

Nanomaterials for photoelectrodes in dye sensitized solar cells-a Review

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ABSTRACT

Dye-sensitized solar cells (DSSCs) are the representative of third generation photovoltaic devices due to its low cost, easy fabrication, and relatively high energy conversion efficiency. Still, there is a lot of scope for the replacement of current DSSC materials due to their high cost, less abundance, and long-term stability. This paper summarizes the recent progress in DSSC technology for improving efficiency, focusing on the active layer in the photoanode, with a part of the DSSC consisting of dyes and a TiO₂ film layer. The application of these nanostructured photoanode materials and their impact on the device efficiency has been described in detail.

Keywords: dye sensitized solar cells, fill factor, photoelectrode, power conversion efficiency

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I. INTRODUCTION

Fossil fuels (coal, oil, and natural gas) form the major energy source for meeting current human needs, yet cause a range of serious environmental issues. Moreover, the ever-growing consumption rate outpaces their regeneration rate, endangering the exhaustion of fossil fuels on earth. Among all new competing energy sources (biomass, solar, wind, hydroelectricity, geothermal energy, and nuclear energy), solar energy is considered to be the most abundant, renewable, and environment-friendly energy form.

Dye-sensitized solar cells (DSSCs) have arisen as a technically and economically credible alternative to the p-n junction photovoltaic devices. Furthermore, DSSCs work better even during darker conditions, such as in the dawn and dusk or in cloudy weather. Such capability of effectively utilizing diffused light makes DSSCs an excellent choice for indoor applications like windows and sunroof [1].

DSSC consists of a mesoporous nanocrystalline n-type semiconductor (typically TiO₂) sensitized with a dye, deposited onto an anode and immersed in redox active electrolyte (generally triiodide/iodide), completed by a counter-electrode (cathode) [2]. Upon illumination, dye molecules anchored onto the TiO₂ surface absorb incoming photons, allowing the photoinduced electron-transfer from their excited state into the TiO₂ conduction band. Then, the electrons are transferred to the counter electrode, thus creating a current. The redox electrolyte reduces the oxidized

dye molecules back to their ground state to enable continuous electron production. Even if power conversion efficiencies (PCEs) above 13% have been reached for liquid-electrolyte DSSCs [3,4] improvements are still required in order to be commercially viable.

Dye-sensitized solar cell (DSSC) offers an efficient and easily implemented technology for future energy supply. Compared to conventional silicon solar cells, it provides comparable power conversion efficiency (PCE) at low material and manufacturing costs. DSSC materials such as titanium oxide (TiO₂) are inexpensive, abundant and innocuous to the environment. Since DSSC materials are less prone to contamination and processable at ambient temperature, a roll-to-roll process could be utilized to print DSSCs on the mass production line. The main objective of this review article is to provide an overview of TiO₂-based photoanode layer used in dye-sensitized solar cells (DSSCs).

II. THE STRUCTURE AND OPERATION PRINCIPLE

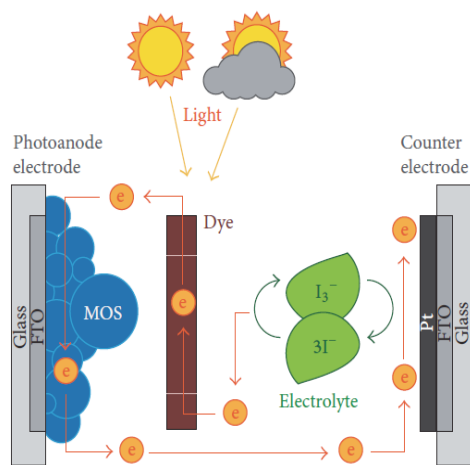


Figure 1: Schematic representation of a DSSC device.

