



Antireflective Self-Cleaning TiO₂ Coatings for Solar Energy Harvesting Applications

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Titanium(IV) oxide (TiO₂, titania) is well-known for its excellent photocatalytic properties, wide bandgap, chemical resistance, and photostability. Nanostructured TiO₂ is extensively utilized in various electronic and energy-related applications such as resistive switching memory devices, flat panel displays, photodiodes, solar water-splitting, photocatalysis, and solar cells. This article presents recent advances in the design and nanostructuring of TiO₂-containing antireflective self-cleaning coatings for solar cells. In particular, the energy harvesting efficiency of a solar cell is greatly diminished by the surface reflections and deposition of environmental contaminants over time. Nanostructured TiO₂ coatings not only minimize reflection through the graded transition of the refractive index but simultaneously improve the device's ability to self-clean and photocatalytically degrade the pollutants. Thus, novel approaches to achieve higher solar cell efficiency and stability with pristine TiO₂ and TiO₂-containing nanocomposite coatings are highlighted herein. The results are compared and discussed to emphasize the key research and development shortfalls and a commercialization perspective is considered to guide future research.

Keywords: antireflection, coatings, renewable energy, self-cleaning, solar cells, titania

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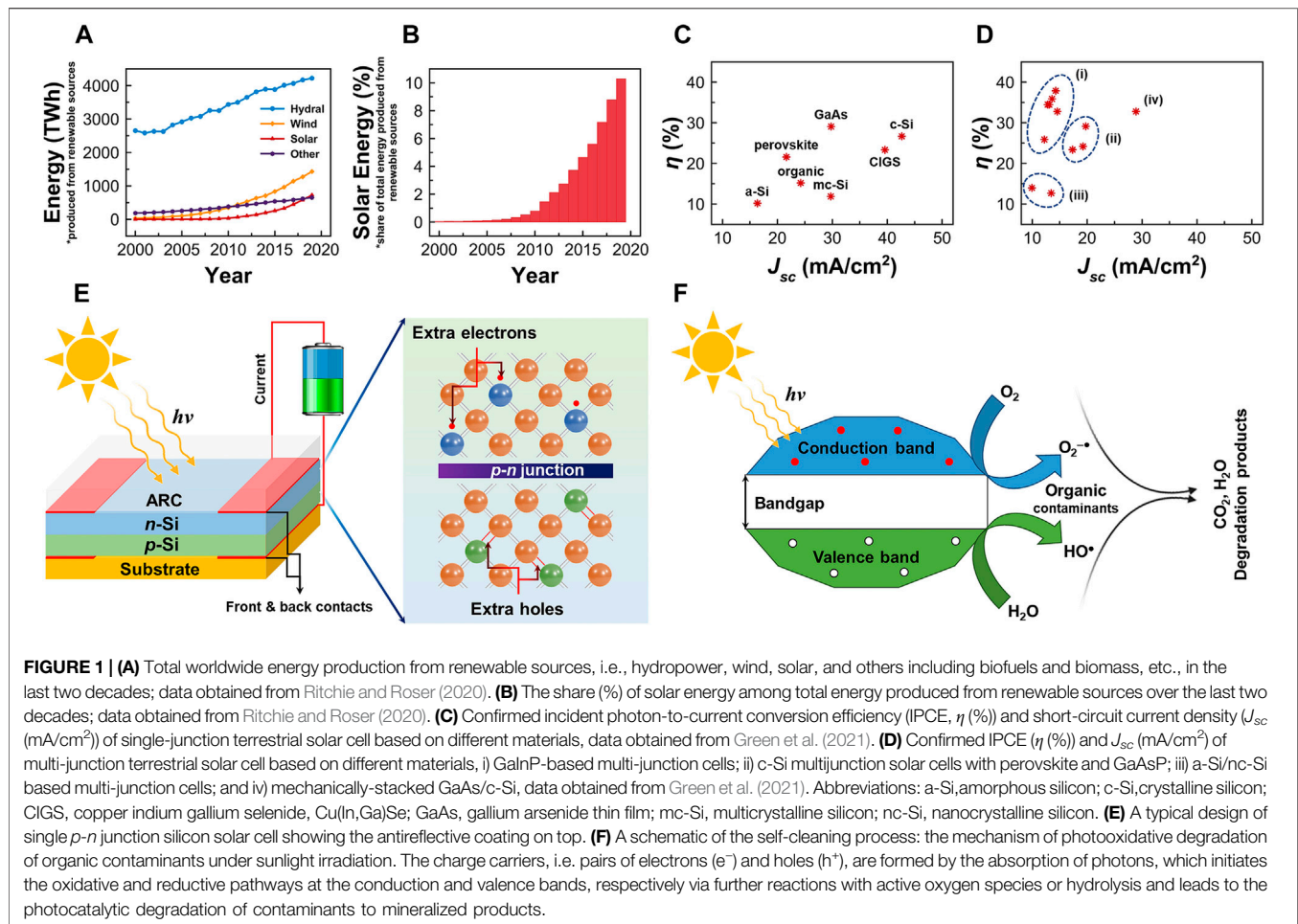
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INTRODUCTION

