

Defect, Morphology, and Interface Engineering of TiO₂ in Dye Sensitized Solar Cells: Recent Progress and Perspectives

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Abstract

Dye-sensitized solar cells (DSSCs) remain promising low-cost photovoltaic technologies because of their simple fabrication, tunable optical properties, and effective operation under low-light conditions. Titanium dioxide (TiO₂) is the most widely used photoanode material in DSSCs owing to its chemical stability, suitable band alignment, low toxicity, and excellent transparency. However, the photovoltaic performance and long-term stability of TiO₂-based DSSCs are still limited by charge recombination, slow electron transport, interfacial losses, and structural degradation. This review summarizes recent advances in defect engineering, morphology engineering, and interface engineering of TiO₂ photoanodes for high-performance DSSCs. Attention is given to the role of oxygen vacancies, Ti³⁺ states, metal/non-metal doping, and heterostructure formation in tailoring the electronic structure and charge transport behavior of TiO₂. The influence of various TiO₂ nanostructures, including nanoparticles, nanotubes, nanorods, nanosheets, and hierarchical architectures, on dye adsorption, light scattering, electron mobility, and recombination dynamics is critically discussed. Furthermore, recent progress in interface engineering strategies such as passivation layers, blocking layers, MXene incorporation, composite photoanodes, and atomic layer deposition are examined in relation to interfacial charge transfer and device stability. Current challenges involving defect-induced recombination, morphology-related transport trade-offs, and long-term degradation are also analyzed. Finally, future perspectives on hierarchical nanoarchitectures, multifunctional interfaces, flexible DSSCs, and hybrid TiO₂ systems are presented. This review provides an integrated understanding of how defect, morphology, and interface engineering collectively govern the performance of TiO₂ photoanodes and offers design guidelines for next-generation high-efficiency and stable DSSCs.

Keywords: titanium dioxide; photoanodes; dye-sensitized solar cells; defects; interface engineering; morphology



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1. Introduction

In dye-sensitized solar cells (DSSCs), the photoanode semiconductor is the primary functional element that controls the processes of charge transfer, electron injection, and light harvesting, all of which eventually affect the device's performance [1]. Michael Grätzel and Brian O'Regan's 1991 research on photoinduced electron injection from dye molecules into a mesoporous TiO₂ network established titanium dioxide (TiO₂) as the

primary semiconductor in dye-sensitized solar cells (DSSCs) [2,3]. TiO_2 is found in the following three phases: anatase, rutile, and brookite. Its anatase phase is favored in DSSCs because of its greater surface area, optimal band alignment, and improved electron transport capabilities [4,5]. Since the pioneering work of Yang et al. and Yu et al. on exposed {001} anatase facets, recent studies have focused on facet-stabilization strategies, fluorine-free synthesis, facet-dependent charge transport, and heterointerface engineering for DSSCs and related photoelectrochemical systems [6,7].

The semiconductor produces a nanocrystalline, mesoporous structure, allowing for high dye loading while also promoting electron percolation through linked particles. The broad bandgap (~ 3.2 eV for anatase) enables transparency in the visible area, allowing light absorption to be controlled by sensitizing dyes rather than the semiconductor itself. TiO_2 has significantly impacted DSSCs, enabling the creation of cost-effective, scalable, and efficient photovoltaic devices for indoor- and low-light applications [6–12].

Titanium dioxide (TiO_2) has continued to be the most popular photoanode material among the other semiconductor materials investigated because of its good conduction band alignment with the majority of sensitizing dyes, excellent chemical and photochemical stability, cheap cost, and environmental friendliness [13–15]. Recent research according to Salim et al. (2023) [16] has shown that alterations to TiO_2 , including defect engineering, heteroatom doping, and nanostructure design, may greatly improve charge transfer and inhibit recombination processes [4]. As illustrated in Figure 1, upon light absorption, electrons are excited from the highest occupied molecular orbital (HOMO) to the lowest unoccupied molecular orbital (LUMO) of the dye molecule [16–18]. For efficient electron injection, the LUMO energy level of the excited dye must be positioned above the conduction band edge of TiO_2 , enabling spontaneous transfer of electrons into the semiconductor [19]. The injected electrons are subsequently transported through the TiO_2 network and collected by the transparent conducting substrate before flowing through the external circuit [20].

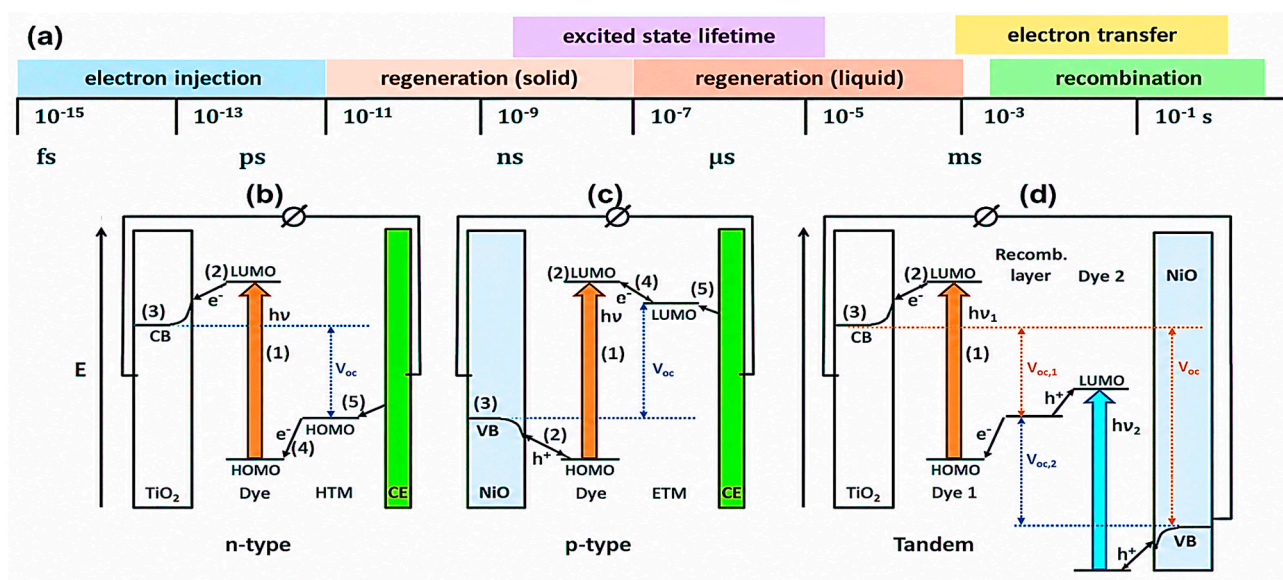


Figure 1. Schematic illustration of the operating principle of dye-sensitized solar cells, showing (a) an illustration of the charge transfer kinetics in ss-DSSCs regarding time, (b) charge transfer occurring in an n-type ss-DSSC, (c) charge transfer occurring in a p-type ss-DSSC and (d) charge transfer occurring in a tandem p-n-DSSC [21].

Given that it functions as the interface for electron injection, the dye adsorption scaffold, and the electron transport medium all at once in a typical DSSC setup, the TiO_2 photoanode is crucial to determining the photovoltaic performance [22]. As a result, charge

production, transport kinetics, and recombination processes inside the cell are impacted by the structural and electrical characteristics of TiO_2 . Fluorine-doped tin oxide (FTO) and other conductive substrates are coated with nanocrystalline mesoporous TiO_2 films in conventional DSSCs [23]. Mesoporous TiO_2 's wide surface area allows for high dye loading, which enables effective electron injection and light harvesting; this is illustratively shown in Figure 2 [24]. But the high density of grain boundaries and defect states created by the nanoparticulate structure can also serve as recombination hotspots, decreasing electron lifespan and overall device efficiency. Consequently, a significant amount of research has been focused on altering the physicochemical characteristics of TiO_2 by means of defect engineering, morphological optimization, and interface modification techniques [25].

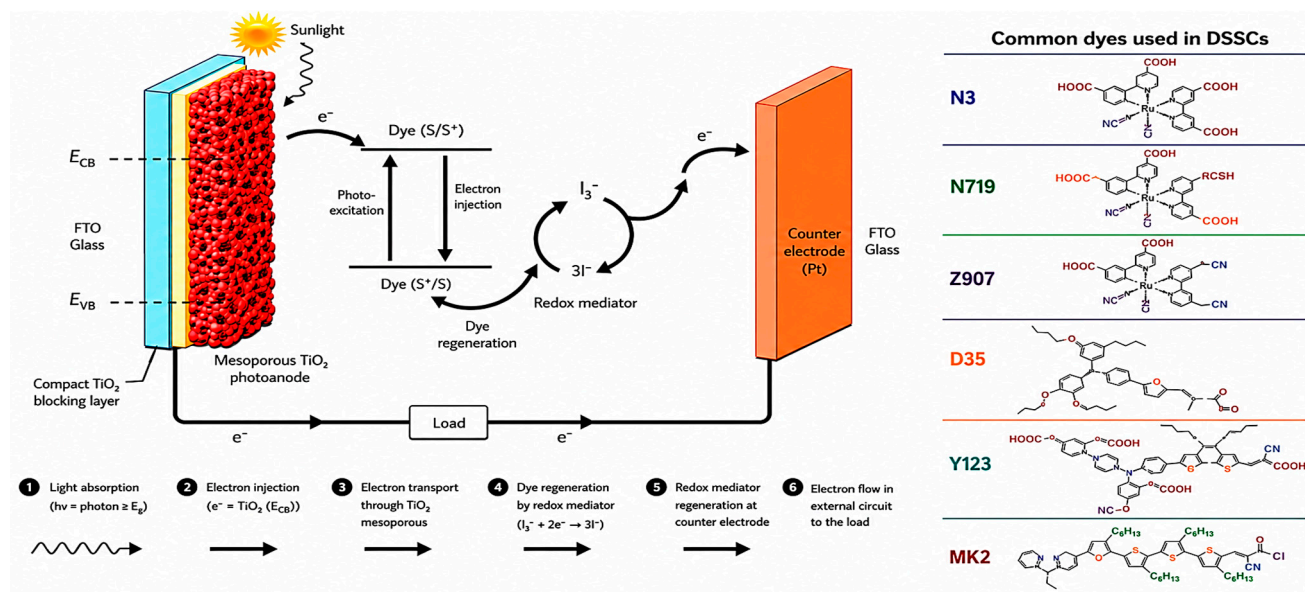


Figure 2. Overview of the DSSC device architecture and photovoltaic operation, showing energy conversion from incident sunlight to electrical output through coordinated interfacial electron-transfer, redox-regeneration processes and common organic and inorganic dyes used in DSSCs [original drawing].

In this review, defect engineering refers to intentional modification of the TiO_2 electronic defect landscape through oxygen vacancy control, Ti^{3+} generation, elemental doping, and lattice disorder manipulation to regulate carrier concentration, trap-state density, and band structure [23].

Morphology engineering refers to the structural tailoring of TiO_2 across multiple dimensionalities (0D, 1D, 2D, and hierarchical architectures) to optimize surface area, electron transport pathways, electrolyte diffusion, and light scattering [26,27].