The device is comprised of four components only:

- (i) Nanostructured *n*-type semiconductor (wide band gap metal oxide) coated over transparent conducting substrate (TCO (ITO [6] or FTO [7]))
- (ii) Visible-light absorber dye (several organic dye can be used, such as N3 [8], N719 [9], N749 [10] (the so called black dye), K8 [11], K19 [12], CYC-B11 [13], and C101 [5])
- (iii) Electrolyte containing redox couple (typically, I^-/I_3^-) in an organic solvent to collect electrons at the counter electrode and effecting dye-regenerating; and [14, 15]
- (iv) Counter electrode (TCO coated with a platinum layer or other suitable catalyst)

The working of DSSCs is a step-by-step sequential phenomenon, carried out by different layers as shown below:

- The dye or the sensitizer is the photoactive material of DSSC and is responsible for the production of energy on exposure to light.
- The dye absorbs the incoming light (sunlight), that is, the photons that cause the excitation of electron from HOMO to LUMO in the dye.
- After that, these excited electrons are then transported into the conduction band of TiO_2 (a white pigment commonly found in white paint) and thus the anode of DSSCs. Meanwhile, the dye rapidly reduced by taking an electron from the electrolyte. The rare-earth elements are one of the most potential dopants for TiO_2 explored to date in order to modify and improve the functionality of TiO_2 in DSSCs.

When the sunlight strikes the solar cell, dye sensitizers on the surface of TiO_2 film get excited and the electrons in turn get injected into the conduction band of TiO_2 . Within the TiO_2 film, the injected electrons diffuse all the way through the

mesoporous film to the anode and are utilized to do useful work at the external load. Finally, to complete the cycle, these electrons are collected by the electrolyte at counter electrode which in turn are absorbed to regenerate the dye sensitizer. The overall performance of the DSSC can be evaluated based on sunlight-to-electric power conversion efficiency (PCE or η) and fill factor

$$FF = \frac{V_{oc} J_{sc}}{P_{in}} \quad (1)$$

$$\eta = \frac{V_{oc} J_{sc} FF}{P_{in}} \times 100\% \quad (2)$$

where J_{sc} is the short-circuit current density ($mAcm^{-2}$), V_{oc} , the open-circuit voltage (V), FF the fill factor and P_{in} the incident light power. J_{max} and V_{max} corresponds to current and voltage values, respectively, where the maximum power output is given in the $J-V$ curve. The photovoltage (V_{oc}) is determined by the potential difference between Fermi-level of electrons in the TiO_2 film and redox potential of electrolyte. Similarly, the photocurrent (J_{sc}) is determined based on the incident light harvest efficiency (LHE), charge injection and collection efficiencies. The fill factor (FF) represents the ratio of the actual maximum obtainable power to the product of the open circuit voltage (V_{oc}) and short circuit current (J_{sc}).

In general, the overall conversion efficiency of dye-sensitized solar cells is tested under standard irradiation conditions (100 mW/cm^2 , AM 1.5). The I-V curve to show the cell performance is shown in Fig2.

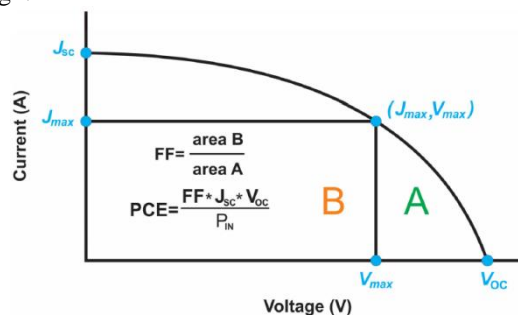


Fig. 2 I-V curve to evaluate the cells performance

Under standard test conditions, the device efficiency can be maximized through optimizing each of these three parameters (V_{oc} , J_{sc} , and FF). For instance, high open-circuit potential can be obtained by using $C_o(II/III)$ redox couple which has more positive redox potential and therefore increases the potential difference. Likewise, the short-circuit current can be enhanced by using panchromatic dye

sensitizers which can absorb broad sunlight covering visible to the near-infrared range in solar spectrum. The fill factor is yet another important parameter that reflects the quality of solar cells. Increasing the shunt resistance and decreasing the series resistance as well as reducing the overvoltage for diffusion and electron transfer will lead to a higher FF value, thus resulting in greater efficiency and pushing the output power of the solar cell closer towards its theoretical maximum. In fact, these parameters are highly dependent on material properties and physical processes within the device. Therefore, theoretical models that can capture the characteristics of physical process and materials properties are critical to optimize various operation parameters and cell configurations.

III. EXPERIMENTAL FINDINGS

Mathew et al.[16] prepared TiO₂ nanoparticle based photoanodes sensitized with molecularly engineered porphyrin based sensitizers (SM315) which exhibits an efficiency of 13%. The sensitizer exhibits enhanced visible and long wavelength absorbance combined with excellent electrolyte compatibility.