The environmental challenges such as global warming, air pollution, and climate change resulting from the burning of fossil fuels and growing awareness about these issues prompted the use of alternative and renewable energy sources. In the last two decades, solar energy has emerged as one such alternative with worldwide solar energy generation reaching 724 TWh in 2019 (Ritchie and Roser, 2020). Albeit it is a small share of the total worldwide energy generation from renewable sources, i.e., hydropower, solar, wind, geothermal, biomass, and biofuels (Figure 1A), solar energy share has grown from 0.04 in 2000 to 10.3% in 2019 (Figure 1B).

The rise in solar energy harvesting is backed by years of research and innovation starting from the development of silicon *p-n* junction solar cells with a significantly improved solar energy conversion efficiency in 1954 (Chapin et al., 1954; Loferski, 1993). Shortly after, silicon solar cells became commercially available (Stabler, 1960), and led the space solar cell industry for decades (Bailey and Raffaele, 2012). However, over the past two decades, several other materials and dual or multiple *p-n* junction cell designs with improved quantum efficiency have been commercialized for terrestrial applications (Green et al., 2021). Nowadays, the performance of these cells has greatly improved, see Figures 1C,D.



Yet, a major limiting factor for the quantum efficiency of solar cells is the optical loss due to reflection. For instance, in the air atmosphere, a polished silicon surface reflects >30% of the incident solar radiations (Thomas et al., 1989). Therefore, to reduce optical loss due to high reflectivity and improve the energy harvesting capability of solar cells, different solutions have been proposed such as the utilization of the antireflective coatings (ARCs), surface texturization, or a combination of both to increase the optical path length of the light within solar cells (Singh and Verma, 2019; Abu-Shamleh et al., 2021; Sagar and Rao, 2021).

Furthermore, in terrestrial sunlight-to-electricity generation applications, solar modules and panels are exposed to dust, organic contaminants, industrial pollutants, and harsh environmental conditions, e.g., sand storms (Said et al., 2015; Quan and Zhang, 2017), which also detriment their efficiency. The development of self-cleaning coatings is desired to address these issues (Soklič et al., 2015; Huang et al., 2018; Zhi and Zhang, 2018). Ideally, a coating material for terrestrial solar cell applications should be robust, transparent, antireflective, and capable of self-cleaning itself.

Titania (TiO₂) is an ideal material due to its excellent physicochemical properties and has been widely investigated in such applications, e.g., high-efficiency solar cells (O'Regan and Grätzel, 1991; Kim et al., 2008), ARCs (Wan et al., 2010; Manea et al., 2013; Salvaggio et al., 2016), and photocatalysis (Fujishima et al., 2000; Fujishima et al., 2008) especially photooxidative self-cleaning (Paz et al., 1995; Muraca et al., 2019). TiO₂ has a bandgap of 3.2 eV and absorbs light in a wide range of frequencies. Also, in photocatalysis, TiO₂ nanoparticles (P25) are considered as the photocatalytic standard.