Interface engineering refers specifically to modification of charge-transfer interfaces in DSSCs, including TiO_2 /dye, TiO_2 /electrolyte, and TiO_2 /FTO interfaces, using passivation layers, heterostructures, ultrathin coatings, and interfacial compositing strategies to suppress recombination and improve carrier extraction [28].

Although research articles have discussed individual aspects of TiO_2 modification in DSSCs, most studies primarily focus on either defect engineering, morphology control, or interfacial modification independently. However, these approaches are strongly interconnected because defect states influence charge transport kinetics, morphology determines electron diffusion pathways and dye adsorption, while interfacial properties govern recombination dynamics and long-term device stability. A comprehensive understanding of the synergistic relationship among these engineering strategies is, therefore, necessary for the rational design of next-generation TiO_2 photoanodes [29–31].

2. Recent Progress in Defect, Morphology, and Interface Engineering of TiO₂ Photoanodes

2.1. Defect Engineering of Titanium Dioxide Semiconductor

In dye-sensitized solar cells (DSSCs), defect engineering has become a crucial tactic for improving the electrical structure and charge transport characteristics of titanium dioxide (TiO₂) photoanodes [32]. Charge carrier dynamics can be impacted by intrinsic flaws within TiO₂'s crystal structure, despite the material's advantageous properties, including chemical stability, high transparency, and appropriate band alignment with several sensitizing dyes. These defects, which are mostly oxygen vacancies, Ti³⁺ states, and impurity dopants, can either increase electron mobility and conductivity or function as recombination sites that lower device efficiency [33]. As a result, contemporary DSSC research has placed a strong emphasis on precisely controlling defect generation. Enhancing electron transport in TiO₂ has been extensively studied by intrinsic defect management, namely, the control of oxygen vacancies. Within the bandgap, oxygen vacancies provide localized energy states that can boost conductivity and carrier concentration.

Enhancing electron transport in TiO₂ has been extensively studied by intrinsic defect management, namely, the control of oxygen vacancies. Within the bandgap, oxygen vacancies provide localized energy states that can boost conductivity and carrier concentration. However, trap states that encourage electron-hole recombination may also be produced by high vacancy concentrations [34]. Therefore, to balance increased conductivity with reduced recombination losses, recent research has focused on controlled defect creation using thermal treatments, chemical reduction, or surface passivation techniques. For example, it has been demonstrated that surface passivation layers like MgO or WO₃ minimize recombination at defect locations while preserving effective electron injection from the dye to the TiO₂ conduction band, resulting in increased carrier lifetime and decreased charge transfer resistance in DSSCs [20]. Doping TiO₂ with metal and non-metal elements is another common defect engineering technique. TiO₂'s electronic structure may be altered by substitutional dopants, which can also produce shallow donor states and enhance visible light absorption. By enhancing electron transport and reducing recombination processes at the photoanode interface, Fe/S co-doped TiO₂ nanoparticles have been shown to dramatically improve photovoltaic performance. The enhanced performance was ascribed to the synergistic effects of sulphur and iron dopants, which modify the TiO₂ band structure and promote charge separation in the photoanode material [35]. Figure 3 shows the improved BET and charge transfer capacitance of such doping.

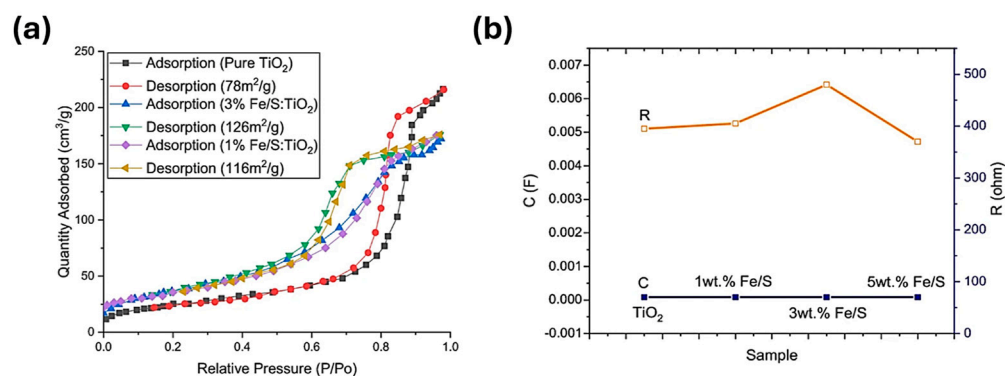


Figure 3. (a) BET of pure and wt.% Fe/S-TiO₂ (b) variation in charge transfer capacitance and polarization capacitance with Fe/S content. Adapted from [35].

In a comparable manner, co-doping techniques using rare-earth and alkaline metals have gained popularity. When compared to undoped TiO₂ systems, magnesium and

lanthanum co-doping in $\text{TiO}_2\text{-ZrO}_2$ composite photoanodes has been shown to improve crystallinity, optical absorption, and charge transport properties, leading to increased DSSC efficiency. The grain size increased with a 1.2% La dopant. This implies that more La incorporation causes the TZ to crystallize, giving the doped materials larger crystallite sizes. The crystallite size (D) was determined using Scherrer's equation, Equations (1) and (3):

$$D = \frac{(0.9\lambda)}{\beta \cos \theta} \quad (1)$$

The dislocation density (δ) and the d-space are computed as follows to obtain more information on the structural properties of the suggested PA samples, Equation (2):

$$\delta = \frac{1}{D^2} \quad (2)$$

$$D = \frac{\lambda}{2 \sin \theta} \quad (3)$$

where D is the crystalline size, β is the XRD peak FWHM appearing at the diffraction angle (θ), and λ is the X-ray wavelength ($=1.54056 \text{ \AA}$).

For the TZ, TZM4, TZM8, and TZM12 samples, the computed D values are, respectively, 21.16 nm, 59.04 nm, 52.57 nm, and 55.80 nm. The addition of magnesium causes these readings to rise. One explanation for the increase in grain size with Mg inclusion is a decrease in dislocation density, which results in fewer crystal lattice defects. A large D and tiny δ show that the films have more crystallization. Reduced trap-mediated recombination inside the TiO_2 lattice and changes in the electronic density of states are the causes of these improvements [8,15].

Defect engineering through semiconductor composite synthesis has also been studied recently. By adding advantageous defect states and changing band orientation, secondary oxides can enhance interfacial charge-transfer mechanisms in TiO_2 . Fe_2O_3 -modified TiO_2 photoanodes, for instance, have shown increased electron transport and higher light absorption because of Fermi-level tuning and lower recombination rates. Figures 4 and 5 clearly prove that defect engineering with Fe_2O_3 reflectance, absorbance, and current density. These compositional changes increase the range of spectrum absorption and facilitate effective charge separation, improving DSSC photovoltaic performance [36,37].

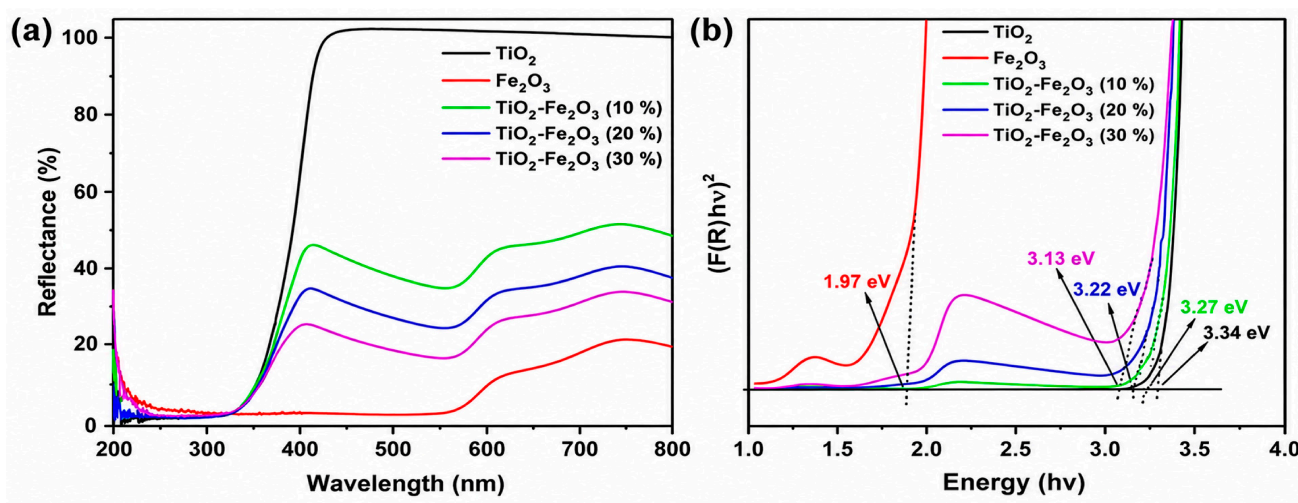


Figure 4. (a) UV-Vis diffuse reflectance spectra (DRS) of pristine TiO_2 , pristine Fe_2O_3 , and $\text{TiO}_2\text{-Fe}_2\text{O}_3$ (10, 20, and 30% wt %); (b) corresponding Tauc plot used to estimate the optical band gaps. The progressive redshift and bandgap narrowing with increasing Fe_2O_3 content are highlighted. Adapted from [37].

