Latest advances in TiO_2-perovskite nanostructured interface engineering. Short review

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The evolving perovskite photovoltaic market includes thin-film perovskite, perovskite/silicon tandem and all-perovskite tandem. The future trends are to enhance the efficiencies of the perovskite solar cells (PSCs) and to explore combining the benefits of previous generations with new materials and approaches of the fourth generation of (PSCs). TiO_2 is considered an efficient and stable electron transport layer (ETL) for perovskite solar cells. Tuning the TiO_2 surface layer in perovskite solar cells is a crucial strategy for enhancing device performance and stability. This involves modifying the TiO_2 layer's properties, such as its surface energy levels, charge carrier density, and interface with the perovskite layer, to optimize electron transport and minimize recombination losses. We present the recent advances in the strategies for modeling the interfaces of TiO_2 -based ETLs in perovskite solar cells to improve electron extraction and transport, reduce recombination losses and enhance stability. We provide an overview of the current state-of-the-art research of TiO_2 carriers' selective surfaces in perovskite solar cells by surface modifications, using bilayer structures, doped TiO_2, nanostructured and hybrid materials as well as discussions on challenges and opportunities in the field.

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1. Introduction

The development of solar energy technologies has been driven by the increased energy demand expected in the coming years and the continuous search for renewable and clean energy sources. The cleanest and most abundant renewable energy source available is considered to be the radiant energy of sun.

The International Energy Agency (IEA) in their World Energy Outlook 2020 report stated the solar energy as the cheapest energy in history. In 2021 the IEA report, predicted an increase of solar power production by 2050 that will become the “world’s primary source of electricity”.

The ever-evolving field of photovoltaics is focused nowadays on the 3rd and 4th generations of solar cells.

The first generation of solar photovoltaics is based on monocrystalline and polycrystalline technology. The early high production costs were driven down based on a continuously technological development. The stability and good performance in time allow this traditional solar device to represent up to 90% of today’s market share in spite of the conversion efficiency around 20–25% under the theoretical efficiency of 29.4% [1]. The major drawbacks rely in complex and high energy production costs and loss of efficiency as heat.

The second generation is based on thin layers (films) of photovoltaic materials like amorphous silicon, cadmium telluride (CdTe), or copper indium gallium selenide (CIGS).

Compared to the first generation, these cells are flexible, lightweight and thinner with decreased costs of manufacturing. However, their efficiencies and shorter lifespan due to degradation in time, are lower as compared to conventional solar cell generation.

The environmental impact of these photovoltaic cells due to their toxicity (for instance cadmium content) adds supplementary costs to manage their end-of-life disposal. However, current, on market today there are CdTe and CIGS based photovoltaic technologies [2].

The photovoltaics field is continuously developing the third-generation of solar cells, aiming to exceed the limits of 1st and 2nd generations as surpassing the Shockley-Queisser limit, lower the manufacture costs and find alternative nontoxic materials. The main representants of the 3rd generation solar cells are: Dye-sensitized solar cells (DSSCs), Perovskite Solar Cells (PSCs), Quantum-Dot Solar Cells.

The efficiencies of perovskite solar cells increased within about a decade from 3% [3], in 2009 to 25.5% in 2020 [4], therefore this technology is considered by many scientists as the “future of solar technology”.

PSCs are called after the absorbent materials known as perovskites, that are a class of materials with a particular structure ABX_3, where cations of various atomic radii (A and B) are bonded by an anion X. In the perovskite solar cells, the perovskite crystals with the role of absorbing the photons are sandwiched between two transport layers, the electron transport layer (ETL) and hole transport layer (HTL) respectively, the latest having the role of transporting electrons and holes generated by the perovskite. Since the number of atoms in their structure can be modified, the different perovskites can be tailored to absorb different wavelength of sun spectrum. There are notably advantages that made the perovskite based solar cells the future of solar cells as:

(a) The fabrication technology is solution based, therefore on a lower energy consumption such as heat. The workable thin layered structures need reduced materials' consumption and consequently the fabrication costs are reduced.

(b) Perovskites can be synthesized to have tunable bandgaps and absorption spectrums. They can be tuned to meet the best arrangement of energy levels within carriers' transport layers ETL and HTL.

(c) Perovskite solar cells are flexible and lightweight increasing thus their applicability.

However, the susceptibility to degradation under exposure of heat, light and moisture is one of the most important drawbacks of perovskite solar technologies. The degradation in time causes not only a diminish of lifespan but also environmental concerns since some components of the perovskite structure are toxic. As a consequence, the manufacturing costs are affected.

The 4th generation of solar cells explores new nanostructured materials (as graphenes), new architecture (as tandem solar cells) together with studies aiming to better achieve the goals of the 3rd generation.

To surpass the efficiency limit for a single junction solar cell, tandem solar cells stacks were fabricated. These cells consist of two or more individual cells composed of different absorption materials that due to their different bandgaps can convert a wider range of solar spectrum into electricity. A promising configuration is Perovskite/Silicon Tandem Cells. However different materials with different bandgaps led to efficiency losses due mainly to transmission and thermalization losses. Apart from these, tandem solar cells suffer from some parasitic resistance losses together with the recombination losses existing in perovskite cells for instance. The power conversion efficiency (PCE) of single-junction PSCs has increased in a decade of research, from more than 26% for single-junction PSCs to almost 34% for perovskite–silicon tandem solar cells and 29.1% for perovskite–perovskite tandem solar cells [5].

