Particular emphasis was placed on identifying the modification strategy that provided the most favorable balance between morphological uniformity, chemical passivation, electrical conductivity, optical compatibility, and interfacial charge transfer. This integrated methodology allowed the influence of ETL modification to be evaluated beyond a single performance parameter and provided a comprehensive basis for understanding the underlying interface-engineering mechanisms.

3. Interface Engineering at the Electron Transport Layer

3.1 Modification of the Electron Transport Layer

The structural and electronic optimization of the electron transport layer (ETL) is vital to mitigating the energetic and chemical losses that occur at the buried bottom interface of planar perovskite optoelectronics (Fan et al., 2016). While colloidal tin(IV) oxide (SnO₂) is widely deployed due to its high optical transparency and low-temperature processability, unengineered (SnO₂) films routinely suffer from high densities of surface hydroxyl groups (-{OH}), surface oxygen vacancies, and microscopic pinholes (Wang et al., 2018). These surface defects introduce deep-level electronic traps that capture photo-generated electrons, leading to severe non-radiative recombination and an energetic barrier that impedes charge extraction toward the transparent

conductive oxide substrate (Luo et al., 2021). To resolve these limitations, this section introduces a dual-functional modification strategy utilizing the integration of two-dimensional ($\text{Ti}_3\text{C}_2\text{Tx}$) MXene

nanocomposites directly into the (SnO_2) matrix, coupled with a surface self-assembled monolayer (SAM) passivation treatment (Palei et al., 2023).