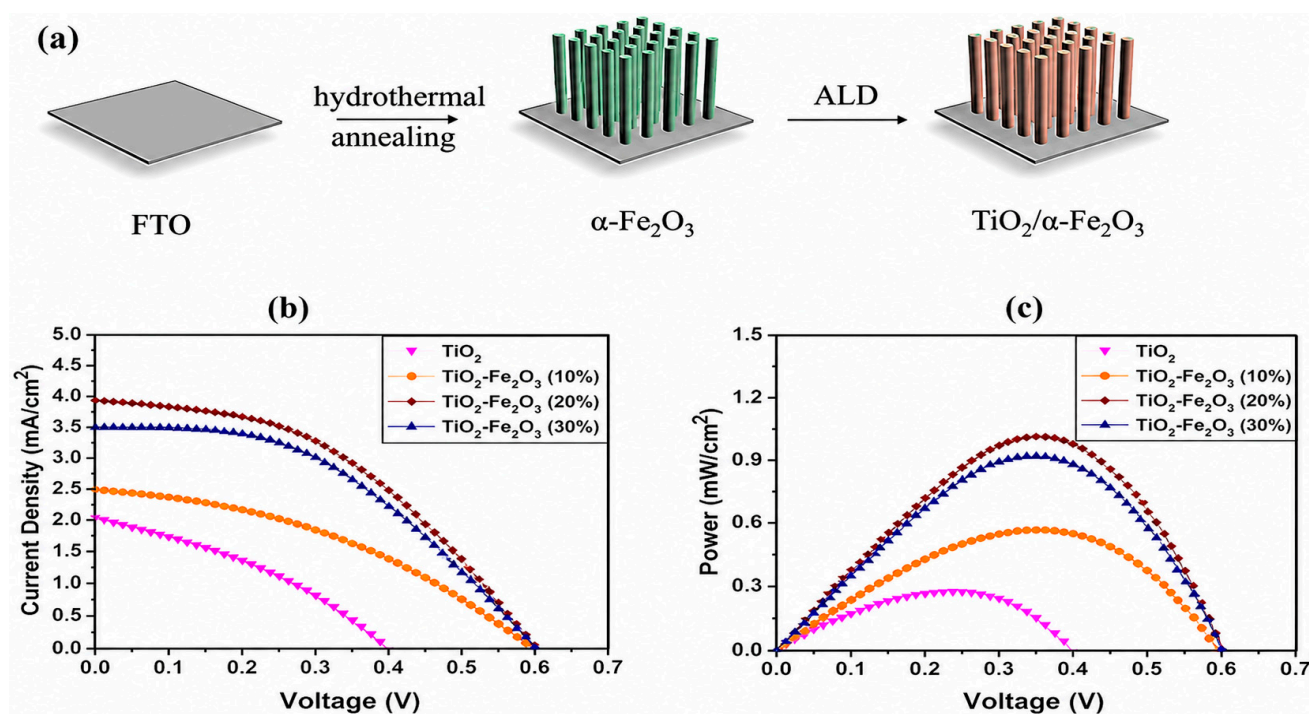


Figure 5. (a) Schematic illustration of the growth evolution of Fe₂O₃/TiO₂ nanorod films on FTO substrates [38]; (b) current density of pristine TiO₂, pristine Fe₂O₃, and TiO₂-Fe₂O₃ (10, 20, and 30% wt %); (c) power voltage of pristine TiO₂, pristine Fe₂O₃, and TiO₂-Fe₂O₃ (10, 20, and 30% wt %) [37].

Surface defect passivation has emerged as a successful tactic for reducing recombination losses at the semiconductor–electrolyte interface in addition to compositional changes. Recombination with oxidized redox species in the electrolyte is typically facilitated by surface states and structural flaws acting as electron trapping sites. By forming energy barriers that stop reverse electron transfer while maintaining effective electron injection from the dye molecules, thin passivation layers or chemical surface treatments can reduce these recombination routes. According to recent research, by lowering charge transfer resistance and stabilizing the photoanode surface, improved passivation layers can dramatically extend electron lifespan and improve overall DSSC efficiency [5,18].

2.2. Morphology Engineering of Titanium Dioxide Photoanodes

One of the best methods for enhancing the efficiency of dye-sensitized solar cells (DSSCs) is morphology engineering of TiO₂ photoanodes. Dye adsorption, light scattering, electron transport, and recombination dynamics are among the crucial activities in DSSCs that are controlled by the morphology of TiO₂. This review focuses primarily on representative TiO₂ nanostructures that have demonstrated the greatest relevance to DSSC performance enhancement, including nanoparticles, one-dimensional nanostructures, nanosheets, and hierarchical architectures, because these morphologies dominate current DSSC research and provide the most effective balance between dye loading, electron transport, light scattering, and recombination suppression [39,40]. The morphology categories of various used TiO₂ semiconducting materials are displayed in Figure 6 [41].

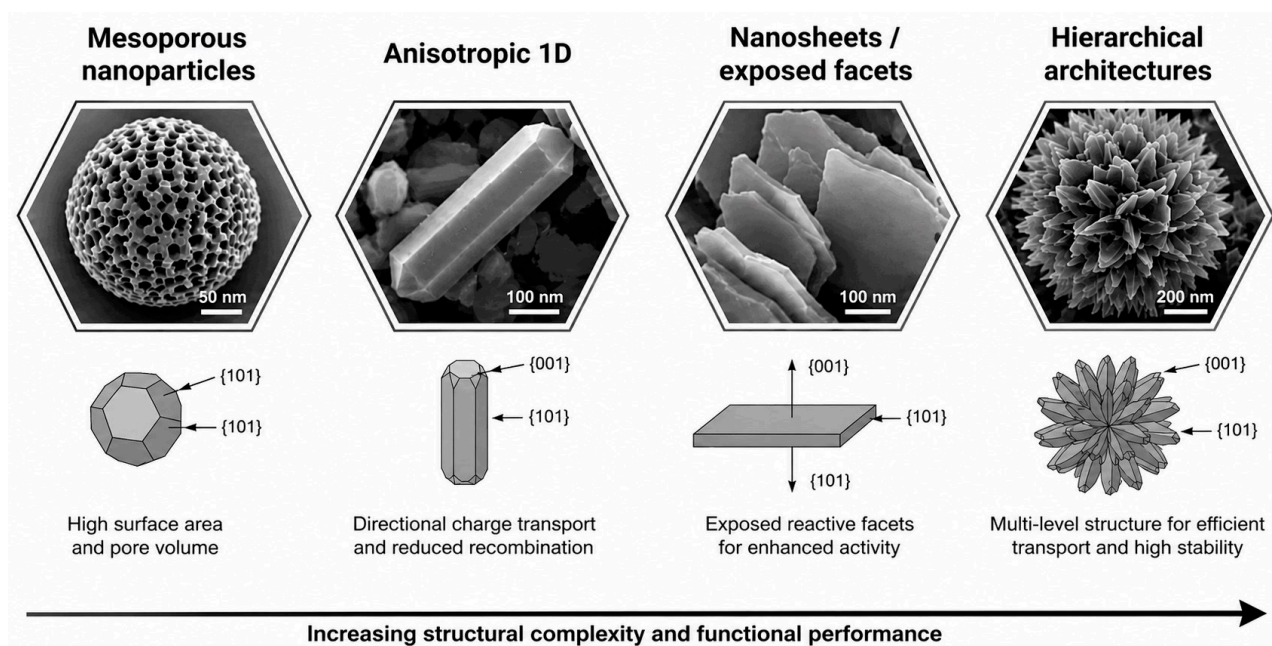


Figure 6. Schematic illustration of the most used TiO_2 morphologies (0D: NPs and nanocrystals; 1D: nanotubes and nanowires; 2D: nanosheets and exposed facets; 3D: hierarchical architectures) [original drawing].

2.2.1. Nanoparticle-Based Mesoporous Films

The most popular photoanode material in DSSCs is still zero-dimensional (0D) TiO_2 nanoparticles because of their great dye-adsorption capacity and large specific surface area. Mesoporous nanoparticle films provide strong photocurrent production and effective photon absorption by giving dye molecules plenty of active sites [41]. Figure 7 shows a typical SEM image of a 0D TiO_2 electrode.

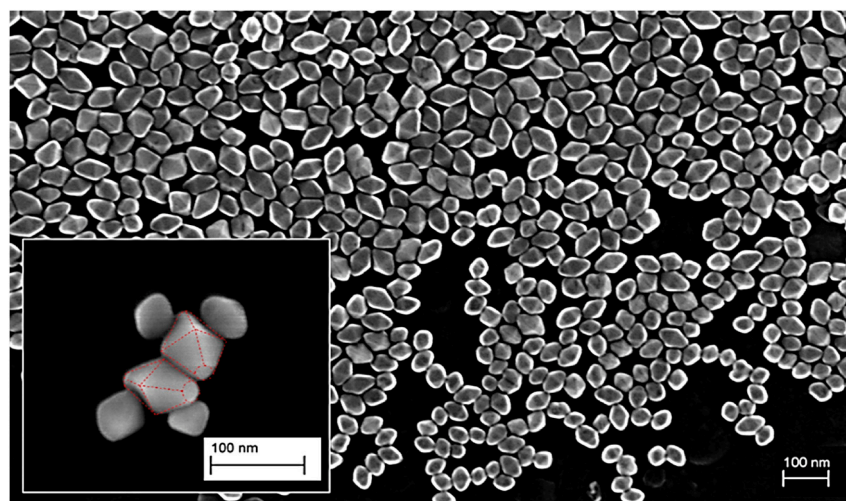


Figure 7. SEM images of TiO_2 NPs deposited on a carbon TEM grid. Insert: representation of bipyramid geometry on particles imaged by a 3 kV In Lens SEM on a silicon substrate. Adapted from [42].

However, near grain boundaries, the haphazard organization of nanoparticles frequently results in complex electron-transport paths and large recombination losses. Optimizing nanoparticle size distribution, crystallinity, and film packing density to enhance charge transfer and interparticle connection has been the focus of recent research. For instance, increased electron diffusion coefficients and improved photovoltaic performance

have been shown in optimized mesoporous TiO_2 nanoparticle films made using controlled sol–gel synthesis and post-treatment procedures [27]. Figures 8 and 9 illustrate the SEM images of mesoporous nanoparticles (NPs) and compact/pressed nanoparticle layers.

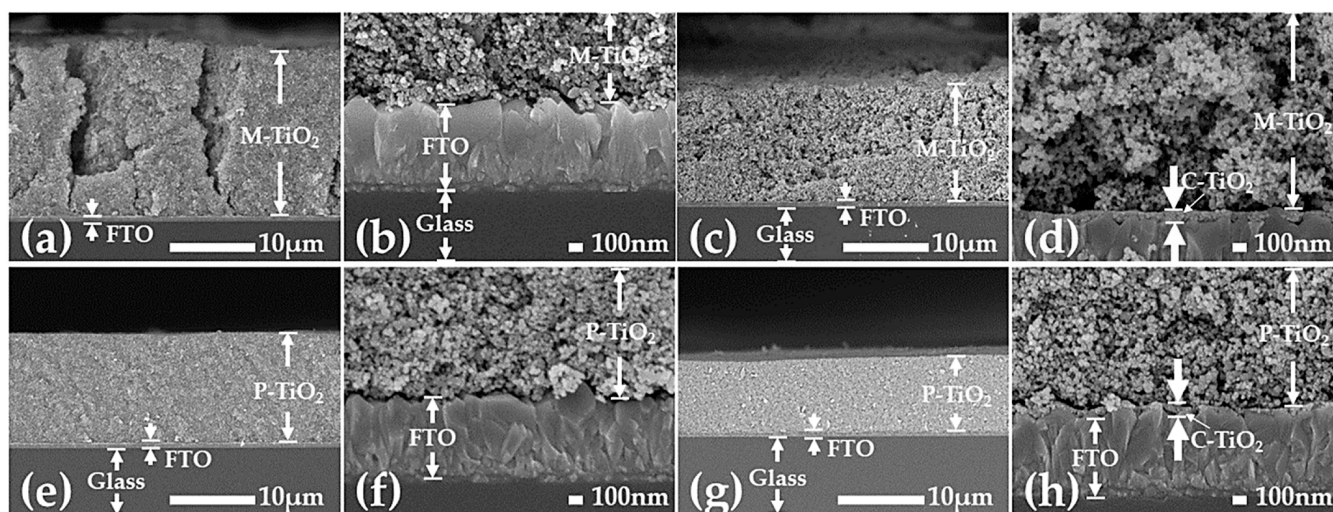


Figure 8. Cross-sectional SEM images of four different structures of photoanode: (a,b) mesoporous TiO_2 layers, (c,d) mesoporous TiO_2 NPs layer stacked on compact TiO_2 thin film, (e,f) pressed TiO_2 NPs layer, and (g,h) pressed TiO_2 stacked on compact TiO_2 thin film. Adapted from [27].