All the record efficiencies reported so far uses TiO₂ nanoparticles as the semiconductor layer and the enhancement in the efficiencies are mainly attributed to the performance of the dye. The one-dimensional nanostructure provides higher light scattering compared to nanoparticles. Although one dimensional photoanodes show great promise in terms of the electron transport and light scattering, the maximum efficiency reported so far using nanotube electrodes is 9.5% [17,18]. The major drawback of the one-dimensional photoelectrodes is the low surface area available for the adsorption of dye molecules.

Varghese et al. [19] found that dye-sensitized solar cells consist of a random network of titania nanoparticles that serve both as a high-surface-area support for dye molecules and as an electron-transporting medium. Despite achieving high power conversion efficiencies, their performance is limited by electron trapping in the nanoparticle film. Electron diffusion lengths can be increased by transporting charge through highly ordered nanostructures such as titania nanotube arrays. Although titania nanotube array films have been shown to enhance the efficiencies of both charge collection and light harvesting, it has not been possible to grow them on transparent conducting oxide glass with the lengths needed for high-efficiency device applications (tens of micrometres).

Microflowers, Nanowires, Fusiforms, Nanopetals, Nanochains, 3D dendrites, Hollow Urchin and Inverse opal [20-27] are some of the other potential TiO₂ nanostructures that have been reported so far

Tong et al.[28] conducted preliminary experiments of introducing intermediate band into the TiO₂-based photoanodes of DSSCs by doping nominal trace amount W⁶⁺ ions. The enhancement of J_{sc} and conversion efficiency might partially be attributed to the formation of intermediate band in the bandgap of TiO₂. In addition, the electron transport and electron lifetime were improved after doping W⁶⁺ in TiO₂, which also benefited the enhancement of J_{sc} and conversion efficiency. A notable improvement of the device performance was obtained when N-type W-doped TiO₂ films were applied as the photoanode of DSSCs. The short-circuit current density (J_{sc}) increased from 12.40 mA cm⁻² to 15.10 mA cm⁻², and the conversion efficiency increased from 6.64 to 7.42% when nominal 50 ppm W-doped TiO₂ was adopted.

Latini et al. [29] prepared solid solutions of scandium in anatase as semiconductor material for DSSC photoanodes by the controlled hydrolysis of titanium (IV) isopropoxide and scandium (III) isopropoxide in hydroalcoholic medium. The final powder was constituted by mesoporous anatase beads doped with Sc. Several DSSCs with photoanodes at different Sc doping were tested both under solar simulator and in the dark. The maximum efficiency of 9.6% was found at 0.2 at. % of Sc in anatase that is 6.7% higher with respect to the DSSCs with pure anatase. In conclusion, the whole picture emerging from the experimental results of the present work indicates that the DSSC performances can be improved through a careful dosage of Sc in anatase together with a strict control of each step of the cell assembly by bothering about the chemistry of materials.

Bakhshayesh et al.[30] reported a facile deposition of uniform photoanode electrodes by a novel anatase-stabilised gel for dye-sensitised solar cells (DSSCs) applications. Highly crystalline anatase-TiO₂ phase is stabilised by indium nitrate at 500°C. The anatase-stabilised DSSC has higher power conversion efficiency of 7.48% than that of unstabilised cell (6.37%).

Subramanian A et al. [31] investigated the use of Titanium nanotubes doped with boron as the photoelectrode for dye-sensitized solar cells. The materials were characterized by SEM, XRD, and UV-vis spectroscopy and their photoconversion efficiencies were evaluated. The chemical compositions of TiO₂ nanotubes (TNA) and boron doped TNA (B-TNA) were identified by the energy dispersive X-ray spectroscopy (EDS). The boron-

doped TiO₂ nanotube arrays showed an enhanced performance with a photocurrent density of 7.85±0.20mA/cm² and an overall conversion efficiency (η) of 3.44±0.10%.

Song J et al.[32] demonstrated a strategy to improve utilization of photogenerated charge in dye-sensitized solar cells (DSSCs) with fluorine-doped TiO₂ hollow spheres as the scattering layer, which improves the fill factor from 69.4% to 74.1% and in turn results in an overall efficiency of photoanode increased by 13% (from 5.62% to 6.31%) in comparison with the control device using undoped TiO₂ hollow spheres. It is proposed that the fluorine-doping improves the charge transfer and inhibition of charge recombination to enhance the utilization of the photogenerated charge in the photoanode.