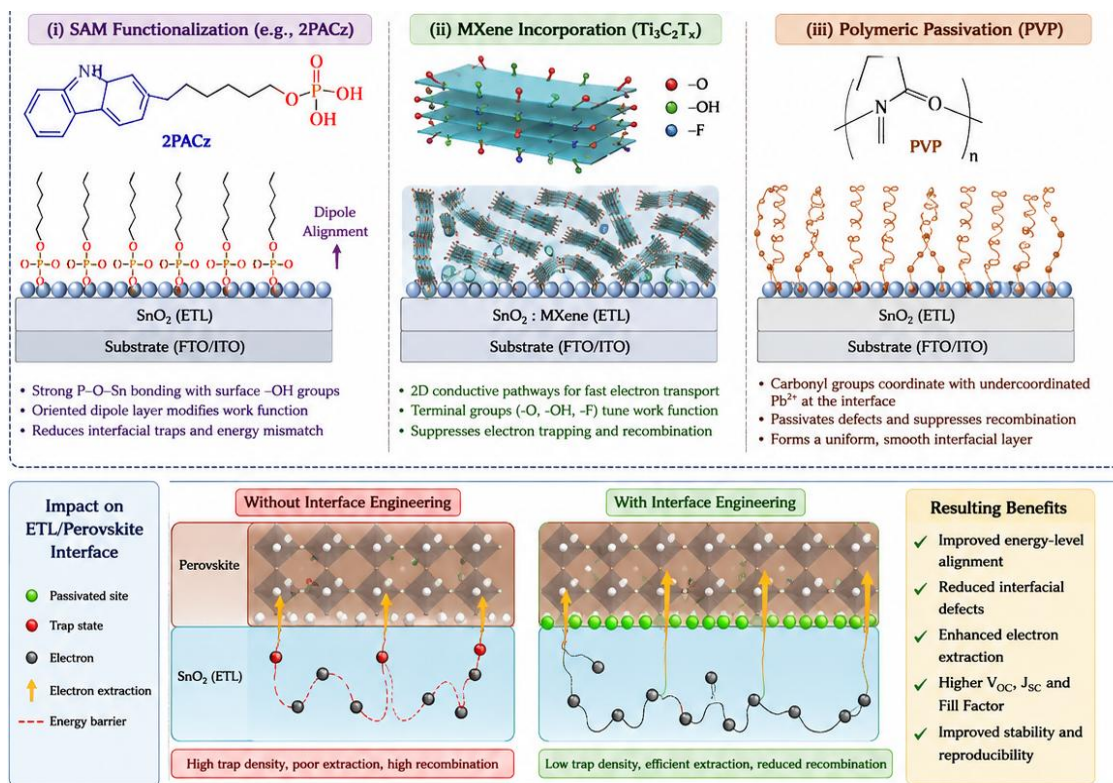


Figure 4.1: Strategies for ETL interface engineering

The incorporation of the highly conductive 2D ($\text{Ti}_3\text{C}_2\text{Tx}$) MXene sheets into the bulk of the colloidal (SnO_2) is executed by blending the materials at optimized volumetric ratios under intense ultrasonic homogenization (Liu et al., 2024). The metallic conductivity of the transition metal carbide sheets introduces an interconnected network of fast electrical pathways within the semiconducting oxide matrix, drastically increasing the bulk electrical conductivity and accelerating vertical electron transport (Palei et al., 2023). This bulk nanocomposite integration effectively lowers the internal series resistance of the ETL, preventing spatial charge accumulation under high photon fluxes and providing a chemically stable foundation for subsequent layers.

3.1 Modification of the Electron Transport Layer

The electron transport layer (ETL) plays a fundamental role in determining the charge-

extraction efficiency and overall performance of perovskite-based solar cells and photodetectors. In the present device architecture, tin oxide (SnO_2) is employed as the principal ETL because of its suitable optical transparency, relatively high electron mobility, favorable energy-level position, low-temperature processability, and compatibility with solution-processed perovskite films. Despite these advantages, the electrical properties of the SnO_2 /perovskite interface can be affected by surface defects, oxygen vacancies, hydroxyl-related species, local energetic disorder, and imperfect interfacial contact. Such imperfections can introduce trap-assisted recombination pathways and hinder efficient electron extraction from the perovskite absorber into the electron transport layer. Therefore, interface modification of SnO_2 was investigated as an effective approach for controlling the electronic and chemical properties of the buried ETL/perovskite junction. The objective was not simply to increase the

conductivity of the ETL, but to simultaneously improve interfacial contact, reduce defect-mediated charge losses, and establish more favorable energy-level alignment for electron extraction. In this study, the modification strategy was investigated

using complementary approaches involving molecular functionalization, conductive nanomaterial incorporation, and polymeric interfacial modification.

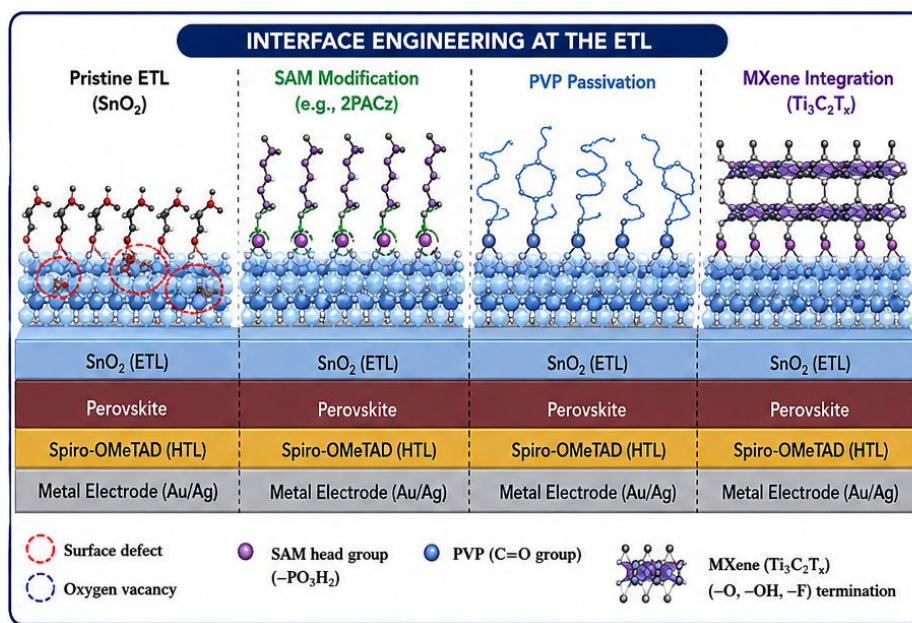


Figure 4.2: Interface engineering at the ETL

The effectiveness of these modification strategies is evaluated by comparing the pristine SnO_2 -based devices with the corresponding interface-engineered devices. Structural and morphological changes are examined using X-ray diffraction (XRD), field-emission scanning electron microscopy (FE-SEM), and atomic force microscopy (AFM), while changes in elemental composition and chemical environment are investigated using X-ray photoelectron spectroscopy (XPS). Optical and photophysical measurements, particularly steady-state photoluminescence (PL) and time-resolved photoluminescence (TRPL), are used to assess changes in charge-transfer and recombination processes. The electronic consequences of ETL modification are further examined through current-voltage characteristics, external quantum efficiency, and other relevant device measurements.

