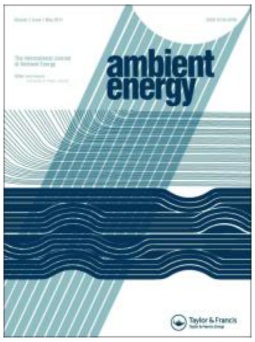

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REVIEW

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Recent developments on green synthesised nanomaterials and their application in dye-sensitised solar cells

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ABSTRACT
The development of highly efficient dye-sensitised solar cells (DSSCs) with good photovoltaic parameters has attracted tremendous attention due to global energy crises. Many novel approaches have been developed to manufacture high-performance DSSCs with an amendment in counter electrodes, electrolytes, dyes, and photoanode materials. Metal nanoparticles and nanostructured metal oxides have widely been used as photoanode material in DSSCs. The ability of nanomaterials to improve electron transport leads to enhanced electron collection efficiency, improving the productivity of DSSCs. Hence, the green synthesis of nanomaterials and their applications in energy generation for a sustainable future is an active area of research. Nanoparticle synthesis using biological agents is cost-effective, less laborious, non-toxic, and environmentally benevolent compared to conventional synthesis routes. The paper gives a critical overview of photoanode materials with a significant emphasis on green synthesised nanomaterials. Also, photoanode modifications and their effect on the performance of DSSCs have been reviewed.

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KEYWORDS
Dye-sensitised solar cells; green synthesis; nanoparticles; natural extracts

1. Introduction

The energy demand is constantly increasing globally due to pullulating population and industrialisation. Due to the industrial development and growth in population, the expected global energy demand will be doubled by the year 2050, specifically in developing countries. Hence, there is a high potential for renewable energy generation from various sources such as wind, solar, biomass, small hydro, and cogeneration bagasse.

Amongst all other abundant and non-polluting renewable energy sources, it is expected that solar energy – will – play a vital role as a future energy source. About 3×10^{24} Joule/year energy in the form of sunlight reaches the earth's surface, nearly 10^4 times more than the world's energy consumption. Thus, the photovoltaic industry frequently requires new scientific and technical solutions for the successful global commercialisation of renewable energy. Still, the production cost, repeatability, reproducibility, size, and stability of devices, and also their efficiencies, have yet to proceed towards sustainable and green technology in the future (Raj and Prasanth 2016). Thus, to enhance the efficiency of solar cells, substantial efforts have been made to fulfil the energy demands.

Among these, thin-film solar cells and silicon solar cells gained significance as first and second-generation energy devices. But the manufacturing cost of these cells is still high, and after use, hardware recycling is still having a problem and producing environmental pollution. Hence, there is an enormous demand for sustainable, organic, and inexpensive photovoltaic devices that imitate plants in energy conversion.

Hence, considerable efforts have been made to improve the efficiency of third-generation thin-film solar cells, known as dye-sensitised solar cells (DSSCs). Compared to first – and second-generation solar cells, most DSSCs are plant-based, inexpensive, non-toxic and do not harm the environment. Plant dyes act as sensitisers that absorb sunlight and convert it into electrical energy (Gokilamani et al. 2014; Safie et al. 2017; Datta et al. 2020; Ganta, Combrink, and Villanueva 2019).

Even though the electricity generation capability of organic dyes has been known since the late 1960s, the first attempt to generate electricity from dye-sensitised semiconductor film of ZnO was sensitised with Chlorophylls (Tributsch 1972), and that's why they are sometimes referred to as 'Artificial Photosynthesis'. The first embodiment of modern-day, dye-sensitised Solar Cell (DSSC) dates back to the late 1980s (Augustynski et al. 1988). Gratzel et al. made a breakthrough cell efficiency of 7% on dye-sensitised solar cells (DSSCs), with colloidal TiO_2 photoanode film in 1991. This facilitated in-depth research on these types of solar cells, which were regarded as one of the best potential renewable energy sources due to their marvellous properties such as semi-transparency and colourful appearances, ease of fabrication, low cost., high efficiencies, and possible plasticity, particularly under indoor illumination or dim light irradiation. Before 1999, there were few reports on DSSCs; however, after

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2000, the cell efficiency of DSSCs of $>10\%$ with mesoporous TiO_2 film was recorded (Hagfeldt and Grätzel 2000; F. Gao et al. 2008); thus, the number of studies on DSSCs exploded. The DSSC with the zinc porphyrin dye and Co (II/III) tris (bipyridine)-based redox electrolyte attained an excellent efficiency record of 13% in 2014 (Mathew et al. 2014). Soon after, with the co-sensitisation of two metal-free organic dyes and a Co (II/III) tris(phenanthroline)-based redox electrolyte, a DSSC achieved the highest efficiency record of 14.30% (Kakiage et al. 2015).