Other oxides such as Ta₂O₅ (Thomas et al., 1989; Sagar and Rao, 2021) could offer better antireflective properties but are not good enough for photocatalytic degradation of environmental contaminants. Therefore, these oxides are often used in combination with TiO₂ to form self-cleaning ARCs. Herein, the role of self-cleaning ARCs based on TiO₂ nanostructures and nanocomposites in reducing the reflectivity and improving the overall energy harvesting capability is discussed. For high-efficiency solar cell applications, the referenced literature is critically analyzed with a particular emphasis on the scalability

of coating methods, industrial viability, cost-effectiveness, and commercialization viewpoints.

ANTIREFLECTIVE PROPERTIES OF TiO₂

TiO₂ Single-Layer Antireflective Coatings

Figure 1E presents the design of a typical single-junction silicon solar cell showing ARC as an integral part of the device. ARCs based on TiO₂ have been known to significantly reduce the solar averaged reflectance of the silicon solar cells (San Vicente et al., 2001; San Vicente et al., 2002). For instance, Cai et al. (2012) produced quasi-omnidirectional ARCs by direct hydrothermal fabrication of rutile-TiO₂ nanorod arrays on Ti-foils. These mesocrystalline TiO₂ nanorods suppressed the reflection to <2.0% in the near-infrared (NIR) and <0.5% in the visible regions. The excellent antireflection properties of TiO₂ nanorod arrays were attributed to the hierarchical tips-on-rod nanostructures that provided an optimal graded refractive index.

Consequently, TiO₂ ARCs with reduced reflectivity can enhance the quantum efficiency, or so-called incident photon-to-current conversion efficiency (IPCE). The researchers have shown that a compact TiO₂ layer (60–100 nm) on dye-sensitized solar cells can improve the IPCE by 50% (Abdullah and Rusop, 2013; Abdullah and Rusop, 2014). The enhancement in IPCE is attributed to the lower optical loss (67% decreased reflectivity), the red-shift of transmittance peak, i.e., greater energy absorption in the visible region, and reduced electron-hole recombination at the interface of TiO₂ (Abdullah and Rusop, 2013; Abdullah and Rusop, 2014).

To facilitate mass production and reduce the cost-to-efficiency ratio, atmospheric-pressure chemical vapor deposition (APCVD) is proposed (Hocine et al., 2013; Hocine et al., 2015). Thus-formed thin TiO₂ ARCs exhibit a 75% decrease in reflectivity and a 27% increase in efficiency of multicrystalline silicon solar cells, which is attributed to the enhanced light transmission by TiO₂ ARC and increased short-circuit current (Hocine et al., 2013). Leem et al. (2014) produced ARCs composed of TiO₂ subwavelength nanostructures through electron beam evaporation and dry etching and observed 33% enhancement in IPCE of GaAs solar cells.

TiO₂-Containing Multi-Layer Antireflective Coatings

Double-layer AR (DLAR) or triple-layer AR (TLAR) coatings further reduce the reflectivity and have long been adopted to boost solar cell efficiency (Jiao and Anderson, 1987; Aiken, 2000; Zhang et al., 2006). Compared to commercial single-layer TiO₂ coatings, DLAR from single-material TiO₂ reduces the solar weighted average reflectance and improves the efficiency of silicon solar cells (Richards, 2003). A notable achievement in this regard was reported by Lien et al. (2006), who deposited TiO₂ SLAR, SiO₂/TiO₂ DLAR, and SiO₂/SiO₂-TiO₂/TiO₂ TLAR coatings on silicon solar cells via a sol-gel process. They observed a 39.5% increase in solar cell efficiency with SiO₂/SiO₂-TiO₂/TiO₂ TLAR coatings.

