Functional structured coatings for perovskite solar cells – a short review of recent advances

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The emergence of new materials such as perovskites has revolutionized the field with their potential for high efficiency and low production costs. Functional coatings play a crucial role in enhancing the efficiency, stability, and scalability of perovskite solar cells. These coatings can improve light absorption, reduce reflection, protect against environmental factors, and facilitate charge transport. They are essential for transitioning perovskite solar cells from research labs to commercial applications. The present paper reviews the recent advancements in synthesis and characterization of functional structured coatings for perovskite solar cells.

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1. Introduction

Photovoltaic technology (PV) plays a key role in the transition from fossil fuels to sustainable energy sources. To meet the goals of the Paris Climate Agreement [1] the annual production capacity should increase to the terawatt (TW) scale [2].

By 2030, zero-carbon solutions could be competitive in sectors representing over 70% of global emissions [1]. To limit global warming to 1.5°C, greenhouse gas emissions must peak before 2025 at the latest and decline 43% by 2030.

Although climate change action needs to be massively increased to achieve the goals of the Paris Agreement, the years since its entry into force have already sparked low-carbon solutions and new markets. More and more countries, regions, cities and companies are establishing carbon neutrality targets. Zero-carbon solutions are becoming competitive across economic sectors representing 25% of emissions. This trend is most noticeable in the ***power and transport sectors*** and has created many new business opportunities for early movers.

The estimated needed global PV generating capacity will be about 70 TW by 2050 [2]. Therefore, the deployment of PV systems must be accelerated to reach a fast growth (>25%) until at least 2032 to avoid a major market downturn in 2050. Verlinder described the future challenges for photovoltaic manufacturing at the terawatt level such as: hydro, wind, solar, geothermal, biomass, synthetic fuel, etc. [2].

The raised global energy demand and the more pronounced effects of climate change is the driving force for the enhancements in PV efficiency. The recent advances in solar PV technologies, are focusing on efficiency improvements, material innovations, and expanded applications.

The improvements in silicon cell technology have enhanced the performance of solar panels while reducing production costs. Recently, significant advancements for high-efficiency photovoltaic cells like multi-junction solar cells, passivated emitter rear cells (PERC), and bifacial solar cells were made [3]. The multi-junction solar cells consist of multiple stacked semiconductor layers, each designed to capture different parts of the solar spectrum. The record efficiency for multi-junction cells has surpassed 47% under concentrated sunlight conditions. The high production costs and complex manufacturing processes of these cells have restricted their use to specialized applications, such as space missions and concentrated solar power systems [4]. PERC technology enhances the performance of conventional silicon solar cells by adding a passivation layer which reduces the recombination losses and improves the overall efficiency of the cell. Efficiency enhancements achieved through PERC technology have led to commercial cells with efficiencies exceeding 23%, compared to the 15-17% efficiency range of traditional silicon cells. Bifacial cells designed to capture light on both the front and rear sides of the panel, effectively utilizing reflected and diffused sunlight have increased the overall energy production and demonstrated efficiency gains of up to 30% compared to traditional mono-facial cells, particularly in installations where ground reflection is significant.

Recently, the introduction of new materials offering potentially cheaper alternatives to traditional silicon-based solar cells made possible the development of new devices such as perovskite solar cells and organic photovoltaics (OPVs). The innovations in materials have significantly impacted the PV field, particularly through the developments of perovskite solar cells, organic photovoltaics (OPVs), and quantum dot solar cells.

The use of perovskite materials with a high absorption coefficient, a long diffusion length, a customizable band gap, minimal carrier recombination loss, and high carrier mobility, represented a significant advancement in the field of photovoltaics [5,6].

Organic materials used in organic photovoltaics (OPVs) offer several advantages, including flexibility, lightweight, and aesthetic appeal. Recently improvements in material design and device architecture have led to efficiencies exceeding 18%, up from previous levels below 10% [7]. Practical applications of OPVs are expanding, with commercial products such as flexible solar panels and integrated photovoltaic materials for textiles becoming more prevalent [8].