This study is an overview of the reported work recently published during 2025. The paper systematizes the advances in enhancing efficiencies, stability and reducing the fabrication costs of:

(I) Carbon-based perovskite solar cells following strategies addressed to the interface TiO_2-ETL engineering: (1) Nanostructured interface by treatments with reactive compounds; (2) Nanostructured interface by additional passivating layers; (3) Nanostructured interface by doping TiO_2-ETL;

(II) Lead halide perovskite cells nano structuring the ETL surface: (1) Nanostructure the interface by composite TiO_2 -ETL and additional layers; (2) Modified interface by 3D TiO_2 hierarchical nanostructure; (3) Nanostructure the interface by doping TiO_2-ETL;

(III) Lead free perovskite cells doped TiO_2 based ETL.

2. Perovskite solar cells

The photovoltaic activity of perovskite solar cells based on the absorption of light by the perovskite has as operating steps: (1) the generation of free charges by photon absorption, (2) electron and holes charge transport and (3) charge extraction. The charge carriers (electrons and holes) are produced by dissociation of the excitons produced on their turn by the perovskite light absorption.

Exciton dissociation occurs at the interface between the perovskite layer and the charge transport layer. If the electron is separated from the hole and injected into the electron transport layer (ETL), it moves to the anode, which in many cases is fluorine-doped tin oxide (FTO) glass. At the same time, the hole is injected into the hole transport layer (HTL) and subsequently migrates to the cathode (usually metal). The working and counter electrodes, respectively, collect the electrons and holes to the external circuit and produce current [6,7].

The research studies are driving by the achieving of the power conversion efficiencies as close as possible to the theoretical limit. However, the power producing process is complex involving many parameters.

An important factor contributing to the overall performance of perovskite solar cells is the device configuration. A perovskite solar cell can have a regular (n-i-p) or inverted (p-i-n) structure, depending on the transport material (electrons/holes) in the outer portion of the cell (first encountered by the incident light). These structures can be divided into two subcategories: mesoscopic and planar structures. The mesoscopic structure includes a mesoporous layer, compared to the planar structure which has only planar layers. Perovskite solar cells without electron and hole transport layers have also been tested. As a result, there are six types of perovskite solar cell architectures reported by research studies: mesoscopic n-i-p configuration, planar n-i-p configuration, planar p-i-n configuration, mesoscopic p-i-n configuration, ETL-free configuration and HTL-free configurations [6].

For a chosen device configuration, the overall performance characteristics as current density (J_{sc}), open-circuit voltage (V_{oc}), fill factor (FF), as well as series (Rs) and shunt (R_{sc}) resistances apart from determining the cell power output, provide an insight of the cell working process and identify the most probable limiting factors that path the way towards further research studies [8-10].

Lately, the need of scale-up and commercialization has driven the interest towards a balance between production costs and efficiencies. Thus, the simple manufacturing process, low cost of components, and good stability of carbon-based perovskite solar cells are continuing to be of interest for the optimization of their technologies [11,12,13].

Flexible perovskite solar cells are ideal for applications ranging from wearables to portable devices and their production technologies are continuously under research for reducing the costs of production and enhance the stability preserving the power conversion efficiency as high as possible [14,15].

Due to their toxicity, instability against moisture, heat and irradiation of organic inorganic perovskites an increased interest was driven towards environment friendly lead-free perovskite solar cells in spite of their low power conversion efficiency as compared with lead-based perovskite cells [16].

3. Titanium dioxide (TiO_2) as electron transport layer (ETL)

The ETL in perovskite based solar cells is responsible for extraction of the electrons generated by perovskite absorption layer and transport them to the electrode with the simultaneous prevention of charge recombination by hole blocking. Additional roles as promoting crystallization of perovskite layer and ensure durability and stability of solar cells are desirable for improved solar cell efficiency [6,7][17].

The achievement of these ETL functions is ensured by some critical characteristics of an ETL as: electron mobility, alignment of its conduction band energy level with the band energy of the perovskite layer, best contact with perovskite layer, low absorption in visible light and good processability as continuous defect free films.

Titanium oxide (TiO_2) is a n-type semiconductor extensively studied as an efficient and stable ETL material for perovskite solar cells due to its properties. The conduction band energy of TiO_2 is lower than most perovskites absorbers therefore a good band alignment with effective electron extraction and suppressing hole injection is possible [17].

TiO_2 is environmentally friendly and accessible from natural resources. The synthesis temperature for TiO_2 can be lowered on the cost of low crystallinity. Also, the deposition techniques for TiO_2 films have been established and scalable providing thus a cost-effective production of PSCs [18].

Porous TiO_2 can be engineered to have an increased interfacial contact area with the perovskite layer enhancing thus the electrons extraction [19,20].

TiO_2 is also well known to have hydroxyl groups contained on its surface/interface with perovskite layer facilitating the nucleation and growth of perovskite layers and passivating surface imperfections on the perovskite [21,22].

TiO_2 can be produced in a variety of morphologies and phases, allowing tailored bandgap characteristics to certain perovskite compositions and device structures [23-25].

However, TiO_2 possesses on its surface, more precise at the interface ETL-perovskite some inevitable detrimental defects that can influence the performance improvement of PSCs [26].

As compared with other ETL, the reserves of titanium oxide are estimated at 650 billion tons, being the ninth most abundant element on Earth, while SnO_2, another oxide with good properties as ETL, is less abundant [17].

Overall, TiO_2-based PSCs imply a viable long-term solution for clean and renewable energy, which encompasses research and development to solve the challenges like stability, toxicity, and costs to improve efficiency and commercialize the technology [26].