Liu et al. (2014) also obtained ~32% enhancement in short-circuit current and IPCE of GaAs multi-junction solar cells with SiO₂/graded-index TiO₂ DLAR top coatings. There are several other reports in favor of reduced broadband reflectance exhibited by DLAR or TLAR coatings based on TiO₂ and SiO₂ (Ali et al., 2014; Li et al., 2014; Lin et al., 2018; Liao et al., 2020; Yuan et al., 2020). Recently, Zhang et al. (2021) tested TiO₂/Al₂O₃ DLAR and TiO₂/Al₂O₃/MgF₂ TLAR coatings for GaInP/InGaAs/Ge triple-junction solar cells. They reported a respective decrease of 18.9 and 22.2% in reflectivity, while an increase of 21.29 and 29.44% in IPCE.

Nonetheless, irrespective of the ARC material, composition, fabrication procedure, cell type, design, etc., a minimum of 8.5% (Li et al., 2014) to 54.5% (Ho et al., 2015) enhancements in IPCE have been reported. Therefore, DLAR or TLAR coatings can be utilized to improve the overall performance of different types of solar cells provided they do not require multiple fabrication steps and do not add to their complexity.

PHOTOOXIDATIVE SELF-CLEANING PERFORMANCE OF TiO₂

The self-cleaning ARCs are desirable for terrestrial solar cell application to withstand harsh environments, air pollution, and organic contaminations (San Vicente et al., 2012). Due to its excellent photocatalytic properties, TiO₂ is widely employed in self-cleaning ARCs for solar cell cover glass and solar panels, self-cleaning windows, indoor air purification systems, sterilization of water, degradation of organic contaminants, and antibacterial applications (Faustini et al., 2010; Banerjee et al., 2015). **Figure 1F** illustrates the mechanism of photocatalytic decomposition of organic contaminants on TiO₂ surface under sunlight irradiation. Photooxidative properties of commercially available TiO₂ particles are generally demonstrated by a conventional dye-degradation experiment under UV illumination (Nakata et al., 2011).

Efforts to move from UV to visible light and efficiently utilize solar energy with TiO₂ are still underway. To achieve this goal, TiO₂ is often doped or mixed with different materials, e.g., non-metals, metals, metallic oxides, etc., for better photocatalytic performance under direct sunlight (Medina-Valtierra et al., 2009; Li and He, 2013; Suárez et al., 2017). For example, TiO₂ films doped with 5% Ni show an increased light transmission in the visible and NIR regions and lower reflectance (Haider et al., 2016). V-doped TiO₂:SiO₂ sol-gel ARCs with 15% V-TiO₂ content also demonstrate low refractive index and excellent photooxidative degradation of methylene blue ($k = 0.033 \text{ min}^{-1}$) (Adak et al., 2017).

TiO₂ doping with metal atoms such as V alters its electronic structure and influences its photoactivity. V atoms may act as traps for photogenerated electrons and holes and reduce their recombination (Adak et al., 2017). The electrons from V⁴⁺ centers are photoexcited to the TiO₂ conduction band, while the resulting V⁵⁺ centers bind with surface oxygen atoms. Consequently, •OH free radicals are formed in the presence of holes (Klosek and Raftery, 2002). Additionally, superoxide-free radicals (O₂^{•-}) are

produced by the reduction of O_2 on TiO_2 surface, which are strong oxidants for organic contaminants.

Therefore, V-doped TiO_2 improves photooxidative decomposition of methylene blue under visible-light illumination (Adak et al., 2017). A similar phenomenon is observed by Naufal et al. (2017) with 2% Sm^{3+} -doped TiO_2 , which exhibits a highly defective surface due to oxygen deficiency and significantly improves photocatalytic oxidation of methylene blue ($k = 0.103 \text{ min}^{-1}$) under sunlight illumination.

Furthermore, graphene or reduced-graphene oxide (rGO) have rarely been incorporated in TiO_2 ARCs despite their excellent electrical properties. A recent study reveals that rGO- TiO_2 surfaces are an outstanding alternative for photooxidative self-cleaning applications (Prabhu et al., 2017). This is supported by a sharp increase in photocurrent response of 0.5 wt% rGO- TiO_2 upon illumination, which suggests the greater separation of photogenerated electrons and holes. Being a zero-bandgap conductor, rGO promotes the transfer of photogenerated electrons and reduces the recombination rate (Wang et al., 2013; Qiu et al., 2015). Therefore, rGO- TiO_2 could act as a potential self-cleaning ARC in energy harvesting devices.