Quantum dot solar cells offer advantages such as tunable bandgaps enabling the design of devices that can absorb a broader range of wavelengths compared to traditional PV materials. Quantum dot solar cells represent a promising frontier in PV technology, with unique advantages in light absorption and conversion efficiency but facing challenges in scalability and material stability [3].

Tandem solar cells are part of the next generation of PV technologies that will allow to fabricate PV modules with efficiencies above 30%. This photovoltaic (PV) technology combines two (silicon-perovskite or perovskite-perovskite sub cells) or more solar cells with different band gaps to capture a wider spectrum of sunlight and convert it into electricity more efficiently than single-junction cells. Currently, several technologies are considered, with different semiconductor materials and different configurations (two terminals, three terminals, or four terminals).

1.1. Perovskite solar cells (PSCs)

PSCs demonstrate certified efficiencies that compete all other thin-film PV technologies, reaching above 25.5% and 32.5% for single-junction and tandem with silicon SCs, respectively [9-11].

Perovskite solar cells (PSCs) are becoming increasingly attractive for commercialization owing to their rapid increase in power conversion efficiency, easily scalable fabrication, reliable operation and compatibility with various perovskite-based tandem device configurations. *Perovskites* have the chemical formula ABX_3, in which A, B and X stand for the monovalent cation (methylammonium ($CH_3NH_3^+$, MA^+), formamidinium ($(NH_2)_2CH^+$, FA^+), cesium (Cs^+) and rubidium (Rb^+)), the divalent metal cation (such as lead (Pb^{2+}) and tin (Sn^{2+})) and the halide anion (such as chloride (Cl^-), bromide (Br^-) and iodide (I^-)), respectively.

Perovskites with a *wide-bandgap* (WBG) around 1.65 – 2.0 eV often involving a Br⁻I mixture with a high Br content (>15%) are used as the top cell in tandem configurations. Perovskites with a *narrow-bandgap* (NBG) of about 1.2–1.3 eV can be obtained through Sn⁻Pb B-site alloying and are often used as the bottom cell in all-perovskite tandem devices. For Sn–Pb perovskites, Sn^{2+} oxidation and the corresponding defect formation represent the biggest challenge [12]. The perovskite layer is responsible for multiple functions, including light absorption, charge generation and separation. An ideal perovskite layer generally has several features: compactness and uniformity, large apparent grain size, suitable bandgap for specific applications, high charge carrier mobility and long charge carrier diffusion length. A perovskite solar cell comprises the perovskite absorbing layer (PAL) placed between the electron transporting layer (ETL) and hole transporting layer (HTL).

An important factor in evaluating the overall performance of perovskite solar cells is the device configuration. The classification of perovskite solar cells according to the transport material (electrons/holes) in the outer portion of the cell is as regular (n-i-p) and inverted (p-i-n) structure, depending on the transport material (electrons/holes) in the out-er portion of the cell (first encountered by the incident light).

Rapid progress was made during the past decade by numerous investigations focused mainly on inner device factors like new materials/composites/structures, device architecture, interfaces between layers, stability.

Enhancement of the conversion efficiency (PCE) involves improving the open-circuit voltage (V_{OC}) and fill factor (FF) as well as the short-circuit current density (J_{SC}).

Since the device architecture, material design, and optical properties of PSCs are consistently being developed and approaches the practical limit, light managing technologies became an important research topic as well as the perovskite solar cell modelling and device characterization [14].

2. Anti-reflective coatings for perovskite solar cells

The energy loss in PSC_s is mainly due to the intrinsic properties of the perovskite and to the thermodynamic constraints [15]. The intrinsic loss is related to the difference between the photons energy and the bandgap of the perovskite, having as results the sub-band transmission or thermalization of hot carriers. The other loss mechanisms include absorber–emitter radiation (emission loss), heat transfer (Carnot loss), and entropy generation (Boltzmann loss), optical loss [16]. The optical losses are related to the reflection loss occurring when the incident light is predominantly reflected in its way through the interface between the air and device and/or when the light is not fully absorbed by the device and it is reflected by the rear electrode. The former occurs owing to the difference in the refractive indices of the two interfacing materials, as refraction and reflection of light occur.