4. Interfaces TiO_2 - perovskite

The boundaries where two distinct materials meet, also called interfaces are well known to have different characteristics as compared with the bulk of each material. These interfaces can exhibit discontinuities due to differences of physical and chemical properties of the different materials, leading to variations in material behavior across the interface. These discontinuities manifest in various forms, like changes in material's density, conductivity, permittivity, induced stress and strain.

Current density (J_{sc}), open-circuit voltage (V_{oc}), fill factor (FF) and the efficiency of the entire cell of PSCs are strongly influenced by the TiO_2 surface morphologies and phases [27-29], the dimensionality of TiO_2 nanostructures [30-32], the thickness and roughness of TiO_2 layer [33-37], the energy level alignment between the ETL-absorber layer and electrodes for efficient charge extraction [38], and by the defects and traps in the perovskite and TiO_2 layer [39-40].

To promote high efficiencies of the cells, the interfaces between ETL and perovskite should possess some attributes like proper energy level alignment, good physical contact, reduced defects, protective coatings from degradation like moisture, heat, UV. However, at the interface between two different materials, the apparition of defects and misalignments are very likely to occur [17].

One critical process is direct linked to the mobile charge species produced by the absorption layer, which in their move near the junctions, confront the possibility of recombination [41]. The two main mechanisms for the recombination of electron-hole pairs are Shockley-Read-Hall (SRH) recombination and Auger recombination. SRH recombination consists in dissipating the energy as heat due mainly to the defects in the semiconductor's structure. The Auger recombination occurs at higher concentrations of charge carriers and consists in the transfer of the energy from recombination to another free electron or hole [17, 41-42].

Both theoretical and experimental investigations demonstrated that misalignment of energy levels at the junctions of the active layer and ETL generates energy hurdles for electron transfer and results in trapping of electrons at interfacial defects. The energy release is emitted as heat and reduces the open-circuit voltage (V_{oc}) [42-43].

Some defects at the interface, such as vacancies, displacements contribute to the distortions of the electronic structure at the interface resulting in the emergence of so called “deep-trap states” (DTS) that act as recombination centers with the consequences as diminished mobility and lifetime of charge carriers [44].

Tuning the TiO_2 surface layer in perovskite solar cells is a crucial strategy for enhancing device performance and stability. This involves modifying the TiO_2 layer's properties, such as its surface energy levels, charge carrier density, and interface with the perovskite layer, focusing on optimizing the electron transport and minimizing recombination losses [41-44].

During the last decade, an overwhelming amount of research has been conducted in an attempt to overcome the limitations in using TiO_2 as an ETL. Review articles present systematized the advances in enhancing performances of perovskite solar cells [45,17,46-50].

The present paper addresses the latest advances on the interface TiO_2-perovskite strategies for enhancing TiO_2-based ETLs perovskite solar cells. The latest reported advances are summarized in Table 1, ordered from the highest to the lowest reported PCE for carbon based, lead halide and lead-free perovskite cells.

Table 1. Latest reported advances on interface TiO_2-perovskite engineering

Interface modifier	**Type of ETL modification (TiO_2 synthesis route)**	**Type of cell perovskite**	Cell Stability (% of initial efficiency)	**Power conversion efficiency (PCE) %**	**Fill factor (FF) %**	**Open circuit voltage (V_{oc}) V**	**Short circuit current density (J_{SC}) $mA \cdot cm^{-2}$**	Ref
Carbon based perovskite cells								
1-phenylguanidine hydrochloride (PGCl)	Intermediate additional layer TiO_2/SnO_2- PGCl (TiO_2 spin coating-500 ^0C)	CsFAMA lead halide perovskite .	75.4% at 300 h (45-50 °C, RH-60-70%)	15.52	77.96	0.94	21.95	[55]
Zinc acetate $Zn(OAc)_2$	Composite ETL and PAL at the interface (TiO_2 spin coating-200 ^0C)	C- $CsPbI_2Br$-	Not reported	14.91	80	1.248	Not reported	[53]
Rubidium acetate (RbAc)	Composite ETL and PAL at the interface (TiO_2 by spin coating-200 ^0C)	C-$CsPbI_2Br$	90 % at 1800 h (RH-15–25 %)	14.28	Not reported	1.29	Not reported	[52]
Er/Yb-TiO_2 nanocones (NCs)	Doped TiO_2 nanocones (hydrothermal synthesis - 450 ^0C)	$(FAPbI_3)(MAPbBr_3)$	86 % at 30 days	13.38	69.19	0.937	19.56	[59]
Sn doped TiO_2	Doped TiO_2 (TiO_2-spin coating-500 ^0C)	C-$FAMACsPbI_3$	Not reported	12.26	76	0.74	21.8	[58]
Tetramethylammonium hydroxide layer (TMAH)	Intermediate additional layer (TiO_2-spin coating-500 ^0C)	C-$CsPbBr_3$	99.72 % at 30 days	10.13	83.24	1.64	7.42	[54]
Lead halide perovskite cells								
Titanium diboride (TiB_2) TiO_2–TiB_2 heterostructure integrated on SnO_2	Composite floral heterostructure TiO_2–TiB_2 (TiB_2 hydrothermal treatment -140 ^0C, TiO_2–TiB_2 heterostructure--450 °C)	(α-$FAPbI_3$) perovskite	Not reported	23.5	78.81	1.201	24.5	[64]
Potassium bitartrate (KBT)	Composite TiO_2-KBT (chemical bath deposition-150 ^0C)	FAMA PbIBr	80 % at 800 h	23.35	Not reported	Not reported	Not reported	[65]
3D hierarchical	TiO_2 3D hierarchical	$CH_3NH_3PbI_3$	Not reported	19.54	74	1.110	23.8	