Tabari-Saadi Yet al. [33] fabricated different structures of TiO₂ photoelectrodes with various arrangement modes of the layers. TiO₂ nanoparticles, synthesized by stabilizing agent free non-hydrolytic sol-gel method, are employed as the under layer, whereas carbon-doped TiO₂ hollow spheres, prepared by hydrothermally grown carbon template, are used as the scattering layer of solar cells. The nanoparticles (22 nm) have anatase structure, while 300–700 nm hollow spheres show mixtures of anatase and rutile phases. X-ray photoelectron spectroscopy confirms that carbon is doped into TiO₂ hollow spheres, resulting in a decrease in band gap energy in the range 2.96–3.13 eV compared with 3.04 eV band gap energy for the nanoparticles.

Dye-sensitized solar cells (DSSCs) were fabricated by Lin J et al.[34] by incorporating transparent electrodes of ordered free-standing TiO₂ nanotube (TNT) arrays with both ends open transferred onto fluorine-doped tin oxide (FTO) conductive glass. The high-quality TiO₂ membranes used here were obtained by a self-detaching technique, with the superiorities of facile but reliable procedures. Afterwards, these TNT membranes can be easily transferred to FTO glass substrates by TiO₂ nanoparticle paste without any crack. This showed an enhanced solar energy conversion efficiency as high as 5.32% of 24- μ m-thick TiO₂ nanotube membranes without further treatments. These results reveal that by facilitating high-quality membrane synthesis, this kind of DSSCs assembly with optimized tube configuration can have a fascinating future.

Song CB et al.[35] fabricated DSSCs based on the novel G-TiO₂NPs/TiO₂ NTs bilayer structure photoanodes by a direct mechanical mixing and spin-coating method. The results showed that the incorporation of 0.1 wt% graphene into the TiO₂ pastes increased the total solar cell efficiency by 44% due to the enhanced electron transferring.

However, higher graphene loading beyond the optimal concentration can cause the decrease of the efficiency due to the light shielding of graphene. This study will provide new insight into the fabrication and structural design of highly efficient DSSCs.

Patle LB et al.[36] prepared dip coated TiO₂ semiconductor meet enough porosity for the application of DSSC. The films deposited were prepared by 10 cycles and observed thickness about 8-9 μ m. The thickness could be increased by increasing cycles but when dried the coated TiO₂ film gets peeled off from the substrate. Even increasing the Cu concentration after 3% the TiO₂ film gets detached from the substrate. This shows that the adhesion of film is very poor for more than 10 cycles as well as Cu concentration more than 3%. Further the investigation has shown the effect of Cu doping in TiO₂ semiconductor reduces the band gap of TiO₂ up to 10%. This decrease in band gap energy of TiO₂ semiconductor corresponds to increase in open circuit voltage of the DSSC. On the other hand, with increase in Cu concentration the short circuit current and photon to current conversion efficiency of the DSSC decreases.

Mao X [37] synthesized photoanode of 3.4 nm-sized SnO₂ nanocrystals (NCs) via the hot-bubbling method. The optimal percentage of the doped SnO₂ NCs was found at ~7.5% (SnO₂/TiO₂, w/w), and the fabricated DSSC delivers a power conversion efficiency up to 6.7%, which is 1.52 times of the P25 based DSSCs.

Llanos J et al. [38] prepared Y_{1.86}Eu_{0.14}WO₆ phosphors using a solid-state reaction method. Their optical properties were analysed, and they were mixed with TiO₂, sintered, and used as a photoelectrode (PE) in dye-sensitized solar cells (DSSCs). The as-prepared photoelectrode was characterized by photoluminescence spectroscopy, diffuse reflectance, electrochemical impedance spectroscopy (EIS) and X-ray diffraction. The photoelectric conversion efficiency of the DSSC with TiO₂:Y_{1.86}Eu_{0.14}WO₆ (100:2.5) was 25.8% higher than that of a DSSC using pure TiO₂ as PE. This high efficiency is due to the ability of the luminescent material to convert ultraviolet radiation from the sun to visible radiation, thus improving the solar light harvesting of the DSSC. The photo-conversion efficiency of dye sensitized solar cells fabricated using different photoanode materials are tabulated in Table 1.