PHYSICOCHEMICAL STRATEGIES FOR IMPROVED TiO_2 Antireflection and Self-Cleaning Properties

In general, two strategies are used to design ARCs (Keshavarz Hedayati and Elbahri, 2016), the conventional stacking layers (single or multiple layers) and ultrathin plasmonic metasurfaces. However, the works reviewed herein only involve conventional single- or multi-layer ARC for solar cells and modules. The formation of DLAR or TLAR coatings enhances the efficiency of solar cells by suppressing reflectivity and improving the short-circuit current. However, several other physicochemical strategies have been adopted for enhanced performance of TiO_2 ARCs. These include the inclusion of a porogen (pore-forming agent) and chemical acid-etching with 5% HF that increase the porosity and surface roughness, respectively, and reduce reflectivity (Rad et al., 2020).

It is believed that the formation of 3D nanoporous structures may achieve zero-reflectance conditions (Bravo et al., 2007; Guldin et al., 2013). For instance, Nakata et al. (2011) concluded that the self-cleaning effect and high transmittance of a thick layer of TiO_2 particles are derived from its porous structure. In this regard, an innovative way to produce highly porous ARC involves the sol-gel synthesis of TiO_2 nanoparticles combined with block-copolymer self-assembly that yields excellent antireflective and photocatalytic properties (Guldin et al., 2013).

The functionalization of TiO_2 surface with silane coupling agents, i.e., tetraethoxysilane and methyltriethoxysilane, is shown to decrease the reflectance of bare- TiO_2 films from 26 to 17.3% and 15.5%, respectively (Purcar et al., 2019). The surface modification of TiO_2 photocatalyst and $SiO_2/TiO_2/SiO_2-TiO_2$ TLAR with fluoroalkylsilane and hexamethyldisilazane, respectively, promotes hydrophobicity and self-cleaning

(Nakajima et al., 2000; Ye et al., 2013). Similarly, metal-doped TiO_2 ARCs are shown to alter the bandgap, increase solar energy absorption and conversion, and improve self-cleaning properties (Haider et al., 2016; Adak et al., 2017).

Another strategy involves metal plasma ion implantation of various metal ions (e.g., Fe, Cr, V) in high-quality TiO_2 films, which activates the TiO_2 surface and improves the sunlight absorption rate (Weng and Huang, 2013). The implantation of Fe^{3+} is particularly observed to decrease the recombination of photogenerated holes and electrons and increase oxygen vacancies (Yu et al., 2006; Weng and Huang, 2013). This also leads to dissociative adsorption of water molecules, which in turn increases surface hydroxyl content and self-cleaning ability of TiO_2 .

A photocatalytic ARC using a heterojunction structure between low bandgap WO_3 (2.8 eV) and higher bandgap TiO_2 (3.2 eV) nanoparticles is developed by Noh and Myong (2014). WO_3 nanoparticles not only generate a self-cleaning effect but act as visible-light sensitizers and transfer the excited electrons to higher bandgap TiO_2 . Thus, WO_3-TiO_2 ARC shows 4.3% higher energy output for silicon-based photovoltaic modules (Noh and Myong, 2014).

Recently, vertically aligned Pt-decorated $p-MoS_2$ nanostructures are fabricated on $n-TiO_2$ nanotube arrays to benefit from Pt/ MoS_2 Schottky heterojunction and MoS_2/TiO_2 $p-n$ heterojunction for visible-light degradation of methylene blue (Dong et al., 2020). These innovations are particularly helpful in understanding the underlying mechanisms to improve the efficiency and lifetime of solar energy harvesting devices. However, there is a need to overcome several practical challenges for large-scale production and application of these devices, as discussed below.