The different types of optical loss (reflection, transmission, and parasitic absorption), have been estimated in numerous studies through simulations and experiments. The ratio between each factor depends on the stacking order of the materials and their thicknesses, as well as the method of simulation and/or experiment. The proportion of total optical loss to the incident light is approximately 13–25% where reflectance, transmittance and parasitic absorption loss occupy about 5–9%, 1–5%, 7–11%, respectively. Therefore, it can be concluded that reflectance and parasitic absorption constitute the majority of the losses, while the transmission loss is minor, especially with the perovskite having a high extinction coefficient [17-21].

Antireflective (AR) coatings are applied to perovskite solar cells to minimize light reflection and enhance light absorption, thereby improving their power conversion efficiency. These coatings work by reducing the refractive index difference between the perovskite cell and the air, which minimizes reflection losses and allows more light to enter the cell and by increasing light's path length within the cell by a scattering mechanism. To mitigate reflection loss, various light management strategies have been pursued, such as incorporating single or double anti-reflective coatings (SL-AR or DL-AR), a thin layer with a gradient refractive index, composite layers, nanostructured coatings [17].

2.1. Structured monolayers/multilayers

In one or more layered structure the ratio of transmitted over reflected energy depends on refractive indexes of the involved materials, on the wavelength, incidence angle, thickness of the additional AR films. According to the Fresnel equation an ideal refractive index for an AR layer should have and index between the refractive indexes of the materials placed in front of and behind it (usually air and soda-lime glass). For different incidence angle and the entire wavelength region there are few materials to meet these characteristics. Some of these materials for AR could be MgF_2 (n = 1.23), polydimethylsiloxane (PDMS) (n = 1.43), SiO_2 (n = 1.46), polymethyl methacrylate (PMMA) (n = 1.5), and Al_2O_3 (n = 1.65). However, the optical losses due to reflection depend also on the thickness of the AR coating and therefore to determine the optimal material for an AR coating simulation are necessary [22].

To start to build a perovskite cell, the composite anti-reflective coatings should be engineered to cover a wider range of wavelengths by incorporating monolayers or multiple composite layers of inorganic materials like MgF_2, SiO_2, TiO_2, Al_2O_3, ZnO or polymers like polymethyl methacrylate (PMMA) polydimethylsiloxane (PDMS) or composites [23]. The optical loss simulation should be conducted as a function of the perovskite active layer composition. Thus, the optical simulation on metal halide perovskite solar cells was conducted and the optical losses, including reflectance, transmission, and parasitic absorption were estimated for different thickness of AR MgF_2 coating [24,25].

Erdin Almuqoddas [26] simulated the integration of various materials for AR as poly(methyl methacrylate) (PMMA), magnesium fluoride (MgF_2), lithium fluoride (LiF), poly(dimethyl siloxane) (PDMS), and calcium fluoride (CaF_2), each offering advantages such as low refractive index, optical transparency, ease of deposition, and cost-effectiveness, rendering them promising for future applications. It was found that the inclusion of PMMA-based SL-AR with a thickness of 70 nm could enhance the PCE values up to 29.7 % [26].

Among different materials that meet the requirements SiO_2 was mostly investigated as a possible AR for perovskite cells. The synthesis process of SiO_2 is easy, inexpensive, and scalable using solution-based techniques, it can be deposited on substrate as different shapes (nanospheres, nanoparticles, mesoporous films, doped in polymers) and last not the least it has the advantage of the experience gained in silicon cells technologies. For $CH_3NH_3PbI_3$ PSCs, the effective refractive index of an AR film of SiO_2 nanospheres deposited by spin-coating of an aged silica sol at 1000 rpm was closest to 1.23, where the antireflective coating on the glass substrate achieves effectively 0% reflection at a wavelength of 550 nm. The power conversion efficiency was improved from 14.81% for reference device without SiO_2 nanospheres to 15.82% for the PSC device with the optimized AR [27].