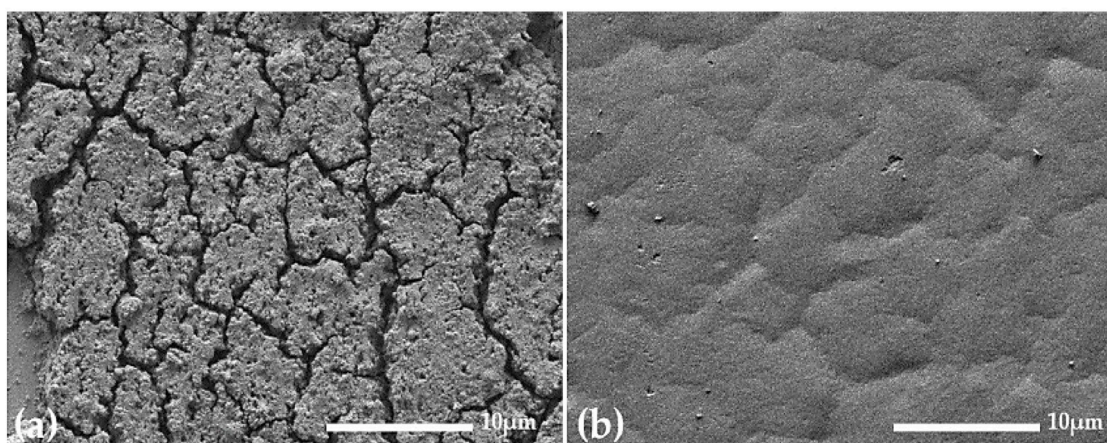


Figure 9. Top view of SEM images of (a) mesoporous TiO_2 NPs layer and (b) pressed TiO_2 NPs layer surfaces. Adapted from [27].

The flat film surface has greater power conversion efficiency because of the compression's role in preventing electrolyte penetration into the FTO substrate's surface. Other investigations also show the fractures on the surface of the mesoporous TiO_2 NPs' layer (Figure 8a). The fast evaporation of water or tert-butanol in the TiO_2 paste caused the fissures. Charge recombination is facilitated by these fissures, which allow the electrolyte to come into contact with the FTO substrate [43]. This study implies that depending on which results are more crucial or desired, TiO_2 fabricated as a pressed/compact layer or its naturally occurring mesoporous layer.

However pressed TiO_2 NPs layer showed the maximum PCE, indicating that the compact and pressed layers successfully increased PCE by avoiding electrolyte contact with the FTO. Because the compact structure of the pressed TiO_2 NPs layer helps electron transmission, the DSSC with photoanode based on the layer has a higher J_{sc} than that based on the mesoporous TiO_2 NPs layer, as shown in Figure 10.

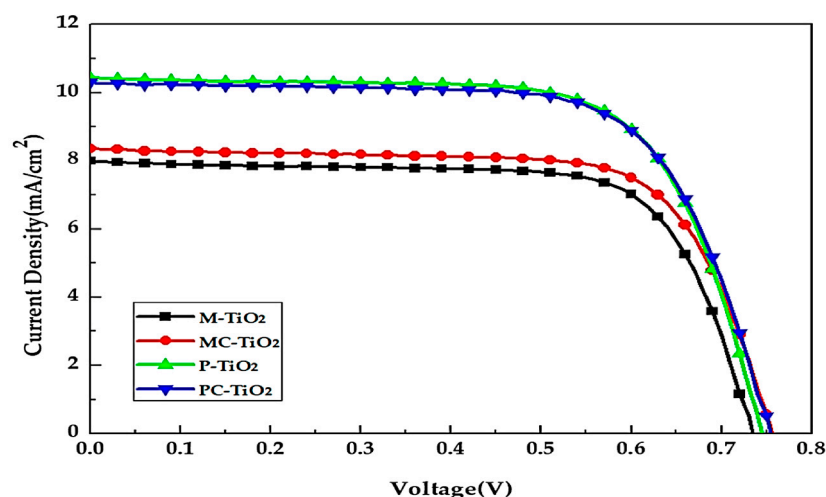


Figure 10. JV characteristics curve of the DSSC with photoanodes based on four different TiO₂ layers. Adapted from [27].

2.2.2. 1D Architectures (Nanorods, Nanotubes, Nanowires)

Because they provide direct routes for electron transport along the structure's axial direction, one-dimensional (1D) TiO₂ nanostructures, such as nanorods, nanowires, and nanotubes, have garnered a lot of interest. One-dimensional topologies allow for quicker electron mobility and less recombination than nanoparticle networks, which need electrons to hop across many grain boundaries [44]. TiO₂-nanotube arrays made by hydrothermal or anodization techniques have shown better carrier lifetimes and longer electron diffusion lengths in DSSCs. Additionally, by giving the dye molecules a direct conduction channel to the conductive substrate, vertically-oriented nanorod arrays promote effective charge collection. In order to improve dye loading while preserving effective electron transport, recent studies have also looked at composite designs that combine nanoparticles with nanorods or nanotubes [45]. Figure 11 shows typical SEM images of nanorods, nanowires, and nanobelts/nanotubes.

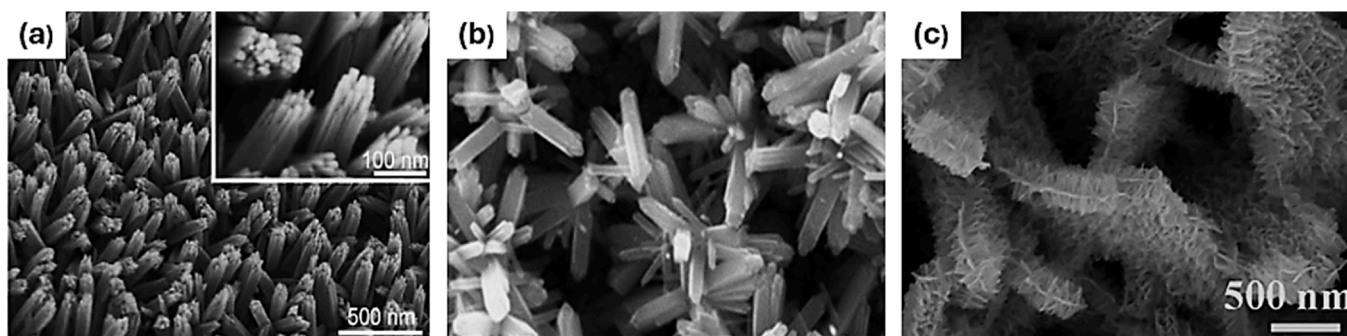


Figure 11. SEM images of (a) synthesized TiO₂ nanowires [46], (b) synthesized TiO₂ nanorods [47], and (c) synthesized TiO₂ nanobelts [48].

The increased density of titania nanoparticles in the nanorods compared to the nanotubes and sintered nanostructures may be the cause of the high values of V_{oc} seen in the nanorod systems. The titania molecules that make up the nanorods are bound and closely packed in homogeneous structural configurations that are perpendicular to the substrate's surface [49]. Compared to the other two architectures, this arrangement guarantees more particles per unit volume. Additionally, the rate of recombination at open-circuit in these structures is lowered because the outside titania particles protect the inner ones from the back reaction with the oxidized species in the redox electrolyte; this improvement has

been proved by the results displayed in Figure 12 [50]. The shorter distance (thin walls) between the trapped electrons in the TiO_2 trap states, and the holes in the redox electrolyte enhances this process, because all of the injected electrons must eventually recombine with the holes in the redox electrolyte at open-circuit; un-trapped electrons rapidly recombine with the holes before they can be trapped again by nearby TiO_2 molecules. By sheltering the electrons caught in the trap states of the inner TiO_2 particles from the holes in the electrolyte, thicker and longer nanotubules can be used to overcome this restriction and reduce recombination at open-circuit [51].

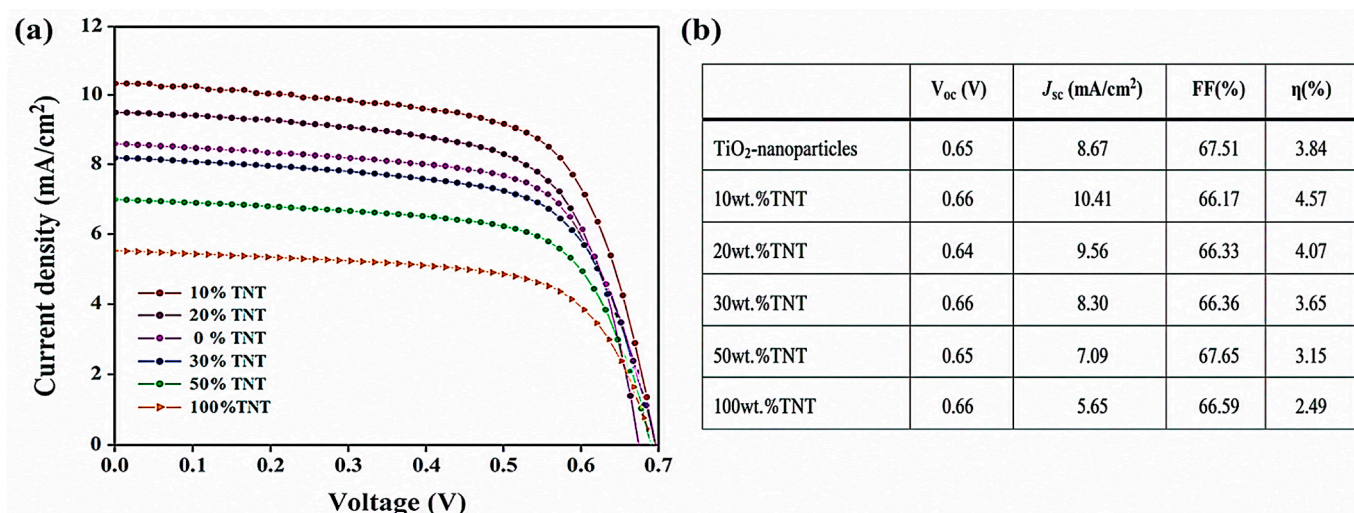


Figure 12. (a) I-V characteristic of the DSSC using hybrid TiO_2 /TNT films; (b) V_{oc} , open-circuit voltage; J_{sc} , integral photocurrent density; FF, fill factor; η , energy conversion of the TiO_2 nanotube (TNT) and TiO_2 nanoparticles. Adapted from [50].

2.2.3. 2D Nanosheets and Exposed-Facet TiO_2

Two-dimensional (2D) TiO_2 nanosheets have shown great promise as photoanode materials because of their exposed crystal facets and strong surface reactivity. Because these high-energy surfaces show improved dye adsorption and quicker electron injection from dye molecules, anatase TiO_2 nanosheets with dominating {001} facets, in particular, have been extensively researched [4], as shown in Figure 13.