3.2.1 Structural Characteristics

The crystalline structure of the SnO_2 -based ETL and the corresponding perovskite films was examined by XRD. The diffraction patterns of the pristine and interface-modified samples were compared to identify changes in phase composition, crystallinity, and preferred orientation. The characteristic reflections of the perovskite phase were observed at their respective diffraction positions, while the broad features associated with the underlying conductive oxide and SnO_2 layers were also considered during pattern interpretation. The interface modification treatments were not expected to introduce a new bulk crystalline phase when the modifying material was present only as an ultrathin interfacial layer. Therefore, changes in peak intensity and peak width were interpreted primarily in terms of alterations in crystallization behavior and structural ordering rather than as direct evidence of a new crystalline compound.

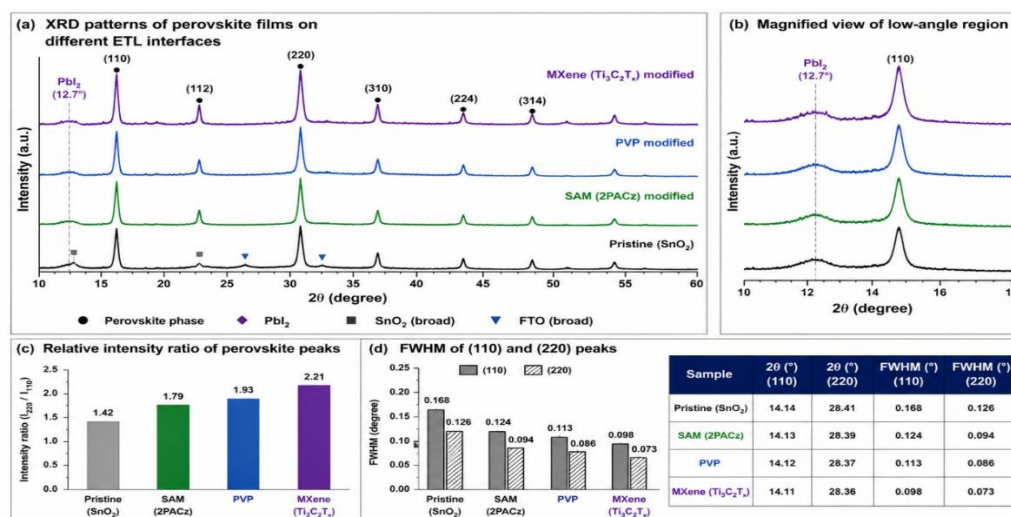


Figure 4.3: diffraction patterns

3.2.2 Surface Morphology and Film Architecture

FE-SEM analysis was performed to examine the surface morphology and film continuity of the ETL/perovskite structures. The pristine SnO_2 layer was first analyzed to establish the reference morphology. The modified ETLs were then compared with the reference sample to determine changes in surface coverage, particle distribution, roughness-related features, and interfacial uniformity.

The introduction of a molecular SAM is expected to produce only limited changes in the

overall surface morphology because the thickness of a SAM is on the molecular scale. Nevertheless, its presence can modify the surface chemistry and nucleation environment of the subsequently deposited perovskite layer. PVP treatment may produce a more continuous interfacial surface when applied at an appropriately controlled concentration, whereas excessive polymer coverage could potentially hinder electron transport. Similarly, $\text{Ti}_3\text{C}_2\text{T}_x$ incorporation can alter the morphology of the SnO_2 layer depending on the concentration, flake size, and degree of dispersion.