Interface modifier	**Type of ETL modification (TiO_2 synthesis route)**	**Type of cell perovskite**	Cell Stability (% of initial efficiency)	**Power conversion efficiency (PCE) %**	**Fill factor (FF) %**	**Open circuit voltage (*Voc*) V**	**Short circuit current density (J_{SC}) mA·cm^{-2}**	Ref
TiO_2 nanorod-	structure over nano-spheres (TiO_2 nanospheres-electrospray deposition, hydrothermal TiO_2 3D hierarchical structure -160 °C)							[69]
N-Acetyl-L-tryptophan (NAT)	Intermediate additional layer N-Acetyl-L-tryptophan (NAT) (chemical bath deposition -110 ^{0}C)	$MAPbI_3$	75.3% at 800 h (RH-30% ± 5%, 25 ± 5 °C).	19.26	Not reported	Not reported	Not reported	[66]
2D-MXene/Janus MoSSe into a mesoporous TiO_2	Composite layer c-TiO_2/mp-TiO_2/MoSSe@MXene@TiO_2 (spin coating- 500 ^{0}C)	$CH_3NH_3PbI_3$	89 % at 500 h	18.33	68.34	1.20	25.14	[67]
TiO_2-GQDs nanocomposite	GQDs doped TiO_2 (QDs - electrochemical process, ETL spin coating 500 ^{0}C)	FAMAPbI$_3$	Not reported	15.10	73.43	0.99	19.82	[74]
Er^{3+} doped TiO_2 (sol-gel)	Er^{3+} doped TiO_2 (Er-TiO_2 nanoparticles sol gel- 550 ^{0}C, ETL-spin coating 110 ^{0}C)	Organic-inorganic perovskite solar cells	Not reported	14.8	0.60	1.06	23.3	[72]
$BaTiO_3/TiO_2$	Composite bilayer $BaTiO_3/TiO_2$ (spin coating 500 ^{0}C)	FAMACs PbBrI-based perovskite	Not reported	14.02	55	0.98	26.01	[68]
Er^{3+} doped TiO_2 (ball-milling)	Er^{3+} doped TiO_2 nanoparticles Er-TiO_2 by ball-milling spray pyrolysis and 450 ^{0}C for TiO_2 film	$CH_3NH_3PbI_3$	Not reported	13.38	65.107	1.012	20.3	[73]
Lead free perovskite cells								
0.5% graphene-doped TiO_2 (TiO_2+Gr)	TiO_2+Gr (0.5 %) ETL device simulation	$RbGeI_3$-based PSC	Not reported	30.14	88.72	1.0622	31.98	[80]
Ce^{4+} dopant ions	Ce^{4+} doped TiO_2 (Ce-TiO_2 particles at 400 ^{0}C, ETL-Spin coated 250 °C)	$(CH_3NH_3)_2CuBrCl$	Not reported	2.21	79.5	0.7707	3.49	[78]
Pr^{3+} ions activated TiO_2	$xPr^{3+}:TiO_2$ (x = 0.07) Pr doped 400 ^{0}C	$(CH_3NH_2)_2CuBr_4$	Not reported	1.6	79.8	0.67	2.97	[79]

5. TiO_2 based ETL in carbon-based perovskite cells

Carbon-based perovskite cells continue to be considered good candidates for commercialization since carbon electrode's fabrication costs are lower and availability is higher as compared with metal electrode perovskite cells [51]. However, the passivating of nonradiative defects at the ETL/perovskite interface aiming to improve the PCE of C-PSCs still remains one of the important challenges [51].

Modifications of interface TiO_2 based ETL and perovskite were focused on enhancing efficiency, stability and reducing the costs occasioned by synthesis at lower annealing temperature.

Latest reported approaches for achieving the up mentioned desiderates are the nanostructuring the interface ETL/PAL by different strategies: (1) Nanostructured interface by treatments with reactive compounds; (2) Nanostructured interface by additional passivating layers; (3) Nanostructured interface by doping TiO_2-ETL.

5.1. Nanostructured interface by treatments with reactive compounds

Treatments with reactive compounds that engineered both ETL and perovskite absorbance layers (PAL) and form nanostructured composite ETL and PAL, was one of the approaches lately reported. The noticeable advance due to this approach with reactive compounds was the reduced fabrication temperature surpassing thus one disadvantage of TiO_2 ETL as compared with SnO_2 for instance. The lower temperature processing of tin oxide films provides an advantage over mesoporous TiO_2, which requires high-temperature annealing (>450 °C). The second advance was the increasing of PCE by reactive compounds that passivate both ETL and perovskite forming nanostructured interface of composite ETL and PAL without introduction of additional layer [52,53].

Yao, Q. et.al. [52] used rubidium acetate (RbAc) molecules as interface modification reactive compound while Hengzhuo Cai et al. [53] used zinc acetate $Zn(OAc)_2$ to optimize the $TiO_2/CsPbI_2Br$ buried interface. In both cases the reactive compound treatment allows a lowered synthesize temperature of 200 ^{0}C and a similar fabrication process for the cell.