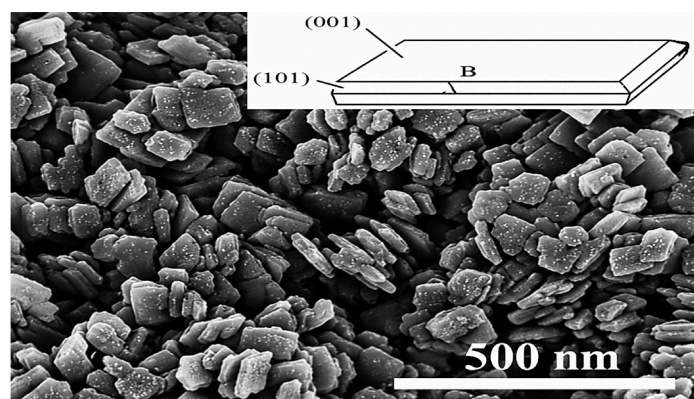


Figure 13. SEM image of images of TiO_2 -NSs (titanium-dioxide nanosheets). Adapted from [4] (Copyright 2010 Royal Society of Chemistry).

In contrast to traditional nanoparticles, nanosheets preserve relatively short electron-transport channels while offering a greater interfacial contact area with dye molecules. Additionally, nanosheets' planar shape may promote better electrolyte diffusion in the

photoanode layer. According to recent studies, TiO₂-nanosheet-based photoanodes perform better in DSSCs due to their increased light harvesting and charge-separation efficiency, as seen in Figure 14 [52].

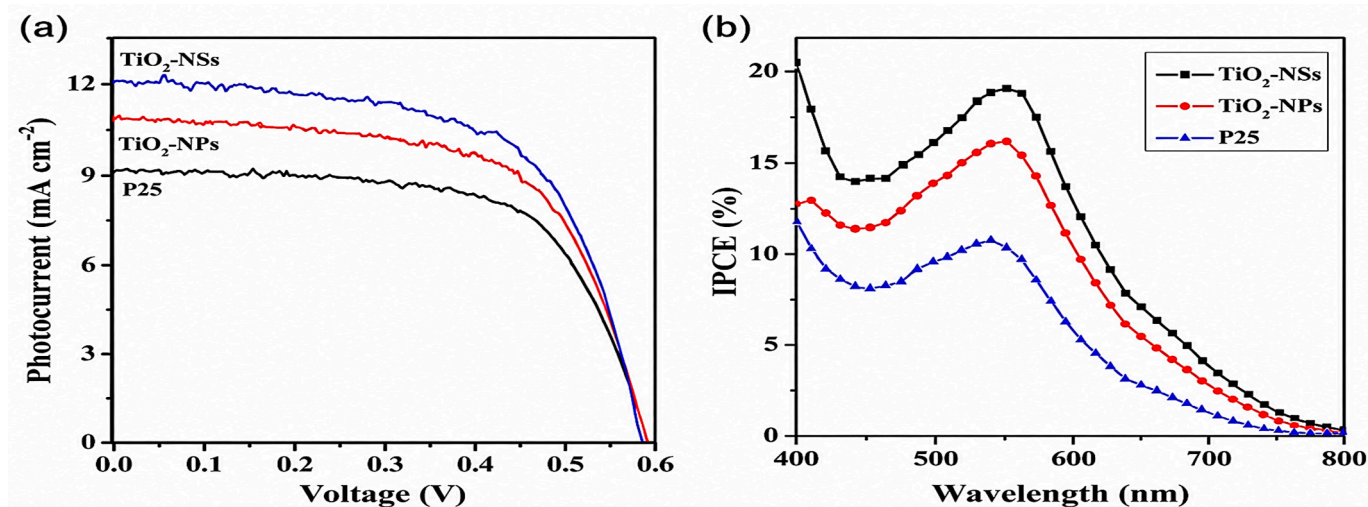


Figure 14. (a) Comparison of the I-V characteristics of DSSCs made from TiO₂-NSs, TiO₂-NPs, and P25 films calcined at 450 °C (b) IPCE of DSSCs made from TiO₂-NSs and TiO₂-NPs and P25 films calcined at 450 °C. Adapted from [4]; copyright permission to reuse (Copyright 2010 Royal Society of Chemistry).

The enhanced performance of TiO₂ nanosheets arises from their high crystallinity, porous structure, and strong light-scattering ability, which improve light harvesting, electron transport, and charge collection. Although TiO₂-NSs exhibit lower dye loading than nanoparticle films, their exposed (001) facets and sheet-like morphology facilitate more efficient charge transfer, electrolyte penetration, and reduced recombination. These findings demonstrate that optimized charge transport and interfacial kinetics can be more important than dye loading alone in determining DSSC efficiency [5].

2.2.4. Hierarchical 3D TiO₂ Architectures

By combining many nanoscale elements into intricate designs that concurrently improve surface area, light scattering, and charge transport, hierarchical three-dimensional (3D) TiO₂ architectures are an advanced method of morphology engineering [53]. Usually, secondary structures, like nanorods, nanosheets, or nanoparticles, are integrated into microscale frameworks, such as spheres, flowers, or porous networks, to create these designs, as can be seen in Figure 15.

By greatly extending the optical path length inside the photoanode, such hierarchical arrangements might enhance light scattering and photon harvesting. Furthermore, because of their optimized pore geometries and linked electron channels, hierarchical TiO₂ designs frequently show enhanced electrolyte penetration and decreased recombination [54–56]. Recent research has shown that by combining the benefits of several dimensionalities, hierarchical TiO₂ photoanodes may reach greater power conversion efficiencies than traditional nanoparticle films. For next-generation DSSC photoanodes, hierarchical morphology engineering is, therefore, increasingly considered a crucial tactic [39–41]. This theory is further supported by the high efficiency rate recorded by hierarchical TiO₂, as shown in Figure 16.

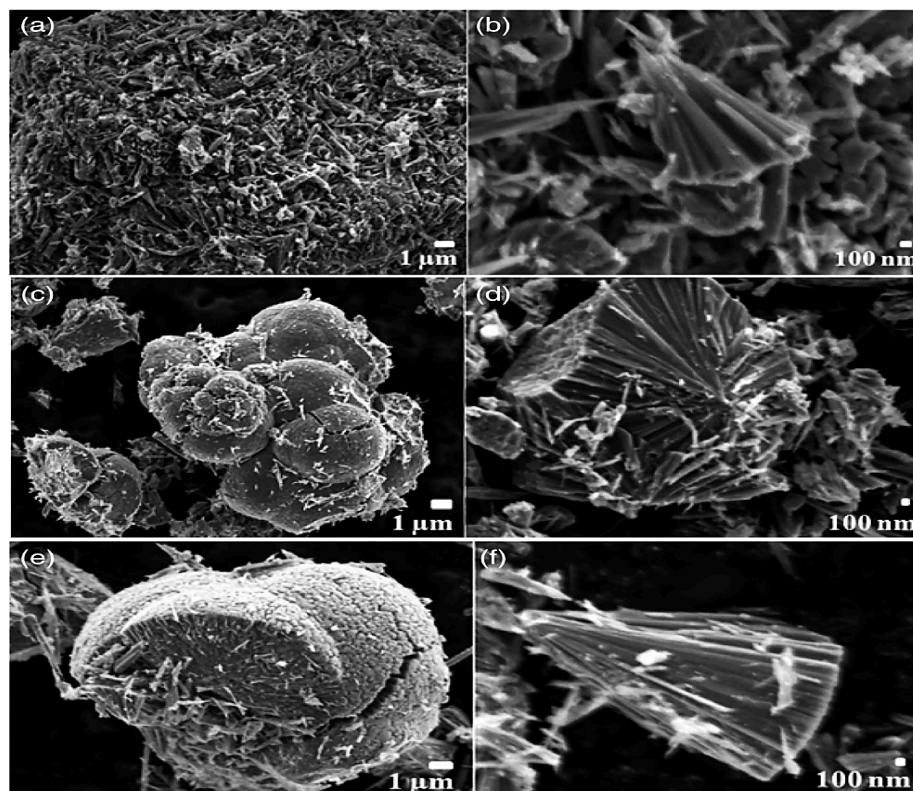


Figure 15. SEM images of the samples prepared at 100 °C (a,b), 130 °C (c,d), and 160 °C (e,f) for 12 h. Growth along the {001} direction, resulting in a crystal morphology, is dominated by the (110) faces. Adapted from [53].

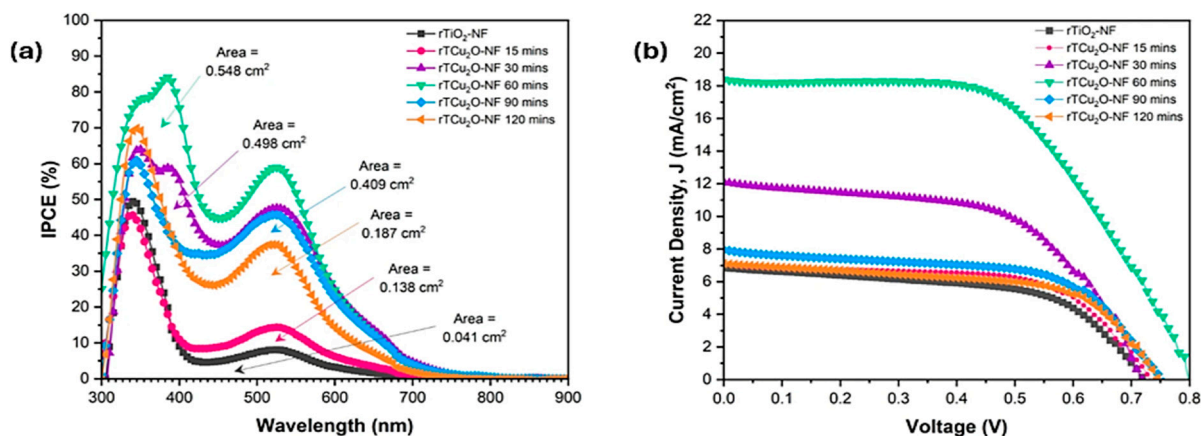


Figure 16. (a) The IPCE spectral response of different hierarchical modified photoanodes: rTiO₂-NF and rTCu₂O-NF 15 min; rTCu₂O-NF 30 min; rTCu₂O-NF 60 min; rTCu₂O-NF 90 min; and rTCu₂O-NF 120 min. (b) The J-V characteristics of different hierarchical modified DSSCs: rTiO₂-NF and rTCu₂O-NF 15 min; rTCu₂O-NF 30 min; rTCu₂O-NF 60 min; rTCu₂O-NF 90 min; and rTCu₂O-NF 120 min. Adapted from [56].

The hierarchical cactus-like TiO₂ nano-thorn architecture significantly improved DSSC performance by enhancing light harvesting and charge collection. The optimized photoanode (60 min growth) exhibited a maximum IPCE exceeding 80%, an integrated IPCE area of 0.548 cm², and a J_{sc} of approximately 18.0 mA cm^{−2}, markedly higher than those of the pristine TiO₂-nanofiber electrode. The improved performance with an efficiency of 8.3% was attributed to the hierarchical morphology, which promoted light scattering, increased photoactive surface area, and facilitated directional electron transport, thereby

enhancing photon utilization while suppressing recombination losses [56]. This study demonstrates the advantages of hierarchical TiO₂ nanoarchitectures for simultaneously optimizing optical absorption and charge transport in DSSCs [57,58].