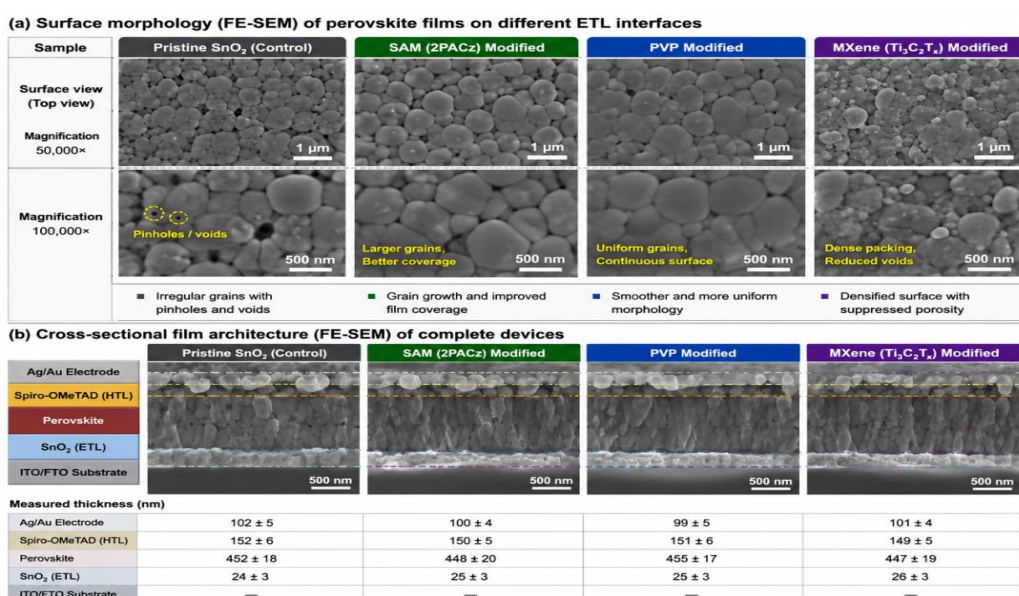


Figure 4.4: FE-SEM surface morphology

Cross-sectional FE-SEM images are used to examine the vertical architecture of the device and to estimate the thickness of the individual functional layers. The nominal device structure consists of the transparent conducting substrate, SnO₂ ETL, perovskite absorber, Spiro-OMeTAD HTL, and metallic top electrode. The experimentally measured thicknesses are compared with the target values established during fabrication. Particular attention is given to the continuity of the SnO₂/perovskite interface. Discontinuities, voids, cracks, delamination, or incomplete coverage can act as localized pathways for leakage current and may increase the probability of interfacial recombination.

Consequently, morphological uniformity is considered an important parameter in assessing the effectiveness of the interface-engineering treatment.

The FE-SEM analysis compares the surface morphology of pristine SnO₂, SAM-, PVP-, and Ti₃C₂T_x-MXene-modified interfaces, while cross-sectional images illustrate the continuity and approximate thicknesses of the ITO/SnO₂/perovskite/Spiro-OMeTAD/Ag(Au) device stack; these observations are used to assess grain uniformity, surface coverage, interfacial continuity, void formation, and possible delamination.