The acetate (Ac−) group belonging to $Zn(OAc)_2$ and RbAc interacts with Cs^+/Pb^{2+}, reducing the number of uncoordinated Cs^+/ Pb^{2+} species on the surface and suppressing thus the formation of trap states together with altering the electronic environment forms. These coordination interactions can neutralize the charge of the defect sites of perovskite and prevent them from participating in nonradioactive recombination, actually passivating the defects on TiO_2 surface as well. Acetate ions on the other hand passivate the defects on TiO_2 surface by occupying the oxygen vacancies and coordinate the Ti^{3+}. The small molecule of Rb^+ is very likely to enter the perovskite structure and fill the Cs^+ vacancies in the perovskite lattice and also to enter the TiO_2 layer. Zn^{2+} is more likely to act as doping element for the TiO_2 surface. At the interface ETL/PAL, Rb^+ or Zn^{2+} form nanostructured compounds with titanium oxide and perovskite. The results of this treatment were the interface modification on both TiO_2 and perovskite sites with the improvement of interface morphology and crystallinity, passivating defects and improving energy level alignment.

To conclude, rubidium acetate and zinc acetate form nanostructured composites with TiO_2 and perovskite layers respectively, reducing the interfacial ETL-PAL defects, increasing the conductivity of TiO_2-ETL and optimizing the energy level alignment. The effects of this treatment were achieving PCEs of 14.28% and 14.91%, open-circuit voltage of 1.29 V and 1.248 V for RbAc [52] and $Zn(OAc)_2$ [53] respectively, as well as a long-time stability of 90 % of the initial PCE value [52].

5.2. Nanostructured interface by additional passivating layers

Introducing interface additional passivating layers was another approach to improve efficiencies and stability of carbon-based perovskite cells.

Li et al. [54] investigated the surface defects manipulation by introducing tetramethyl-ammonium hydroxide $[(CH_3)_4]N^+(OH)^-$ molecular layer (TMAH) between TiO_2-ETL and perovskite. TMAH molecule was suggested to passivate the surface defects of TiO_2 by its OH^- group and permeates the perovskite completing its crystalline structure by tetramethylammonium (TMA^+) adjusting thus the energy level by TMAH dipole layer. The authors proved an increase of PCE -TMAH configuration by up to 10.13 % [54], minimizing the energy loss and achieved high V_{OC} of 1.64 V. Modification of the perovskite layer was demonstrated by the authors to preserve 99.72 % of the initial efficiency after 30 days of simulated atmospheric environment. The PCE performance was not very high. However, it is based on the very good quality of perovskite film and opens the way for further improvement for TiO_2 surface passivation. (TMA^+) with a similar structure with perovskite achieves a defects free PAL layer with very good stability of the cell in atmospheric environment. The reported Voc of 1.64 V proved the very good quality of perovskite film in accordance with its stability. However, there are 2 drawbacks. One is related to the very high toxicity of TMAH and the second that TiO_2 is deposited at elevated temperature (500 ^{0} C). The low short circuit current density (J_{SC}) of 7.42 $mA \cdot cm^{-2}$ suggests a lower electron extraction and transport by TiO_2 due either to the defects on the surface or to a lower conductivity.

Wenxiang Zhou et al. proposed a method to optimize the interface radiative recombination and promote the perovskite layer growth using an organic compound 1-phenylguanidine hydrochloride (PGCl) introduced between the double electron transport layer of TiO_2/SnO_2 and perovskite [55]. The PGCl was noticed to be chemically connected to TiO_2/SnO_2 ETL and perovskite layers by electrostatic coupling and hydrogen bonding. The oxygen vacancy density was reduced by replacing with the Cl^- ions from PGCl. The iodine ions in perovskite precursor solution form hydrogen bonding, with PGCl improving the dispersion and crystal growth of the perovskite film. The carbon-based PSC device takes advantages from both double TiO_2/SnO_2 ETL and the interface modification with PGCl showing a PCE of 15.4%. In addition, the used modifier material PGCl proved to stabilize PbI_2 in perovskite [55].

The engineering of the ETL/PAL interface with different reagents that form either composite with the layers or additional films are comparatively presented in Table 2. According to the latest studies, the highest and lowest PCE values of ~15% (PGCl) and 10% (TMAH), respectively, were fabricated at 500 ^{0}C with a 30 days' stability of 75% and 99,7%, respectively. However, a PCE of ~14.9 % and stability of 90% could be obtained by RbAc and $ZnO(Ac)_2$ treatments at lower annealing temperature of 200 ^{0}C. These results path the way towards further studies most probably starting with the passivating mechanism not fully investigated so far.

Table 2. PCE, stability and annealing temperature for interface modifiers for carbon-based perovskite cells

Passivating reagents	**PCE%**	**Synthesis temperature (^{0}C)**	**Stability (%)**	**Reference**
$Zn(OAc)_2$	14.91	200	Not reported	[53]
RbAc	14.28	200	90.00	[52]
TMAH	10.13	500	99.72	[54]
PGCl	15.52	500	75.40	[55]

5.3. Nanostructured interface by doping TiO_2-ETL

Introducing dopants was demonstrated by many studies that can improve electron mobility, conductivity, and charge extraction as previously reported [56-57]. Latest reports continue this approach and test different dopants to modify the ETL in carbon-based perovskite cells.

Rana et al. [58] reported the performances of Sn doped TiO_2 as ETL in carbon-based perovskite cells. Their systematic Taguchi method was utilized to optimize four imperative fabrication parameters: electron transport layer (ETL) type, perovskite absorber layer composition, hole transport layer (HTL) type, and perovskite layer annealing temperature. In their validation experiment the authors reported a PCE of 12.26 %. However, the modified conductivity and energy level alignment of TiO_2 with perovskite is due to both doped ETL and to the passivating materials integrated into the perovskite layer [58].