2.3. Interface Engineering of Titanium Dioxide Semiconductor

One important tactic to enhance the stability and performance of dye-sensitized solar cells (DSSCs) is interface engineering of TiO₂ photoanodes. In DSSCs, electron injection, charge transport, and recombination dynamics are controlled by the interface between TiO₂, dye molecules, and the electrolyte [59]. Device efficiency is decreased by poorly designed interfaces because they frequently produce surface flaws and trap states that speed up electron recombination with oxidized dyes or electrolyte species. In order to maximize charge transfer and inhibit recombination, current research has concentrated on passivation layers, heterostructure interfaces, and interfacial compositional adjustment [60,61].

Further evidence of the effectiveness of interface engineering has been demonstrated using indium (In)-doped and tungsten (W) TiO₂ photoanodes. EIS, Mott–Schottky, electron lifetime, and OCVD analyses revealed enhanced interfacial charge-transfer properties and reduced electron recombination after indium (In) and tungsten (W) incorporation. The optimized Ti photoanode exhibited the lowest charge-transfer resistance, longest electron lifetime, and slowest photovoltage decay, indicating more efficient charge separation and electron transport. Mott–Schottky measurements confirmed the n-type nature of all samples and showed that indium doping modified the interfacial electronic properties. These results highlight the potential of doping as an effective strategy for improving charge collection and overall DSSC performance through interface engineering. Figure 17a shows the Nyquist curves and Figure 17b shows the Mott–Schottky analysis. Likewise, Figure 18a depicts the open-circuit voltage decay (OCVD) measurement, and Figure 18b shows the electron lifetime measurements [19,62].

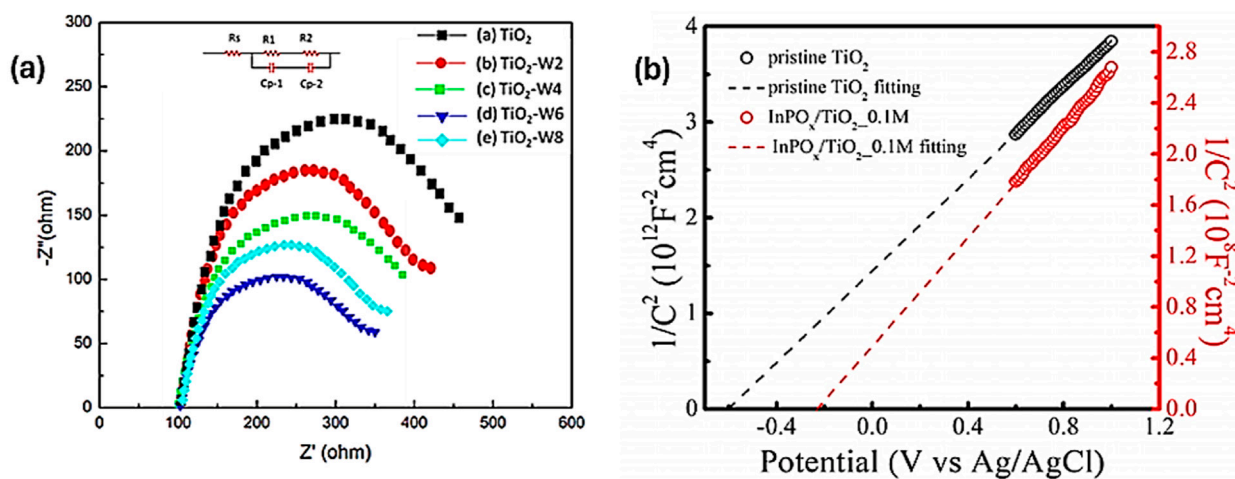


Figure 17. (a) Nyquist plots of W-doped TiO₂ DSSC (adapted from [62]); (b) Mott–Schottky analyses for the photoanodes fabricated by different interfacial In-doped concentrations (adapted from [63]).

The improved photovoltaic performance can be attributed to the beneficial influence of indium incorporation on the electronic and electrochemical properties of TiO₂ [61]. Indium doping increases the carrier concentration and electrical conductivity of the photoanode, thereby facilitating electron transport and reducing charge-transfer limitations within the semiconductor network. Mott–Schottky analysis revealed that the carrier density increased to the order of $10^{12} \text{ F}^{-2} \text{ cm}^4$ after in incorporation, indicating enhanced charge-carrying capability and improved electronic conductivity. These modifications promote more efficient electron injection from the dye molecules and accelerate charge transport

through the TiO_2 framework. However, excessive indium content adversely affects device performance because high dopant concentrations introduce additional electron-scattering centers that hinder charge transport and increase recombination losses.

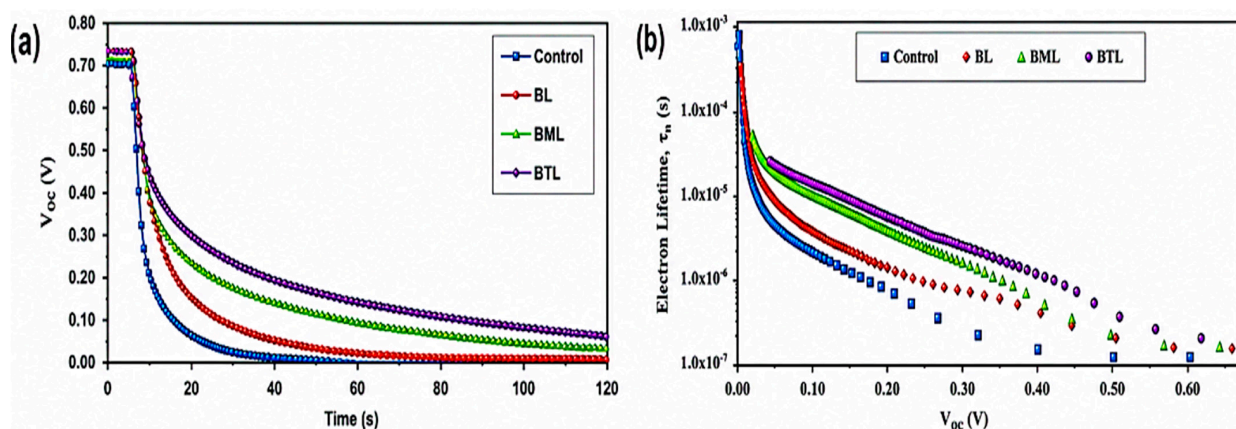


Figure 18. (a) OCVD and (b) electron lifetime for fabricated solar cells with different interfacial photoanodes. Adapted from [64].

Consequently, according to Ayaz et al. (2024) [19], photoanodes containing more than 4% In exhibit a gradual decline in photovoltaic performance. At the optimal doping concentration, the combined improvements in conductivity, interfacial charge-transfer kinetics, and electrochemical properties resulted in a power conversion efficiency of 8.11%. Furthermore, the introduction of a compact TiO_2 blocking layer further suppressed interfacial recombination at the FTO/photoanode interface, leading to an additional enhancement in efficiency to 8.62% [19]. These findings demonstrate that controlled indium doping, combined with appropriate interfacial engineering strategies, provides an effective route for optimizing charge transport and maximizing DSSC performance [39,62].

In interface engineering, surface passivation using wide-bandgap or insulating coatings has also shown promise. In order to provide energy barriers that prevent electron back-transfer to the electrolyte while preserving effective electron injection from the dye, materials like MgO , ZrO_2 , and SnO_2 are often placed on TiO_2 surfaces [65,66]. Figure 19 depicts that TiO_2 nanoparticles with exposed {001} facets achieved markedly higher DSSC efficiency (7.06%) than randomly shaped TiO_2 nanostructures (3.47%), mainly due to enhanced dye adsorption and improved light harvesting, which significantly increased photocurrent generation.

EIS analysis indicated only minor changes in charge-transfer resistance, suggesting that improved optical properties were the dominant factor governing performance enhancement [67]. This suggests that electron transport and recombination were not the main reasons for the improved efficiency. Open-circuit voltage decay measurements showed that the random TiO_2 structures exhibited slightly longer electron lifetimes. However, the difference in electron lifetime was too small to account for the large increase in photocurrent observed for the {001}-facet nanoparticles. Overall, the study concluded that exposing the high-energy {001} facets of anatase TiO_2 enhances DSSC performance primarily by increasing dye adsorption and light absorption efficiency [68], as shown in Figure 19. The passivation layer lowers the density of surface trap states that promote recombination, which results in this improvement. It has been demonstrated that these tactics increase photocurrent and total DSSC efficiency [20].

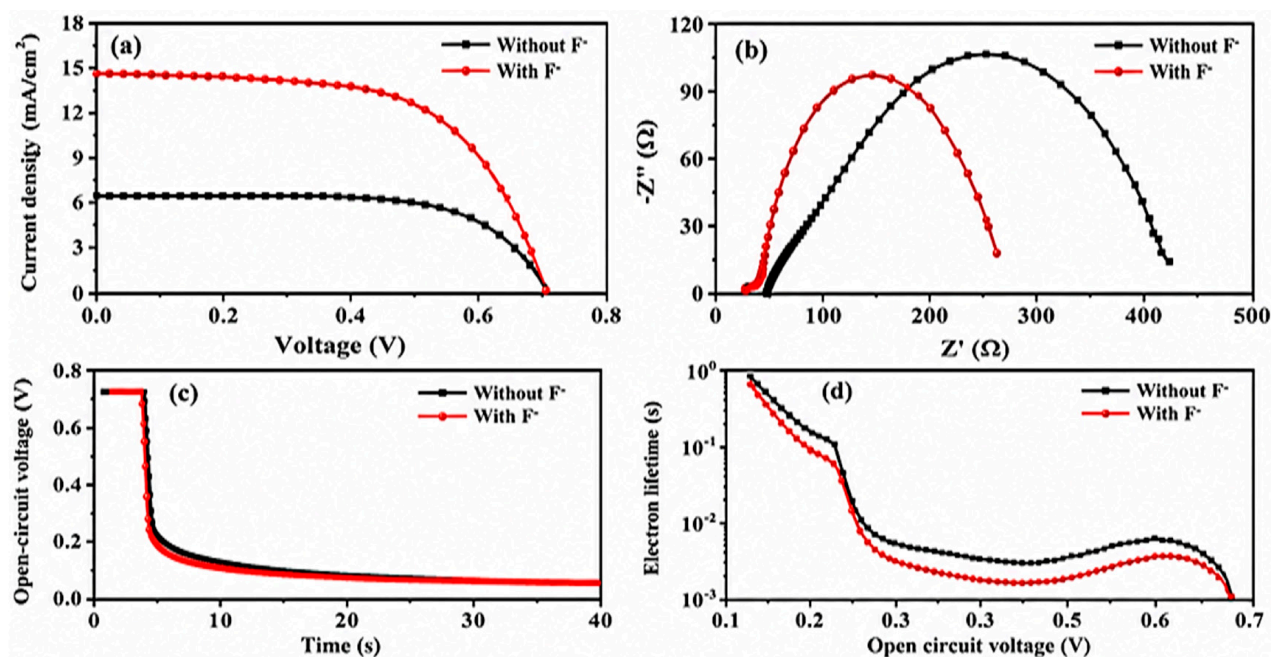


Figure 19. Photovoltaic characteristics of the DSSCs based on random-shape TiO₂ nanostructures (noted “Without F⁻”) and truncated-octahedron TiO₂ nanoparticles with exposed {001} facets (noted “With F⁻”): (a) I-V curves; (b) Nyquist plots by EIS measurements; (c) open-voltage decay measurement upon turning off the illumination; (d) electron lifetime determined from the data of (c). Adapted from [67].