Table1: Performance of dye sensitized solar cell using various photoanode materials

Photoelectrode	Jsc(mAcm ⁻²)	Voc(V)	FF	Eff(%)	Reference
TiO ₂ nanoparticle	18.1	0.91	0.78	13	16
TransparentTiO ₂ nanotube	18.5	0.77	0.64	9.1	17
TiO ₂ nanorod	16.52	0.772	0.746	9.5	18
TiO ₂ nanotube on Ti coated TCO	15.8	0.73	0.59	6.89	19
TiO ₂ microflowers	1.255	0.573	0.26	3.72	20
TiO ₂ nanowires	17.5	0.75	0.47	6.2	21
TiO ₂ fusiform	10.13	0.68	0.57	4.68	22
TiO ₂ nanopetals	10.54	0.7	0.7	5.3	23
TiO ₂ nanochains	15.95	0.75	0.61	7.46	24
TiO ₂ 3D dendrites	14.52	0.72	0.68	7.2	25
TiO ₂ hollow urchin	17.17	0.612	0.647	7.16	26
TiO ₂ inverse opal	6	0.75	0.62	2.8	27
TiO ₂ doped with tungsten	15.1	0.73	0.67	7.42	28
TiO ₂ doped with scandium	19.1	0.752	0.68	9.6	29
TiO ₂ doped with indium	16.97	0.716	0.61	7.48	30
TiO ₂ doped with boron	7.85	0.66	0.66	3.4	31
TiO ₂ doped with flourine	11	0.754	0.76	6.31	32
TiO ₂ doped with carbon	20.38	0.73	0.57	8.55	33
ONT/FTO	10.65	0.7	0.7	5.32	34
TiO ₂ NPs/TiO ₂ NTs	16.59	0.69	0.56	6.29	35
TiO ₂ doped with copper	6.84	0.591	0.56	2.28	36
7.5%SnO ₂ doped TiO ₂	14.53	0.79	0.58	6.7	37
TiO ₂ , Y _{1.86} Eu _{0.14} WO ₆	12.3	0.757	0.43	3.9	38

IV. APPLICATION OF DSSC

The dye-sensitized solar cells market is bifurcated on basis of application into portable charging, BIPVs, BAPVs, embedded electronics, outdoor advertising, solar chargers, wireless keyboards, emergency power in military, AIPVS, and others (light intensity meters and consumer appliances).

BIPVs category dominated the dye-sensitized solar cells market, contributing a share of more than 20.0%, in terms of value, in 2017. This can be attributed to the properties of dye-sensitized solar cells such as low-weight, easy installation, and a high performance-to-weight ratio. These enable easy installation and retrofitting of these

cells in building envelopes like rooftops, louvres, glazed facades, and ceilings.

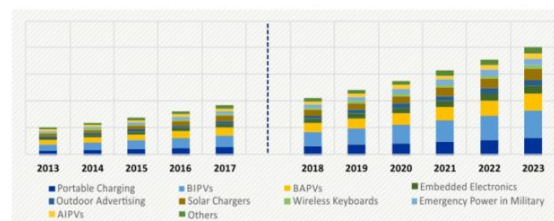


Fig3:DSSC Market by application, square meter(2013-2023)

The low-cost fabrication of dye-sensitized solar cells and no requirement of structural support material for modules such as beams envisages economic advantage in the

building construction, due to possibility of price substitution of a building element from the price of panels. These factors are likely to drive the market in future.

Low power production efficiency of dye-sensitized solar cells can be viewed as the key factor restraining the dye-sensitized solar cells market growth. Silicon-based cells have a higher cost, but they are more efficient and have a power production efficiency of 12–15%, which is a slightly higher than the efficiency of these cells (which is around 11%). The global annual growth rate in DSSC market by application in a 10-year period is shown in Figure 3.

V. CONCLUSION

Dye-sensitized solar cells (DSSCs) have been widely studied due to several advantages, such as low cost-to-performance ratio, low cost of fabrication, functionality at wide angles and low intensities of incident light, mechanical robustness, and low weight. This article provides a comprehensive summary of the techniques and modifications done in the TiO₂ photoanode layer to improve the performance of DSSCs. In particular, this review highlights a huge pool of studies that report improvements in the efficiency of DSSCs using TiO₂, which exhibit better electron transport. However, a brief explanation has been given to greater understand the working and components. The application of these nanostructured photoanode materials and their impact on the device efficiency has been described in detail. Research and innovation in the dye-sensitized solar cells market help in mass commercialization of these cells.

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