NOTABLE ACHIEVEMENTS AND PRACTICAL CHALLENGES

Table 1 presents notable achievements and selected examples to provide an overview of the impact of TiO_2 ARCs on enhancing antireflectivity and efficiency. TiO_2 is an n -type semiconductor due to inherent oxygen vacancies (Nowotny et al., 2006). Therefore, TiO_2 layers help electron injection and ease electron transport by providing an electrical path for the photogenerated electrons (Chong et al., 2005; Dong et al., 2010). In solar cell applications, $n-TiO_2$ ARC deposited on $p-Si$ (**Figure 1E**) combines to form a heterojunction, thereby allowing one type of charge carriers (electrons) and blocking holes to improve the efficiency of the solar cells (Avasthi et al., 2013; Singh et al., 2017). TiO_2 ARCs can also get rid of the environmental contaminants through photooxidative degradation in the presence of sunlight (Naufal et al., 2017).

In addition to its self-cleaning properties, the hardness of TiO_2 ARC makes it a scratch-resistant shielding cover for solar cells and prevents mechanical damage (Wang et al., 2015; Mazur et al., 2016). An ultrathin ($\sim 23 \text{ nm}$) layer of nanoporous TiO_2 deposited on SiO_2-TiO_2 ARC not only ensures self-cleaning but acts as a protective barrier against abrasion and

TABLE 1 | Notable achievements in the development of TiO₂-based antireflective coatings (ARCs) and figures of merit showing the resulting enhancement in solar cells' performance.

Cell type	Coating method	ARC material(s)	ARC thickness (nm)	Reflectivity measurement		Cell characteristics				Ref
				Range, λ (nm)	Reflectance ^a (%)	J_{sc} (mA/cm ²)	V_{oc} (V)	FF (%)	η (%)	
Dye-sensitized solar cell (ITO/p-TiO ₂)	Without ARC	TiO ₂	60	-	-	6.17	0.662	6.17	2.30	Abdullah and Rusop (2013)
	RF magnetron sputtering				-	8.93	0.669	0.58	3.44	
Multicrystalline silicon solar cells	Without ARC	TiO ₂	60	300–1,150	35.0	28.63	0.561	70	11.24	Hocine et al. (2013)
GaAs solar cell	APCVD				8.6	33.86	0.585	72	14.26	
	Without ARC	TiO ₂ (SWS)	~63	350–900	>40	18.38	1.011	79.22	14.74	Leem et al. (2014)
	Electron beam evaporation and dry etching				9.5	23.65	1.012	79.46	18.98	
					6.2	24.82	1.012	78.42	19.66	
Monocrystalline silicon solar cell	Without ARC	TiO ₂ (DLAR)	56.8	400–1,000	37.0	16.2	0.61	76.4	11.36	Lien et al. (2006)
	Sol-gel process and dip coating				9.3	24.8	0.61	76.2	14.49	
					6.2	25.8	0.61	76.6	14.99	
					3.2	27.1	0.61	76.7	15.85	
Silicon solar cell	Without ARC	TiO ₂ (DLAR)	~100	400–900	36.0	24.8	0.62	79.1	12.2	Chen et al. (2012)
	DC reactive magnetron sputtering				10.3	34.2	0.63	77.9	16.8	
					3.7	37.2	0.63	78.2	18.4	
Monocrystalline silicon solar cell	Without ARC	SiO ₂ (DLAR)	81.1	400–1,000	35	9.24	0.442	69	2.8	Ali et al. (2014)
	RF magnetron sputtering				15	12.35	0.504	72	4.5	
					7	16.13	0.520	75	6.2	
CIGS: Cu(In,Ga)Se ₂ solar cell	Without ARC	TiO ₂ -SiO ₂	Six stacks of	250–2,500	-	33.36	0.510	65.85	11.20	Li et al. (2014)
	Sol-gel process and dip coating				-	35.12	0.521	66.40	12.15	
Crystalline silicon solar cell	Without ARC	TiO ₂	20	400–1,050	-	26.10	0.54	-	10.96	Ho et al. (2015)
	Electron beam evaporation				-	30.38	0.55	-	12.84	
					-	32.16	0.55	-	13.69	
					-	39.89	0.55	-	16.93	

^aThe solar weighted average reflectance (%) as reported in the relevant literature. APCVD, atmospheric pressure chemical vapor deposition; ARC, antireflective coating; DLAR, double-layer antireflective coating; ITO, indium tin oxide; PECVD, plasma-enhanced chemical vapor deposition; p-TiO₂, porous TiO₂; RF, radio-frequency; SLAR, single-layer antireflective coating; SWS, subwavelength structures; TLAR, triple-layer antireflective coating.

mechanical damage, thus providing high chemical and mechanical durability (Miao et al., 2013).