Apart from solid-state passivation layers, electrolyte additives such as 4-tert-butylpyridine (TBP), N-methylbenzimidazole (NMB), and imidazolium derivatives provide molecular-level passivation of the TiO₂ surface and play important roles in regulating interfacial charge-transfer processes in DSSCs [69]. Their effectiveness, however, depends strongly on the redox electrolyte employed. In conventional iodide/triiodide electrolytes, TBP is widely used to suppress electron recombination by adsorbing onto TiO₂ surface sites and shifting the TiO₂ conduction-band edge toward more negative potentials, thereby increasing Voc. NMB exhibits similar behavior and often produces stronger conduction-band shifts due to its higher basicity [64,65]. In cobalt-based DSSCs, TBP and related nitrogen-containing additives are frequently employed to mitigate the faster recombination kinetics associated with cobalt redox mediators [70,71]. However, excessive additive concentrations may influence cobalt-complex diffusion and mass transport, requiring careful optimization [72]. In quasi-solid-state and gel-polymer electrolytes, TBP and NMB remain beneficial for interfacial passivation, although their influence on charge transport can differ from that observed in liquid electrolytes because ionic mobility is partially governed by the polymer matrix [73]. Therefore, the selection and concentration of these additives must be tailored to the specific electrolyte chemistry and device architecture [69,74,75].

Engineering heterointerfaces using hybrid or composite photoanodes, as shown in Figure 20, is another significant avenue. Electronic conductivity and interfacial charge transfer can be improved by combining TiO₂ with conductive materials such as MXenes, conductive polymers, or transition-metal oxides. For example, the incorporation of Ti₃C₂T_x MXene into TiO₂ photoanodes creates a conductive interface that decreases recombination losses and promotes electron transport.

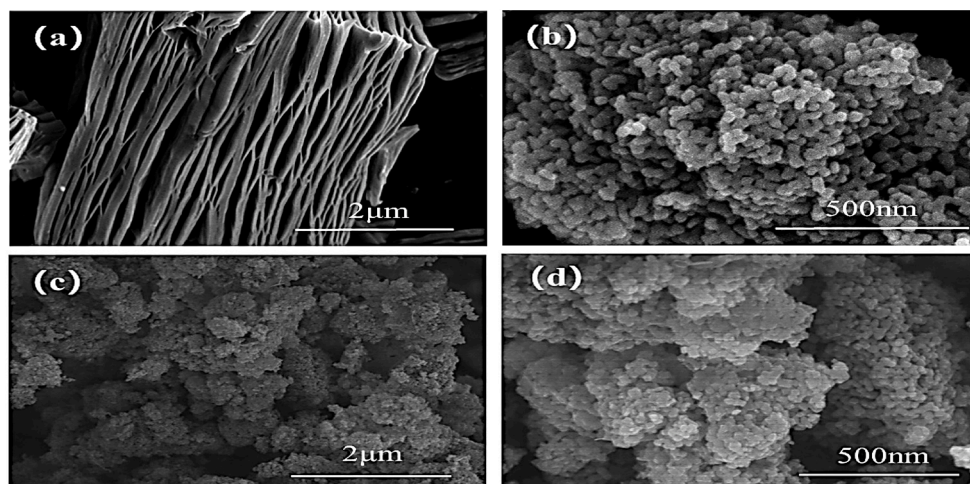


Figure 20. Field-emission SEM images of (a) MXene, (b) TiO_2 and (c,d) $\text{TiO}_2/\text{MXene}$ (4:1) composites. Adapted from [76].

According to studies using density functional theory, these hybrid interfaces alter TiO_2 's conduction-band alignment, encouraging more effective electron injection from dye molecules as seen in Figure 21. Because of enhanced carrier separation and longer electron lifetimes, experimental systems employing MXene-modified TiO_2 photoanodes have shown significant gains in power conversion efficiency [5,20,77]. This is observed in Figure 21, while Figure 22 proves the electrical improvement of said hybrid interfaces.

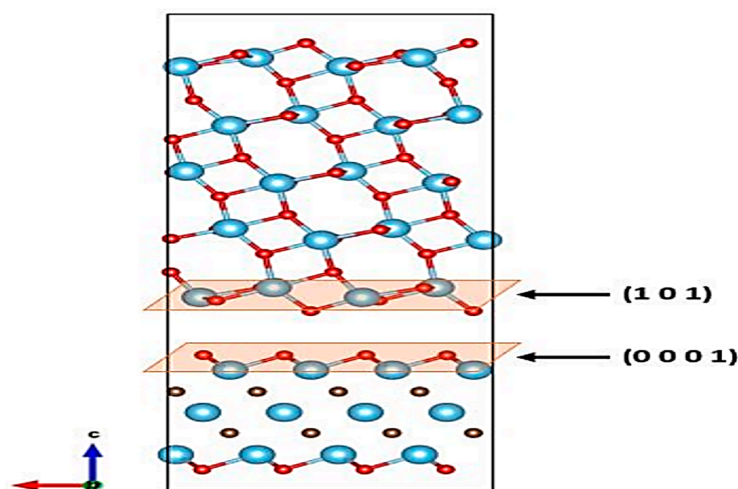


Figure 21. Structure used for DFT simulation (side view). The interaction planes are schematically shown (orange): (101)- TiO_2 material and (0 0 0 1)- $\text{Ti}_3\text{C}_2\text{Tx}$. Adapted from [77]; copyright permission to reproduce (Copyright 2022 American Chemical Society).

Other methods for modifying interfaces include chemical doping and defect engineering. TiO_2 's electronic structure may be adjusted, surface states can be changed, and charge mobility can be enhanced by adding heteroatoms or dopants [78,79]. For instance, it has been reported that Fe/S-modified TiO_2 nanoparticles improve photoinduced charge separation and facilitate electron transport at the TiO_2 /dye interface, increasing DSSC performance. These changes increase the efficiency of light-to-electricity conversion and decrease recombination pathways, as shown above in Figure 3 [35]. Three samples of each Fe/S wt% samples analyzed and annotated with (1), (2) and (3) as seen in Figure 22.

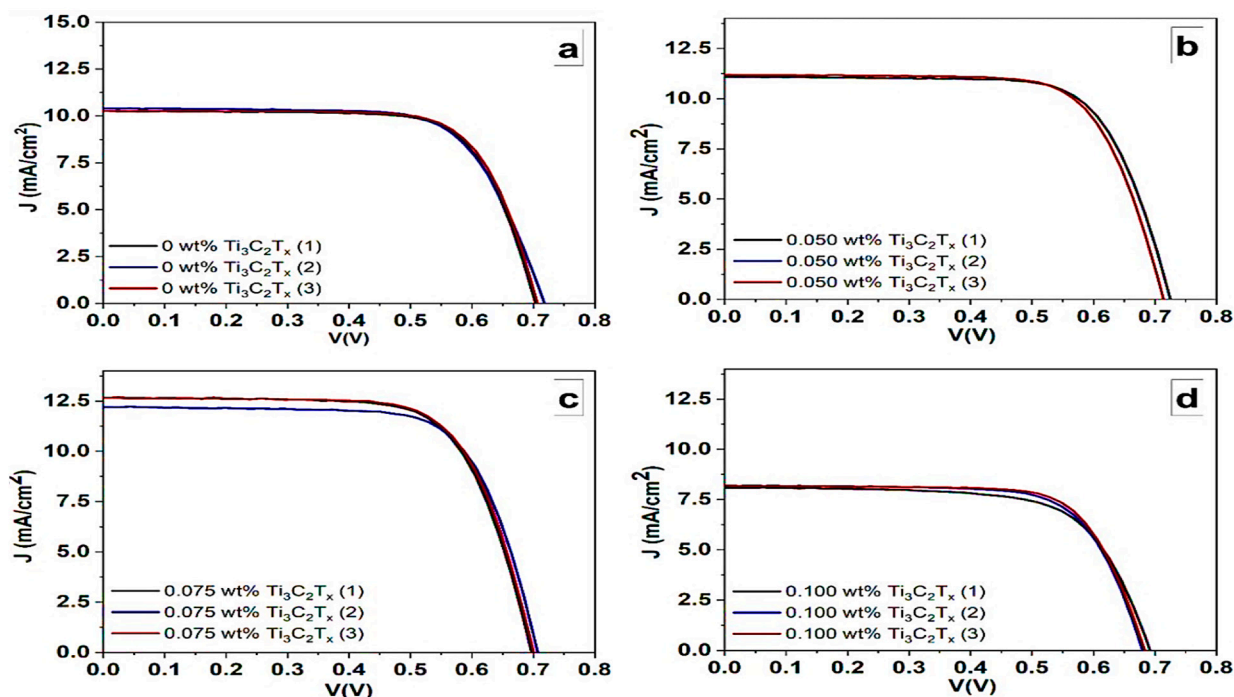


Figure 22. J-V curves of DSSCs using photoanodes based on bare TiO₂ (a) and with addition of 0.05 wt% (b), 0.075 wt% (c), and 0.10 wt% (d) of Ti₃C₂T_x. Adapted from [78]; copyright permission to reproduce (Copyright 2022 American Chemical Society).

In addition to chemical modification, interface engineering via structural design is becoming more popular. One-dimensional TiO₂ nanorods or nanotubes combined with mesoporous TiO₂ matrices are examples of hierarchical nanostructures that offer better dye loading and electron-transport channels. These designs preserve effective electron collecting routes while increasing the effective interfacial area for dye adsorption. Multidimensional TiO₂ nanoarchitectures are attractive candidates for next-generation DSSC photoanodes because they greatly improve light harvesting and charge transfer, according to recent evaluations [80].

2.4. Influence of Fabrication and Coating Techniques on TiO₂ Interface and Morphology Quality

The fabrication method used for TiO₂ photoanodes plays a critical role in determining film morphology, porosity, interface quality, defect density, adhesion, and scalability. Conventional techniques such as doctor blading and screen-printing produce highly porous films with excellent dye-loading capability and remain attractive for large-scale DSSC manufacturing; however, they may suffer from thickness non-uniformity and crack formation during drying [81,82]. Spin coating provides smoother and more uniform films with lower defect densities but generally produces thinner photoanodes with limited porosity. Spray pyrolysis offers uniform large-area coatings, good adhesion, and controllable crystallinity, making it suitable for scalable production. Advanced techniques such as atomic layer deposition (ALD) and sputtering provide superior interface control, dense compact layers, and effective defect passivation, thereby reducing interfacial recombination [79]. Meanwhile, hydrothermal growth enables the synthesis of highly crystalline one-dimensional and hierarchical TiO₂ nanostructures that facilitate directional electron transport and improved charge collection. Overall, the choice of deposition technique directly influences charge transport, recombination kinetics, and device reproducibility, highlighting the importance of process optimization alongside material design [26,76]. Table 1 summarizes the influence

of TiO₂ fabrication techniques on photoanode quality and DSSC performance, highlighting the trade-off between scalability, morphology control, and interface engineering [83].