The replacement of mesoporous TiO_2 with one-dimensional Er/Yb-doped TiO_2 nanocones as electron transport layer (ETL) was the strategy adopted by Lv et al. [59] for a better energy level alignment at the ETL/perovskite interface and demonstrate the superior conductivity, a reduced density of defect states and a better energy level alignment at the ETL/perovskite interface leading to increased short-circuit current density (J_{sc}) and open-circuit voltage (V_{oc}). The one step hydrothermal method implying 450 ^{0}C thermal treatment post deposition was used to fabricate TiO_2 and TiO_2 doped nano cones directly on FTO glass. The Er/Yb-TiO_2 NCs possess high conductivity and conduction band edge facilitating a good energy band alignment. However, the obtained 13.38% PCE is not very high, even if it is based on the synergy effects of RE doping and surface nanostructure.

6. TiO_2 based ETL in lead halide perovskite cells

The research studies on lead halide perovskite cells addressed the main challenges as long term stability to moisture, heat, and light and keep the performances when scale up to commercial production. A key area of research is modeling [60-62] engineering the interface ETL- PAL [63]. Thus, the latest studies were centered on nano structuring the ETL surface aiming to increase the PCE by adjusting the energy level alignment and increasing conductivity. The identified approaches were: (1) Nanostructure the interface by composite TiO_2 -ETL and additional layers; (2) Modified interface by 3D TiO_2 hierarchical nanostructure; (3) Nanostructure the interface by doping TiO_2-ETL.

6.1. Nanostructure the interface by composite TiO_2 -ETL and aditional layers

Composite TiO_2 as a nanostructured ETL or as an additional interlayer ETL/PAL was a recent approach aiming to enhance both efficiency and stability of the cells. The fabrication process was achieved with different reagents. In their reported work, Nouf Alharbi et al. [64] proposed to take the advantages of TiO_2/SnO_2 bi-ETL by modifying the TiO_2 interface. Thus, the authors integrated TiO_2 as a floral heterostructure TiB_2 -TiO_2 in a bi-layer electron transport layer of SnO_2/TiO_2–TiB_2/perovskite/Spiro-OMeTAD/Ag arrangement. The TiO_2/SnO_2 bi-ETL experiences a mismatch in their conduction band minimum that generates an energy barrier at the interface, obstructing electron flow and resulting in charge accumulation. The incorporation of TiB_2 produces an intrinsic electric field which promotes effective charge separation. It was demonstrated an enhancement of lattice compatibility with the α-$FAPbI_3$ perovskite, therefore reducing interfacial strain and facilitating uniform film growth, enhancing thus the durability of the perovskite solar cells. Their manufactured n-i-p PSCs configuration achieved a PCE of 23.5 % [64]. However, the device fabrication temperature was 450 ^{0}C and the overall efficiency of the cell was also due to the very good conductivity of SnO_2.-However the energy level alignment perovskite-SnO_2 was achieved by the TiB_2 -TiO_2

and makes this aproach interesting for further developments.

The interface engineering by the addition of molecular potassium bitartrate (KBT) to the TiO_2 to form a composite ETL significantly enhances TiO_2 surface morphology, leading to a reduction in surface roughness and defect states [65]. The carboxyl and hydroxyl functional groups belonging to the potassium salt of tartaric acid form bonds with titanium interface defects and optimize the energy level alignment of TiO_2/perovskite. KBT also reduced the concentration of non-radiative recombination centers at the interface, by passivating the uncoordinated Pb^{2+} and I^- belonging to the perovskite layer with COO^- and OH^- moieties. The preparation of the cell was achieved at 150 ^{0}C with a 23.35% PCE. The KBT apart from the lower annealing needed temperature, passivated both defects on TiO_2 and on organic/ inorganic perovskite site. Due to optimized defects free ETL/PAL interface over 80 % of the initial efficiency was maintained after 800 h of continuous run.

An acetylamino-functionalized interlayer formed by small molecules was proposed by Y. Chen et al. [66]. The organic modifier N-Acetyl-L-tryptophan (NAT) formed bonds with both Pb in perovskite stabilizing the layer and with TiO_2 passivating the defects on the surface and within the ETL layer. The fabrication of TiO_2 based ETL was performed at 100 ^{0}C. The manufacture device was demonstrated to preserve 75.3% of its initial efficiency [66].

A bilayer ETL formed of compact TiO_2 and composite porous TiO_2 impregnated with two-dimensional (2D) nitrides and carbides (2D MXene) and molybdenum sulfide selenide (MoSSe) nanosheets was proposed by Nouf Alharbi et al. [67]. The recently discovered materials called the 2D layered Janus TMD referred to a XMY compound (where X and Y are different chalcogen elements and M is a transition metal) makes easier for the material to produce an intrinsic electric field. Improved electron mobility and extraction, as well as decreased recombination are the consequences of the MXene/TiO_2 interface. The further integration of MoSSe promotes energy level alignment. The most efficient device developed by the authors consisted of a c-TiO_2/mp-TiO_2/MoSSe@MXene@TiO_2 electron transport layer and demonstrated a PCE of 18.5 %, maintaining 89 % of its initial efficiency after 500 h in ambient air [64]. It is worth to be mention and further improve the high achieved device stability which is a consequence of the very good energy level alignment [67].