A double-sided antireflection coating was developed to synergistically improve the transmittance of polymer–ITO electrodes. The flexible electrode polyethylene naphthalate (PEN)-ITO was sandwiched between double-sided antireflection coatings, SiO_2 NPs on the PEN side and polymer–metal oxide composite layer (PMO) on ITO side [28].

Wang et al. fabricated mesoporous and hollow silica antireflection coatings by spin coating and demonstrate the reduced Fresnel reflection at the air/substrate interface of PSCs. They reported and enhanced PCE from 19% to above 20% [29].

Yalun Wang et al. improved the transmittance of the glass substrate from 90% to 95% and got a PCE enhancement for PSCs by preparing a porous antireflective coating and a bilayer broadband antireflective coating with mesoporous silica nanoparticles [30].

The optical loss including reflection loss, absorption loss, and transmission loss in printable mesoscopic perovskite solar cells (p-MPSCs) was analyzed by Xiadong Wang et al. [31].

The authors developed a mesoporous SiO_2 antireflection coating printed on the fluorine-doped tin oxide (FTO) glass substrate and reduced the optical reflection at the air/glass interface. Porosity and thickness of the SiO_2 film was modulated to obtain a graded refractive index interface. The authors reported an increase of the transmittance of FTO glass by ≈2%–4% in the spectral range of 350–800 nm at normal incident angle with the highest transmittance improved from 85% to 89%. The SiO_2 coating also exhibits wide-angle and broadband antireflection properties. The coatings successfully help p-MPSCs to obtain about an average 3% enhancement in the short-circuit current density (J_{SC}) and PCE [31].

The working wavelengths of organic/inorganic hybrid materials range from the deep ultraviolet to the near-infrared region, covering the entire visible region and qualifies them as antireflection films for perovskite solar cells. A TiO_2 doped polyvinyl alcohol (PT) film was used to mitigate reflection loss. The refractive index of the hybrid material was controlled in the range of 1.65–1.95 at a wavelength of 550 nm. The PT/Cellulose Acetate (CA) and PT/Polymethyl Methacrylate (PMMA) double-sided antireflection films were designed and prepared on a large area (10 × 10 cm^2) glass substrate with a high transmittance of 98% and 99.3%, respectively. The CA/PT/CA and PMMA/PT/PMMA antireflective hybris films remained stable after aging for 240 days [32].

Theoretical studies and simulations complement experimental investigations and provide valuable guidance for selecting ARCs with appropriate materials and thicknesses to enhance solar cell performance. Various materials for AR were considered, including poly(methyl methacrylate) (PMMA), magnesium fluoride (MgF_2), lithium fluoride (LiF), poly(dimethyl siloxane) (PDMS), and calcium fluoride (CaF_2), each offering advantages such as low refractive index, optical transparency, ease of deposition, and cost-effectiveness, rendering them promising for future applications. The simulations for integrating on $MAPbI_3$-based inverted p-i-n PSCs of an SL-AR composed of PMMA, with a thickness of 70 nm, lead to a notable increase in PCE by 29.7 %. Additionally, the DL-ARC design using PMMA/PDMS (poly(dimethyl siloxane) is anticipated to achieve a maximum PCE of 30.3 % with a thickness of 150 nm in the second layer [26, 33].

The polymer-based substrate such as polyethylene naphthalate (PEN) used in flexible PSCs (FPSCs) has a higher refractive index than that of the glass-based substrate in rigid PSCs (RPSCs), which leads to inferior light transmittance. The high refractive index induces an increase of reflectance at the substrate interface, reducing the light entering the FPSC and leading to a loss in short-circuit current density (Jsc). In addition, direct development of flexible substrate is also highly challenging because it requires significant modifications to conventional PSC fabrication processes or changes to the properties of substrate. A sticker-type multilayer nanostructured anti-reflective (GSMA) film derived from the wings of the glasswing butterfly was proposed to manage the optical losses for FPSCs. The butterfly wings consist of double layers of chitin and wax, with nanostructures between each layer. This unique structure induces a highly smooth change in refractive index, which is beneficial for light transmittance according to Fresnel equation. The GSMA film effectively improved the optical properties of PSC substrates, reducing reflectance (~5.01%) and enhancing light transmittance (~6.17%) in indium tin oxide (ITO)/PEN [34].