Table 1. Relationship between TiO₂ fabrication methods and key photoanode characteristics in DSSCs.

Technique	Porosity	Interface Quality	Defect Control	Scalability	Reference
Doctor Blading	High	Moderate	Moderate	High	[84]
Screen Printing	High	Moderate	Moderate	High	[79]
Spin Coating	Low–Moderate	High	High	Moderate	[85]
Spray Pyrolysis	Moderate	High	Moderate–High	High	[86]
Hydrothermal Growth	High	High	High	Low	[80]
Sputtering	Low	Very High	High	Moderate	[81]
ALD	Low	Excellent	Excellent	Low	[60,87]

3. Common Misinterpretations in the TiO₂ DSSC Literature

Several commonly reported correlations in TiO₂-based DSSC research are often oversimplified and may lead to misleading conclusions. First, a higher TiO₂ surface area does not necessarily lead to higher power conversion efficiency, as excessive surface area can increase trap-state density and electron recombination despite improved dye adsorption [20]. Similarly, oxygen vacancies are often considered beneficial because they enhance conductivity and electron transport; however, excessive vacancy concentrations can create recombination centers that shorten electron lifetimes and reduce device performance. Higher dye loading is also not always advantageous, as thicker or highly porous films may hinder electron collection and increase transport losses. Likewise, exposed anatase {001} facets have been reported to improve dye adsorption and photocurrent [14]. Still, their benefits depend strongly on crystal quality, surface chemistry, and recombination kinetics rather than facet exposure alone. Another frequent misconception concerns electrochemical impedance spectroscopy (EIS), in which reduced charge-transfer resistance is often interpreted as evidence of improved charge transport without adequately accounting for concurrent changes in recombination dynamics [19]. Finally, increases in short-circuit current density (J_{sc}) may occur at the expense of higher recombination rates and lower open-circuit voltage (V_{oc}), highlighting the need to evaluate all photovoltaic parameters simultaneously. Recent reviews emphasize that DSSC performance should be assessed through a comprehensive analysis of charge transport, recombination resistance, electron lifetime, and energy-level alignment rather than isolated metrics [88].

3.1. Challenges and Future Perspectives

Despite decades of improvement, TiO₂-based DSSCs continue to encounter difficulties that hamper their efficiency, stability, and large-scale commercialization. Recent investigations have shown that defect states, morphological complexity, and interfacial recombination processes all have a significant impact on charge transport and device stability. Addressing these issues through rational materials engineering is a critical research focus for the next generation of DSSCs [23,24,89,90].

3.1.1. Challenges in TiO₂ Engineering for DSSCs

Charge Recombination Induced by Surface and Bulk Defects

TiO₂ defect states have two functions in DSSCs. On the one hand, electron transport and electrical conductivity can be improved by oxygen vacancies and Ti³⁺ centers. Conversely, high defect density creates trap states that encourage charge recombination

between oxidized species in the electrolyte and photoinjected electrons in TiO_2 . Overall power conversion efficiency and open-circuit voltage are greatly decreased by this recombination [91]. One of the main loss mechanisms in DSSCs is the existence of interfacial trap states, according to recent research. To prevent electron recombination at the FTO/ TiO_2 interface, blocking layers and interface engineering techniques are frequently needed [67,68]. Sputtered TiO_2 blocking layers, for instance, have been demonstrated to enhance device performance and decrease reverse electron transport [81]. Controlling defect concentration precisely is another new problem. Non-radiative recombination centers result from uncontrolled defect development during synthesis or thermal processing, while modest defect concentrations can increase electron mobility. Therefore, reduced recombination losses and increased conductivity must be balanced in defect engineering.

Morphological Trade-Off Between Surface Area and Electron Transport

Light scattering, electron transport routes, and dye loading are all significantly influenced by the shape of TiO_2 nanostructures. Mesoporous TiO_2 nanoparticle films that offer a large surface area for dye adsorption are used in conventional DSSCs. However, in these networks of nanoparticles, electron transport happens via a random hopping process that raises the likelihood of recombination [80]. To enhance charge transmission, alternative morphologies such as nanotubes, nanorods, and hierarchical structures have been investigated. By offering direct electron routes, these designs lessen trap-limited diffusion. However, compared to networks of nanoparticles, they usually provide less surface area for dye adsorption, which results in less light harvesting [92–94]. According to recent experimental research, hybrid architectures that combine nanoparticles with hierarchical scattering layers can boost electron transport and dye adsorption at the same time, improving photovoltaic efficiency [10,26]. Achieving the ideal balance between large surface area and effective electron transport is still difficult despite these developments. The development of fabrication methods that allow for exact control over particle connectivity and pore structure is currently ongoing.

Interfacial Energy Alignment and Charge Transfer Limitations

Rapid electron injection from the excited dye into the TiO_2 conduction band and low recombination at the dye/ TiO_2 and TiO_2 /electrolyte interfaces are necessary for effective DSSC functioning. However, this process is frequently hampered by interfacial imperfections and improper energy alignment. To enhance interfacial charge transfer, composite photoanodes have been investigated [95,96]. For instance, depending on their composition and crystallinity, TiO_2 /ZnO hybrid structures have been demonstrated to affect electron transport and recombination kinetics. On the other hand, excessive secondary oxide incorporation may result in increased recombination pathways or undesirable band alignment. Additionally, a crucial concern is the durability of dye– TiO_2 bonding during prolonged light and electrolyte exposure. Interface deterioration can hasten device breakdown and progressively lower electron injection efficiency [62,66].

Stability and Long-Term Degradation

DSSCs frequently experience long-term deterioration because of interactions between the photoanode, dye molecules, and electrolyte, despite TiO_2 's chemical stability. Dye desorption, electrolyte leakage, and structural alterations in the porous TiO_2 network are examples of degradation processes. Recent experimental and computational research has shown that factors including dye content, porosity, and TiO_2 film thickness have a major impact on long-term degradation rates. The intricate connections between structural factors and DSSC stability have even been examined using machine learning algorithms. Therefore, improved control over material microstructure and a deeper comprehension of

degradation processes are necessary to increase the longevity of TiO₂ photoanodes [97]. The morphology and interfacial properties of TiO₂ photoanodes strongly influence both the efficiency and long-term stability of DSSCs [33]. Although mesoporous nanoparticle films provide high dye-loading capacity, their abundant grain boundaries and defect sites can accelerate charge recombination and structural degradation. In contrast, one-dimensional TiO₂ nanostructures and hierarchical architectures offer improved electron transport, reduced recombination, and enhanced structural stability [93]. Furthermore, interface engineering through passivation layers, blocking layers, and surface coatings suppresses electron back-transfer and mitigates electrolyte-induced degradation. Therefore, the simultaneous optimization of TiO₂ morphology, defect chemistry, and interfacial characteristics is essential for developing durable high-performance DSSCs [23].

3.2. Future Perspectives for Advanced TiO₂ Photoanodes

3.2.1. Hierarchical and Multiscale Morphology Design

It is expected that the upcoming generation of DSSC photoanodes will use hierarchical TiO₂ structures that incorporate several length scales. These architectures combine effective electron-transport channels with a high capacity for dye adsorption. For instance, hydrothermally produced hierarchical TiO₂ aggregates can function as scattering layers that improve light harvesting while preserving meso-porosity for dye loading. To maximize electron transport across the photoanode, future studies may use nanostructured scaffolds like nanotubes, nanosheets, or three-dimensional networks.

3.2.2. Advanced Interface Engineering

The improvement of DSSC performance will continue to depend heavily on interface engineering. Heterostructure creation, ultrathin blocking layers, and surface treatments can all greatly lower recombination losses. The use of metal–organic frameworks (MOFs) or other porous materials with TiO₂ to improve electron transport and charge separation is an emerging strategy. Improved electron transport characteristics and compatibility with flexible substrates have already been shown by MOF/TiO₂ composite photoanodes made at low temperatures. Furthermore, molecular surface modification and atomic layer deposition (ALD) methods may allow for exact control of interfacial characteristics [29,30].

3.2.3. Integration with Emerging Materials and Flexible Devices

Additionally, flexible, and wearable solar systems are the focus of future DSSC research. Improved mechanical stability and low-temperature processing of TiO₂ photoanodes are necessary to accomplish this aim [77]. Effective charge transfer in flexible devices may be made possible by hybrid materials that combine TiO₂ with conductive frameworks or organic–inorganic composites. Moreover, combining TiO₂ DSSCs with hybrid perovskite systems or tandem solar designs may create new possibilities for high-efficiency photovoltaic solutions [97].

4. Conclusions

Recent advances in TiO₂ photoanode engineering have significantly enhanced DSSC performance by improving defect chemistry, morphology, and interfacial properties. Strategies such as defect engineering, elemental doping, and composite formation have improved charge transport, light harvesting, and recombination suppression. Simultaneously, the development of nanostructured and hierarchical TiO₂ architectures has enabled improved dye adsorption, light scattering, and electron transport. Interface engineering approaches, including passivation layers, heterostructures, MXene-based composites, and atomic layer deposition, have further reduced interfacial recombination and enhanced charge separation.

Despite these advances, challenges remain in balancing defect concentration, electron transport, dye loading, and long-term stability. Future progress will depend on integrating morphology, defect, and interface engineering with scalable fabrication methods and emerging materials, such as MOFs, MXenes, conductive polymers, and hybrid photovoltaic architectures. Together with advances in computational modeling and in situ characterization, these developments are expected to accelerate the realization of efficient, stable, and commercially practical TiO₂-based DSSCs.

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Abbreviations

The following abbreviations are used in this manuscript:

TiO ₂	Titanium Dioxide
0D	Zero-Dimensional
1D	One-Dimensional
2D	Two-Dimensional
ALD	Atomic Layer Deposition
DSSC	Dye-Sensitized Solar Cell
DRS	Diffuse Reflectance Spectra
DC-DSSC	Direct Contact Dye-Sensitized Solar Cell
Fe ₂ O ₃	Iron Oxide
SW-DSSC	Sandwich Structure Dye-Sensitized Solar Cell
NP	Nanoparticle
NW	Nanowire
NR	Nanorod
MOF	Metal–Organic Framework
MgO	Magnesium Oxide
FTO	Fluorine-Doped Tin Oxide
I-V	Current-Voltage
J-V	Current-Voltage
SnO ₂	Tin Oxide
T-Z	TiO ₂ –ZrO ₂
ZrO ₂	Zirconia
ZnO	Zinc Oxide

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