Reza et al. [68] proposed a composite nanostructure of $BaTiO_3$/TiO_2 bilayer as electron transport layer. The structure includes a mesoporous TiO_2 layer that serves as a barrier to prevent charge recombination and a mesoporous $BaTiO_3$ layer that facilitates efficient electron transport. Both mesoporous TiO_2 and $BaTiO_3$ layers are deposited by spin coating method using the synthesized nanoparticles and developed paste formulation. The higher band gap energies of ETLs containing $BaTiO_3$ as compared to the pristine TiO_2 ETL inhibited electron-hole recombination. The optimized band alignment between TiO_2 and $BaTiO_3$ leads as reported to improved electron extraction as well as to the enhanced infiltration and growth of the perovskite resulting in an 82 % increase in photo-conversion efficiency compared to conventional TiO_2-based devices [68]. The use of $BaTiO_3$ could be further investigated for its potential for enhancing the performance, stability, and commercial viability of PSCs.

As a general overview, composite TiO_2 –ETL or composite TiO_2 interlayer fabricated with the use of moieties containing $COOH^-$ and OH^- anions, can be fabricated at thermal treatment temperatures lower that 500 ^{0}C and can passivate Pb^{+2} and I^- at the perovskite layer interface, achieving PCE of 23.35 % and 19.26% in the case of TiO_2-KBT and TiO_2/Try, respectively. Stabilities in normal environment condition after 30 days of run were also enhanced (Table 3).

For TiO_2-TiB_2 ETL, the 23.5 % PCE value, even if it is not related only to the TiO_2 composite, the interface structure is worth to be investigated since it achieved a very good Jsc value of 24.5 mA·cm^{-2} (Table 1).

In the case of mTiO_2/MXenes/MoSSe interlayer, the 89% stability reflects a good defect free interface achieved at 500 ^{0}C and the interface engineering approach could provide information on interface modification mechanism (Table 4).

Table 3. PCE, stability and annealing temperature for composite TiO_2 ETL with interface modifiers in lead halide perovskite cells

Composite ETL	**PCE %**	**Annealing temp (^{o}C)**	**Stability (%)**	**Reference**
TiO_2-TiB_2	23.50	450	Not reported	[64]
TiO_2-KBT	23.35	150	80.00	[65]
TiO_2-$BaTiO_3$	14.02	500	Not reported	[68]

Table 4. PCE, stability and annealing temperature for composite TiO_2 based interlayers ETL/perovskite with interface modifiers in lead halide perovskite cells

Composite TiO_2 based interlayer	**PCE %**	**Annealing temp (^{o}C)**	**Stability (%)**	**Reference**
TiO_2/Try	19.26	110	75.30	[66]
TiO_2/MXenes/MoSSe	18.33	500	89.00	[67]

6.2. Modified interface by 3D TiO_2 hierarchical nanostructure

Employing nanostructured TiO_2, such as nanorods or nanocrystals, are known to enhance electron transport and improve perovskite film quality.

Khalid et al. [69] performed an exclusive combination of electrospray and hydrothermal low temperature methods for growing 3D hierarchical TiO_2 nanostructures as ETLs for PSCs. Their 3D hierarchical TiO_2 nanorod-sphere nanostructure-based ETL produced an efficient mesostructured perovskite device with an average power conversion efficiency (PCE) of 19.54% and a maximum of 21.3%. The performance was attributed to the 3D hierarchical structure of the ETL. The nanorod shape reduced electron recombination and increased the overall effectiveness of charge extraction from the perovskite. Because TiO_2 spheres possessed a large surface area, there were many locations available for deposition of the perovskite absorber layer, which increased light absorption and photocurrent production [69]. The proposed cell architecture was demonstrated to be a low cost (synthesis temperature of hierarchical TiO_2 nanostructures was 160 °C) and highly efficient. The synthesis procedure qualifies the device to be further optimized for commercial use.

6.3. Nanostructure the interface by doping TiO_2-ETL

Passivating interface defects of TiO_2-lead perovskite cells through doping the ETL layer was the subject of many studies aiming to enhance the cell efficiencies and improve its stability. The Rare Earth TiO_2 doped ETL still are interesting options for efficient perovskite cells with scalable and economic viable fabrication technologies [70,71].

Er^{3+} may substitute the Ti sites, interstitially, despite its larger ionic radius because it may be energetically more advantageous than the creation of oxygen vacancies. Er^{3+} ions can introduce intermediate energy levels within the TiO_2 band-gap, facilitating improved light absorption and photogenerated carrier dynamics. The impact of Er-doped TiO_2 synthesized *via* a ball milling method was recently reported [72]. The ETL was prepared by spin coating of Er-TiO_2 nanoparticle suspension. Ball milling method produced tetragonal anatase phase of Er doped TiO_2, increasing the efficiency of PSCs by modifying the structure of the TiO_2. The authors claim a potentially effective route for the production of Er^{3+} doped TiO_2 nanoparticles with high crystallinity as paste. The device fabricated with 0.5 %Er content showed a PCE of 13.38%. The proposed ball milling method is worth to be considered as a cheap and scale-up fabrication technology.

The sol–gel route was used for fabrication of Er^{3+}-doped TiO_2 in perovskite cell [73]. The absorption in the ultraviolet and visible light regions was enhanced compared to the undoped ETL layer. The reported device efficiencies were 14.8 % [73].