2.2. Composite structured layers

ZnO, due to its very wide band gap and high refractivity was a good option as an antireflection coating on silicon-based solar cells. Composite structured of cobalt-doped ZnO thin films, offer superior anti-reflection properties, making them promising candidates for enhancing solar cell efficiency. The anti-reflection performance was improved, using an AR of ZnO films doped with 6 wt% cobalt complexes ($C_{16}H_{11}N_3Cl_2Co$) by achieving a 7.97 % reduction in reflectance compared to those doped with commercial cobalt [35].

2.3. Nanostructured architectures

The integration of perovskite cells with silicon cells in tandem solar cells while simultaneously reducing losses from reflection and parasitic absorption represents a challenge. To reduce optical losses, silicon cells usually have microscopic pyramid patterns etched on their surface by highly corrosive chemical agents, which lessens the reflection of the entire device and increases its current. As perovskites are sensitive to a range of chemical substances, less effective planar anti-reflective coatings tend to be applied. The use of the direct nanoimprinting method makes it possible to produce the complete device in just one technological process, which is key for reducing overall device costs. Apart from enhancing the efficiency, the application of this procedure for this layer does not harm the perovskite. A thermally assisted UV roller nanoimprint lithography (TUV-Roller-NIL) was proposed and found to be a suitable and scalable process for the structurization of perovskite solar cells. A TUV-Roller-NIL-based fabrication of the honeycomb texture on epoxy-based negative photoresist material (SU-8 photoresist) increased the PCE with 2.2% absolute as compared to the planar reference sample, beating the commonly used MgF_2 planar layer [36].

During last decade anti-reflective nanostructured architecture coatings as the biomimetic moth-eye, nanocone arrays, inverted pyramids were proposed due to the properties of these various forms of microstructures to make the light transmission between the substrate of the device and air to meet a gradient refractive index and permit the absorption of the diffuse light [37].

Lately, a honeycomb-like textured SU-8 photoresist layer (SU-8 is an epoxy-based negative) photoresist was applied using a roller nanoimprint technique onto a planar perovskite solar cell to minimize reflection losses. The results show that the applied honeycomb pattern reduces the solar-weighted reflectance from 13.6% to 2.7%, which enhances the current density of the unmodified cell by 2.1 mA cm^{-2}, outperforming the commonly used planar MgF_2 antireflective coating by 0.5 mA cm^{-2} [38].

It was reported that moth eyes have a perfect sub-wavelength antireflective structure on the corneal surface with great transmittance over a broad spectral range from ultraviolet, through the visible part of the spectrum, to the terahertz region [39]. Recently, biomimetic moth eye nanostructures have been artificially fabricated with 3D sub-wavelength structures in the form of nanorods, nanograss, nanotips, nanocones, etc. from polymer, Si, SiO_2 and TiO_2 [40,41].

A nanopatterned MgF_2/SiO_2 AR structure fabricated by combining the single-layer MgF_2 interference effect with a two-dimensional (2D) SiO_2 moth-eye AR structure was applied to the front side of a glass/ITO structure to reduce the surface Fresnel loss from the interface between air and the glass. The PCE of PSCs with the optimal MgF_2/SiO_2 AR structure increased by 12.50% [42].

A large-area scalable manufacturing of microlens array (μLA) -based light management scheme with the lens array placed external to the device without disturbing the internal active layers can enhance absorption and photocurrent in perovskite solar cells by >6%, through light focusing within the active layer. A typical polymer (e.g. polystyrene, polyurethane) index-matched to glass is spin-coated on a thick glass substrate. The polymer is then stamped using a Polydimethylsiloxane (PDMS) mold replicated from a flexible master pattern under the effect of temperature and pressure. The PDMS mold is then released to reveal the inverse of the PDMS pattern on the polymer surface [43].

2.4. Integrated structures to manage the optical losses

Antireflective structures, inverse opal and diffraction gratings simultaneously integrated into PSCs increase the light absorbance of the PSCs from 88.0 % to 93.74 %.