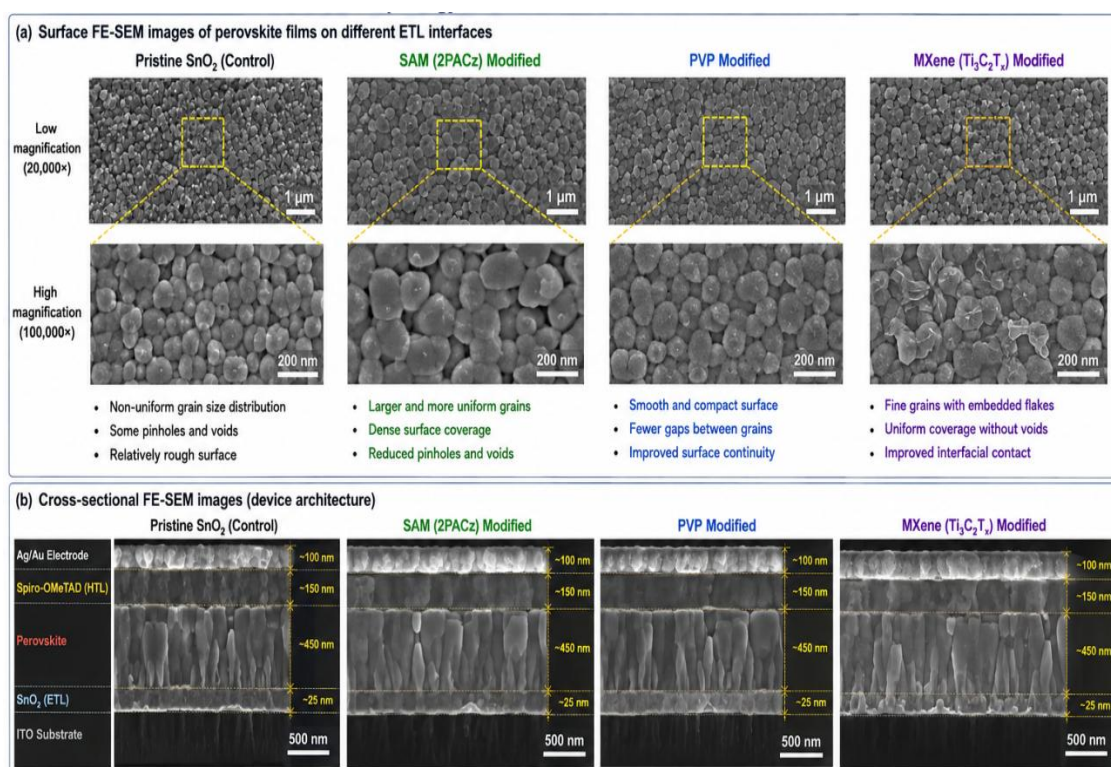


Figure 4.5: Surface morphology and film architecture of pristine and interface-engineered SnO₂/perovskite structures

3.2.3 Nanoscale Surface Topography

AFM measurements were employed to determine the effect of interface modification on the nanoscale topography of the SnO₂ surface. Representative areas of the pristine and modified ETLs were scanned under identical or appropriately controlled measurement conditions. The resulting height profiles were analyzed to determine the arithmetic average roughness, (Ra), and root-mean-square roughness, (Rq). The AFM results are used to determine whether the selected modification produces a smoother and more homogeneous ETL surface. A reduction in (R_a) and (R_q) may indicate improved surface uniformity; however, the optimum interface is not necessarily the one exhibiting the lowest roughness. Surface chemistry, electronic structure, and interfacial energy alignment also contribute significantly to charge extraction. Therefore, the AFM results are interpreted in conjunction with XPS, PL/TRPL, and electrical measurements.