A temperature and humidity stress test reveals that DLAR with a denser TiO₂/ZrO₂ barrier layer is more durable showing only 1% reduction in transmittance after 96 h (Li et al., 2013). Albeit the persistence of optical activity over a long period is not usually tested, a few cases reported in the literature reveal that TiO₂ ARCs resist aging and maintain low reflectivity ($\leq 1\%$) after 1,200 h of testing (Xu et al., 2013).

However, there are several practical challenges associated with the fabrication of ARCs. Considering the commercialization perspective and industrial-scale fabrication of ARCs, it is important to maintain the cost-efficiency balance and ease in the fabrication of uniform TiO₂ ARCs. For solar cells and modules, self-cleaning ARCs are generally deposited by vacuum methods such as physical/chemical vapor deposition, sputtering, and thermal evaporation. These methods are

reproducible and yield extremely stable ARCs with the controlled thickness (Shahiduzzaman et al., 2017; Pae et al., 2018). However, these are expensive and are less versatile.

The sol-gel process is an alternative wet-chemical route that has been largely studied for the fabrication of self-cleaning ARCs (San Vicente et al., 2001; San Vicente et al., 2002). Compared to vacuum processes, the sol-gel method is low-cost and offers greater flexibility in the choice of raw materials, solvents, molar ratios, and experimental conditions, e.g., pH, temperature, etc. (Wang and Shen, 2010). However, the sol-gel process can have reproducibility issues along with a possibility of poor adhesion and mechanical strength due to excessive solvent evaporation. To improve mechanical properties and avoid abrasion damage, post-deposition heat treatment is necessary (Langlet et al., 2001), which may increase cost and fabrication time/steps. Also, the coating procedure for sol-gel materials (dip- or spin-coating) is tedious and irreplicable.

Similarly, extremely high gains in efficiency of solar cells have been reported with innovative ARC-fabrication techniques such as electron beam evaporation and dry-etching (Leem et al., 2014). The fabrication of plasma ion-implanted or vertically-aligned composite nanostructures is also fascinating and delivers excellent results (Weng and Huang, 2013; Dong et al., 2020). However, these methods are less suitable for mass production at the industrial scale due to inherent reproducibility issues and an increased number of steps involved and cost. Therefore, an important aspect of future research is to find an optimal cost-to-efficiency balance through innovative yet simple, scalable, reproducible, and cost-effective methods for the fabrication of TiO₂ ARCs.

CONCLUDING REMARKS

The quantum efficiency of terrestrial solar cells is fundamentally restricted to approximately 30% due to high reflectivity, i.e., nearly 35% (Diedenhofen et al., 2012; Sheng et al., 2013), albeit higher efficiencies are theoretically and practically achievable with multi-junction solar cells (Pakhanov et al., 2018; Geisz et al., 2020). The optical loss due to reflectivity and environmental conditions can be countered through self-cleaning ARCs to achieve the true potential. TiO₂ is a choice of material that can simultaneously perform both functions: antireflectivity and self-cleaning. Overall, the properties of TiO₂ ARCs make them ideal candidates for high-efficiency, long-life solar cell production, and commercialization. However, there is a need to design scalable and cost-effective approaches for their fabrication.

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DATA AVAILABILITY STATEMENT

All relevant data presented in this study are included in the article, further inquiries can be directed to the corresponding author.

AUTHOR CONTRIBUTIONS

AA conceived the idea, secured funding, administered the project, performed a literature survey and analyses, written the first draft, reviewed and edited the manuscript. AH, IU, and MS are co-investigators and helped in literature survey, review, and editing. AR reviewed and edited the work. The authors have read this work and approved it for publication.

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Conflict of Interest: The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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