The AR was formed of SiO_2 microspheres with hollow structure with a core of 30 nm and a shell thickness of 20 nm. The two-dimensional (2D) inverse opal SnO_2 electron transport layer and the diffraction grating structure was formed on perovskite by using a nanoimprinting technique using PDMS as a grating template. The composite triple demonstrated the performance improvements, including a 5.74 % light absorption enhancement, and a 20 % increase in power conversion efficiency (PCE) [44].

A double-layer SiO_2 film with graded refractive index was prepared using pure SiO2 film as first layer and polyethylene glycol (PEG)-modified SiO_2 film as the second layer. The double layered coating is multifunctional film with anti-reflection, high hardness, and self-cleaning properties [45].

Mingfang Huo et al. proposed a TiO_2 polyvinyl alcohol hybrid film sandwiched between cellulose Acetate (CA) or polymethyl methacrylate (PMMA). Due to the intrinsic high absorbance of the organic/inorganic hybrid films stack in the UV region, such a design should play a major role in preventing the UV-induced degradation [46].

In the future, photovoltaic industry will favor multifunctional film with anti-reflection, high hardness, and self-cleaning properties due to the complex outdoor environment.

Double-layer broadband SiO_2 antireflective film with high transmittance increase and excellent mechanical properties was successfully prepared via an acid-catalyzed sol–gel method using polyethylene glycol (PEG) as pore-forming agent. The porous PEG2000-modified SiO_2 films were selected as the outer layer for coating onto the pure SiO_2 film to form double-layer SiO2 films with graded refractive index. The water contact angle (WCA) was also reduced to 6° while that of pure SiO_2 film was 51.3°, endowing the double-layer SiO_2 film superhydrophilic and anti-fogging performance [47].

Transparent luminescent Eu-complex down-converting material (LDC) into siloxane matrix was used as an external coating layer of carbon-based perovskite solar cells (C–PSCs). The coating enhances the solar cells' power conversion efficiency (PCE) and simultaneously acts as an anti-reflective and UV-shielding layer. Eu-complex down-converting material UV light absorption and re-emission in visible light, as well as its antireflection property had as result an increase of short circuit current (Jsc) [48].

3. Conclusion

This short review presents the recent advances in materials/structures for optical loss management in perovskite photovoltaic cells. SiO_2 among different materials that meet the requirements for optical management coatings was mostly investigated as a possible AR for perovskite cells since its synthesis process is easy, inexpensive, and scalable using solution-based techniques, it can be deposited on substrate as different shapes (nanospheres, nanoparticles, mesoporous films, doped in polymers) and it has the advantage of the experience gained in silicon cells technologies.

Anti-reflective nanostructured architecture coatings as the biomimetic moth-eye, nanocone arrays, inverted pyramids and honeycomb structures were recently investigated as films for light management in perovskite cells due to their properties of these various forms of microstructures to make the light transmission between the substrate of the device and air to meet a gradient refractive index and also to permit the absorption of the diffuse light.

Recently the strategies to suppress the reflection loss at the interface cell/ air aiming to enhance the perovskite solar cell efficiency is combined with the techniques that improve the long-term stability under environmental factors such as humidity, temperature, and light exposure. Thus, multifunctional films that promotes enhancement of efficiency together with an optimal balance between efficiency and stability are continuously developed.

Possible future research trends could be:

- Design of nano multifunctional new structured films with antireflection, encapsulating and protection synergetic effect for perovskite photovoltaics.
- Theoretical and experimental research on functional films and coatings for improving the stability, efficiency, and scalability of perovskite solar cells.
- Multifunctional films and coatings produced by low costs solution-based and scalable printing techniques.
- Experimental and processing advancements in high-performance coatings for PSCs exposed to environmental stressors such as moisture, heat, UV radiation, and mechanical strain.
- Computer modeling and simulation to predict the properties, performance, durability, and reliability of functional films and coatings in PSC devices.
- Development of novel test methods to evaluate the interplay between stability, efficiency, and scalability, with an emphasis on accelerated aging tests and correlations between laboratory results and field performance.

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