Combining TiO_2 with other materials like carbon dots can improve electron transport and stability. Enhanced grain size and crystallinity of the perovskite layer, improved photovoltaic performance, reduced trap states, and faster charge transfer dynamics were proved by Sukrit Sucharitakul et al. [74] by incorporation of TiO_2-GQDs nanocomposite in the TiO_2 electron transport layer using an electrochemical method. The proposed one-step coating process for TiO_2-GQDs nanocomposite provides a scalable and commercially viable method for enhancing PSCs through ETL engineering. Devices utilizing the TiO_2-GQDs nanocomposite exhibited increased open-circuit voltage, short-circuit current density, and fill factor, demonstrating enhanced charge transfer and reduced recombination [74].

7. TiO_2 based ETL in lead free perovskite cells

The non-toxic lead-free perovskite cells are subject of recent studies to overcome the environmental issued occasioned by the lead perovskites in spite of their lower PCE as compared to the lead ones [75-77].

In lead - free cells TiO_2 is the preferred ETL. However, the challenges apart from enhancing the performances given mainly by the perovskite absorber, the improvement of TiO_2 surface by passivating strategies are current under study.Ce^{4+} doped TiO_2 was reported to achieve a PCE of 2.21 % in copper-based perovskite cells [78].

Pr^{3+} doped TiO_2 was explored as ETL in lead free cells aiming for aiding the visible light absorbing for Cu-perovskites, since at excitation with UV radiation, Pr^{3+} has emission in red region [79]. The authors demonstrated the incorporation of Pr^{3+} ions into TiO_2 lattice with effects as lowering the conduction band and reducing the TiO_2 band gap. In addition, Pr^{3+} was reported to occupy the interstitial sites at the perovskite interface passivating thus the defects states.

TiO_2-graphene composite was investigated using Solar Cell Capacitance Simulator-one Dimension (SCAPS-1D) [80]. The reported simulated results showed notable improvement of PCE of 30.14 %, by incorporation of graphene in ETL layer achieving thus a suitable ETL bandgap that improved charge carrier transport, minimized interfacial defects, and increased overall device stability.

8. Conclusions

The development of TiO_2 based ETL as a result of its outstanding electron mobility, high stability, and low cost was extensively studied during the latest years. Different recently reported strategies were developed aiming at overcome the drawbacks of TiO_2 as ETL in PSCs and to maximize power conversion efficiency and stability. These issues have shown promise to be solved by methods such as surface modifications, doping, and interface engineering.

The latest reported studies continue the impressive work done during the last decade on studying the interfaces of TiO_2 -based ETLs in perovskite solar cells aiming at improving electron extraction and transport (by surface modification, increased contact area, doping), reducing recombination losses (by interface passivation, defect reduction), enhancing stability (air stability, long-term performance) by intermediate layers at the interface ETL-PAL or by forming compounds with both ETL and PAL.

This paper overviews the reported work focusing on latest published during 2025 and systematizes the advances in enhancing efficiencies, stability and reducing the fabrication costs of:

(i) Carbon-based perovskite solar cells following strategies addressed to the interface TiO_2-ETL engineering: (1) Nanostructured interface by treatments with reactive compounds; (2) Nanostructured interface by additional passivating layers; (3) Nanostructured interface by doping TiO_2-ETL.

(ii) Lead halide perovskite cells nano structuring the ETL surface: (1) Nanostructure the interface by composite TiO_2 -ETL and additional layers; (2) Modified interface by 3D TiO_2 hierarchical nanostructure; (3) Nanostructure the interface by doping TiO_2-ETL.

(iii) Lead free perovskite cells doped TiO_2 based ETL.

For carbon-based devices, the latest reported PCEs were in the range of 10.13-23.5%, preserving 75-99.72% of initial efficiency after 30 days of continuous run in normal conditions. PCEs of 14.28-14.91 % were achieved for 200 ^{0}C fabrication temperature.

In the case of lead halide perovskite cells, 13.38-23.5 % PCEs with stability of 75-89% of initial efficiency after 30 days of continuous run were reported. For maximum device fabrication temperature of 160 ^{0}C, the PCEs were in the range of 19.26-23.35 %.

Lead-free devices with 2.21% PCE were achieved by doped TiO_2-ETL.

Possible future directions

Using TiO_2 as ETLs in perovskite solar cells has proven to have enormous promise and offers a plausible route to their commercialization, hence TiO_2-based PSCs have the potential to develop into a workable and long-term solution for supplying the rising demand for clean and renewable energy with more research and development.

Possible future directions for enhanced performance of TiO_2 based ETL in perovskite photovoltaics are summarized below:

- Continuous tailoring the TiO_2 surface morphologies with larger surface area (as nano cones, 3D nanostructures) in synergy with doping elements;
- Explore doped or nanocomposite TiO_2 based ETL (as Rare Earth, CQDs, graphene), that enhance charge transport, diminish recombination, and increased stability;
- Further development of passivation solutions for ETL/PAL interface with reactive compounds (as organic salts) to reduce surface and trap defect states, enhancing thus the electrons extraction and stabilizing at the same time the perovskite layer;
- Investigating fabrication methods at lower temperature (<200 ^{0}C) for TiO_2 based surface nano-architecture with increased surface area and perovskite contact;
- Development of scalable and economical fabrication techniques (as solution-based technologies, ball-milling) for TiO_2 ETLs necessary to enable PSCs to be produced on a large scale;
- Concentrate the research on environmentally acceptable and sustainable ways to synthesize TiO_2-based ETL, (without the use of toxic reagents) with stability effect on perovskite layer reducing thus the cell's negative effects on the environment;
- Modeling the key fabrication parameters to optimize the method for electron transport layer (ETL) type, perovskite absorber layer composition, hole transport layer (HTL) type, by designed experiments, that can identify the optimal fabrication conditions to maximize power conversion efficiency (PCE).

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