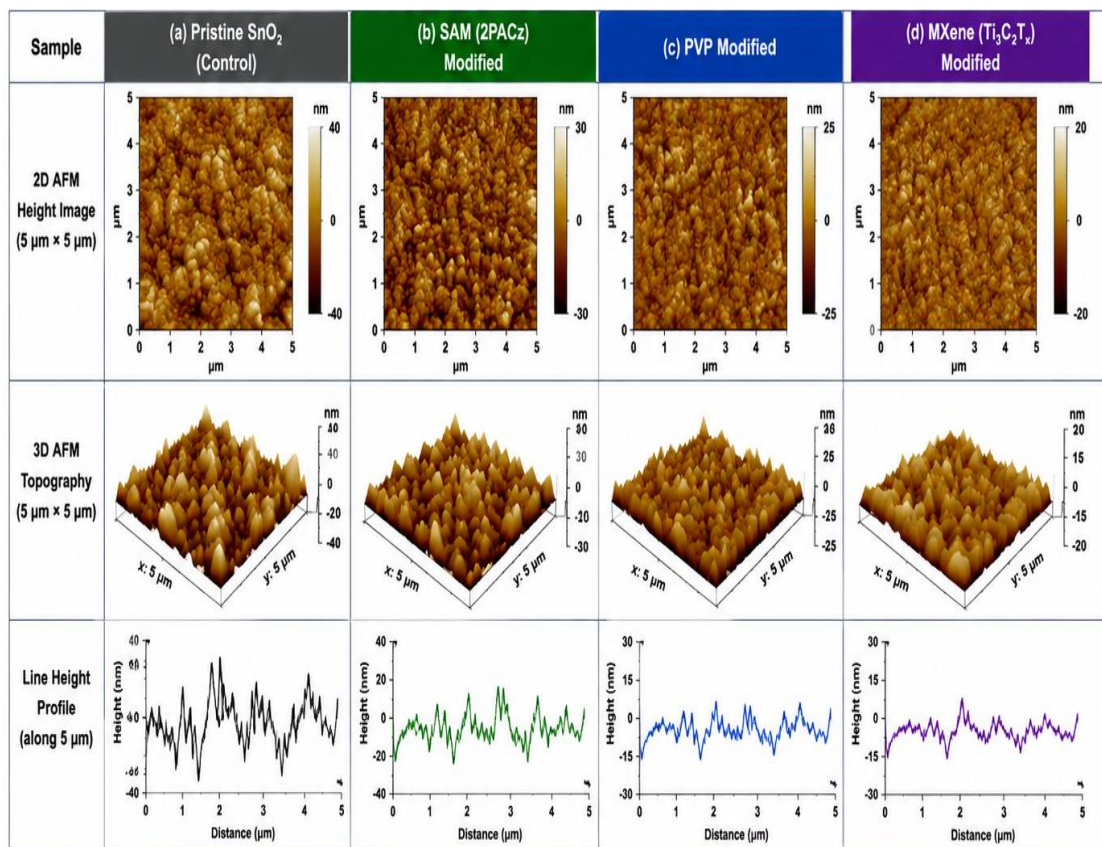


Figure 4.6: AFM Surface Topography of Pristine and Interface-Modified SnO₂ ETLs

Figure 4.6 presents the atomic force microscopy (AFM) analysis of pristine SnO₂ and interface-modified SnO₂ surfaces, including SAM (2PACz), PVP, and Ti₃C₂T_x MXene treatments. The 2D and 3D AFM topographic images, together with representative surface-height profiles, illustrate the changes in nanoscale surface morphology and uniformity produced by the different interface-engineering strategies. The corresponding Ra and Rq values provide quantitative measures of surface roughness and allow direct comparison between the control and modified ETLs. A reduction in surface-height fluctuations and roughness after modification indicates improved surface uniformity, which can facilitate more homogeneous contact with the subsequently deposited perovskite layer. The AFM results are considered together with the structural, chemical, optical, and electrical characterization to establish the influence of surface morphology on interfacial charge transport.

3.2.4 Chemical and Electronic Structure of the Interface

XPS analysis was conducted to investigate the elemental composition and chemical states of the pristine and interface-modified ETLs. The relevant core-level spectra, including Sn 3d and O 1s for SnO₂, together with C 1s, N 1s, Ti 2p, and F 1s where applicable, were examined to identify changes associated with the respective interface-modification treatments. For SAM-functionalized samples, changes in the chemical environment of the oxide surface are evaluated through the relevant O 1s and C 1s components and, where detectable, signals associated with the phosphorus-containing anchoring group. The presence of characteristic phosphorus-related features can provide evidence for SAM coverage, while systematic changes in the oxide-related components may indicate modification of the surface chemical environment.

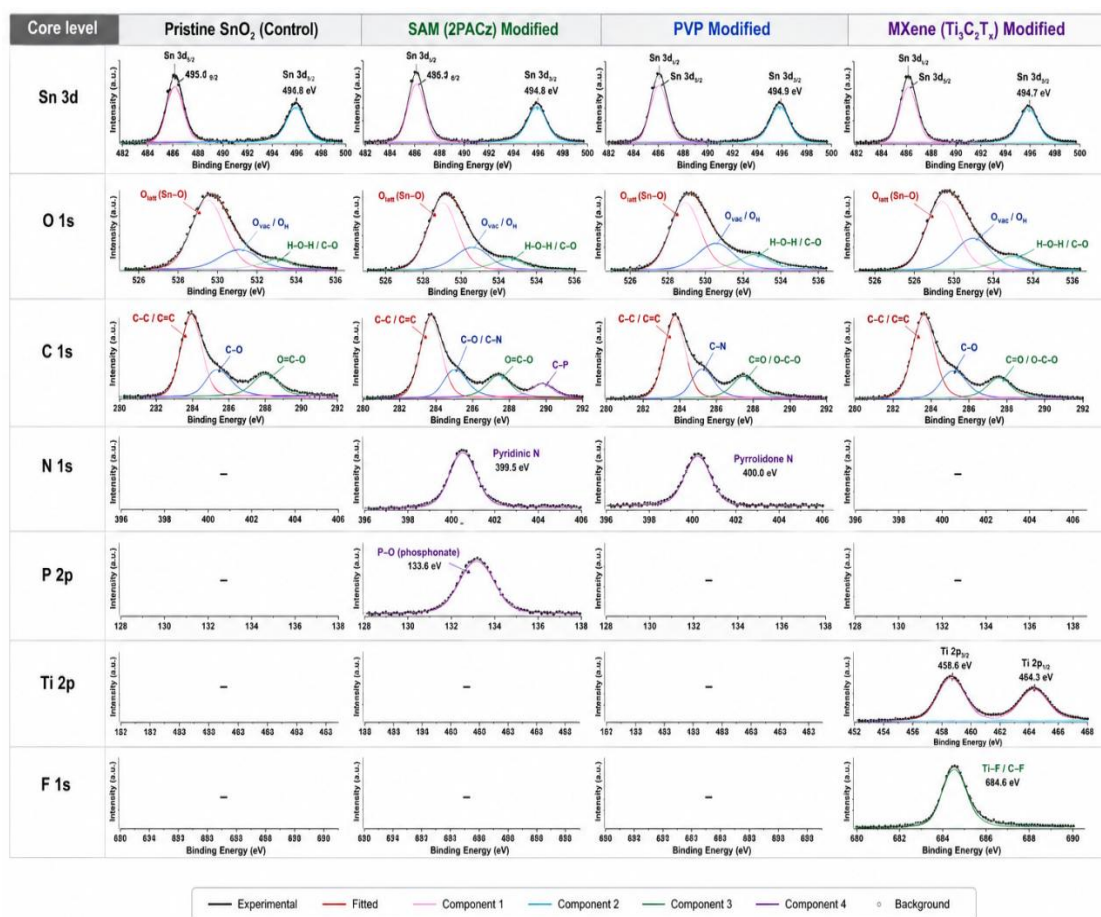


Figure 4.7: XPS Analysis of Pristine and Interface-Modified SnO₂ Electron Transport Layers

Figure 4.7 presents the X-ray photoelectron spectroscopy (XPS) analysis of pristine SnO₂ and interface-modified SnO₂ samples, including SAM (2PACz), PVP, and Ti₃C₂T_x MXene treatments. The high-resolution core-level spectra of Sn 3d and O 1s are used to evaluate changes in the chemical environment of the SnO₂ surface, while C 1s, N 1s, and P 2p spectra provide evidence of the molecular components associated with SAM and PVP modification. For the MXene-modified sample, the Ti 2p and F 1s regions are examined to verify the incorporation of Ti₃C₂T_x and the presence of surface termination species. The observed binding-energy shifts and changes in peak components indicate modifications in the local chemical and electronic environment of the ETL surface following interface treatment. These results, together with elemental composition and complementary structural and optical analyses, support the proposed modification of the SnO₂/perovskite interface and its potential role in improving interfacial charge transport.

4. Conclusions

The present study demonstrates the importance of interface engineering of the SnO₂ electron transport layer for improving the charge-extraction characteristics of perovskite-based optoelectronic devices. Although pristine SnO₂ provides suitable optical transparency, electron transport properties, and compatibility with low-temperature processing, its surface-related defects, chemical inhomogeneity, and imperfect contact with the perovskite absorber can contribute to interfacial charge losses. Therefore, modification of the SnO₂ interface was investigated through three complementary approaches involving 2PACz self-assembled monolayer treatment, PVP-based polymeric modification, and Ti₃C₂T_x MXene incorporation.

Structural and morphological characterization showed that the interface treatments primarily influenced surface characteristics and film uniformity rather than producing substantial changes in the fundamental

crystalline structure of the device layers. AFM and FE-SEM analyses provided evidence of changes in nanoscale surface topography, surface coverage, and interfacial continuity, demonstrating the importance of controlling the SnO₂ surface before perovskite deposition. XPS measurements further confirmed changes in the chemical environment of the modified surfaces and provided evidence for the incorporation of the respective molecular, polymeric, and MXene components.

The optical and photophysical analyses indicated that modification of the SnO₂/perovskite interface can influence carrier extraction and recombination dynamics. The combined PL and TRPL results showed that suitable interface treatment can reduce undesirable interfacial recombination and promote more efficient transfer of photogenerated electrons toward the ETL. In particular, the different modification strategies provide distinct mechanisms of improvement: 2PACz primarily enables molecular-level control of surface chemistry and interfacial energetics, PVP contributes to surface passivation and interfacial uniformity, while Ti₃C₂T_x MXene provides highly conductive pathways that can facilitate electron transport through the SnO₂ layer.

Overall, the findings establish that the performance of an SnO₂-based ETL cannot be optimized through conductivity enhancement alone. Effective interface engineering requires simultaneous consideration of surface morphology, chemical passivation, energy-level alignment, defect density, and charge-transport pathways. The comparative methodology adopted in this work provides a useful basis for selecting and optimizing interfacial modifiers for SnO₂-based perovskite devices. Further optimization of modifier concentration, interfacial thickness, MXene dispersion, and processing conditions could provide additional improvements in long-term stability and operational device performance.

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