These findings further encouraged research studies on each component of DSSCs, including photoanode, photosensitiser, electrolyte, and counter electrode, to develop economical photovoltaic devices to compete in the solar cell market. The DSSC is an emerging technology which can be applied in many electronic markets comprising smart homes, smart buildings, wireless sensor networks, and smart cities (RapidFire Consulting. 3gsolar. 2018). However, the life span and power conversion efficiency of DSSC is unrivalled with the silicon solar cell. As a result, new materials must be developed to make this technology viable, usable, and cost-effective. The power conversion efficiency of DSSC is determined by light-harvesting efficiency, charge collection efficiency, light scattering ability, and charge recombination rate. These characteristics are straightforwardly allied with the photoanode of DSSCs.

The photoanode material is an essential element of DSSCs. The photovoltaic performance of photoanode is greatly influenced by the crystallinity and morphology of the nanomaterial being used as a photoanode. Chemical and physical methods are used to manufacture photoanodes with different morphologies. But, these are expensive and use toxic and hazardous materials during nanomaterials synthesis. On the other hand, the biological methods, including bio-sources such as bacteria, fungi, yeast, and plant extract as a reducing agent, are very cost-effective and do not comprise the use of toxic and hazardous materials during the processing. Therefore, several researchers are adopting these green methods for the synthesis of photoanode materials. Thus, the major focus of the current manuscript is on the application of green synthesised photoanode materials and their effect on the performance of DSSCs. The purpose of this review is to understand better the impact of photoanode material properties on the photovoltaic performance of DSSCs. In this review, the important photoanode materials, including green synthesised nanomaterials, are discussed. Also, the progress in DSSC device efficiency has been reviewed in detail. Finally, the challenges and prospects of DSSCs are discussed in the summary of this review.

2. Construction and working principle of DSSC

Figure 1 shows a sandwiched structure of Dye-Sensitised Solar Cell (DSSC) between transparent conductive materials like indium tin oxide (ITO) glass and fluorine-doped tin oxide (FTO) glass, a mesoporous photoanode with dye-sensitised titanium dioxide (TiO_2) film, an electrolyte comprising iodide/triiodide ($I^-/I3^-$) redox couple, and a counter electrode with platinum (Pt) catalyst. The operating cycle of DSSC is briefed as follows:

The dye molecules present on the surface of the nanocrystalline TiO_2 photoanode absorb the incoming photon. An electron from a molecular ground state S^0 is excited to a higher excited state S∗ as these particles absorb an incoming photon. The excited electron is injected into the TiO_2 particle's conduction band, oxidising the dye molecule S^+. The injected electron passes through the porous nanocrystalline structure to the glass substrate's transparent conducting oxide layer (negative electrode, anode), then to the counter-electrode through an external load (positive electrode, cathode). The electron is transferred to triiodide in the electrolyte at the counter-electrode, resulting in iodine. The cycle is completed by reducing the oxidised dye by the iodine in the electrolyte (Ehrmann and Blachowicz 2019).

The functioning sequence can be summarised in chemical reaction terminology as (Matthews, Infelta, and Grätzel 1996):

Anode: $S + h\nu \rightarrow S^*$ Absorption
$S^* \rightarrow S^+ + e^-(TiO_2)$ Electron injection
$2S^+ + 3I^- \rightarrow 2S + I_3^-$ Regeneration
Cathode: $I_3^- + 2e^-(Pt) \rightarrow 3I^-$
Cell: $2e^-Pt + h\nu \rightarrow e^-(TiO_2)$

The performance of DSSC is measured with the following parameters:

(a) Open circuit photovoltage (V_{oc}): Open circuit voltage is the voltage measured at zero current in the cell.
(b) Short circuit photocurrent (I_{sc}): It is the photocurrent measured at zero voltage. In general, it is presented in the form of the short circuit current density (J_{sc}) defined as the short circuit photocurrent divided by the active cell area.

$$J_{sc} = \frac{J_{sc}}{\text{Area}}$$

(c) Fill factor (FF): It is the ratio of the maximum power output (P_{max}) to the product of J_{sc} and V_{oc}.

$$FF = \frac{P_{max}}{J_{sc} \times V_{oc}} = \frac{I_{mpp} \times V_{mpp}}{J_{sc} \times V_{oc}}$$

where I_{mpp} and V_{mpp} are the photocurrent and photovoltage corresponding to the maximal power point, respectively, in the J–V curve.

(d) Energy conversion efficiency (η): It is the ratio of maximum power output (P_{max}) to the incident radiation power (P_{in}) on the solar cell surface. It is the product of V_{oc}, J_{sc} and FF divided by the incident irradiation power.

$$\eta = \frac{P_{max}}{P_{in}} \times 100$$

Basic characterisation techniques for DSSC are Photocurrent density-photovoltage (J–V) measurement. The J–V measurement can be performed under light illumination of 100 mW/cm^2 using AM 1.5 solar simulator.

A typical J–V curve is shown in Figure 2. During the J–V measurement, the open circuit photovoltage (V_{oc}), short circuit photocurrent (J_{sc}), fill factor (FF) and efficiency (η) can be determined.

3. Photoanode materials

Photoanodes act as electron collectors and are made of semiconductor metal oxide material. The overall power conversion

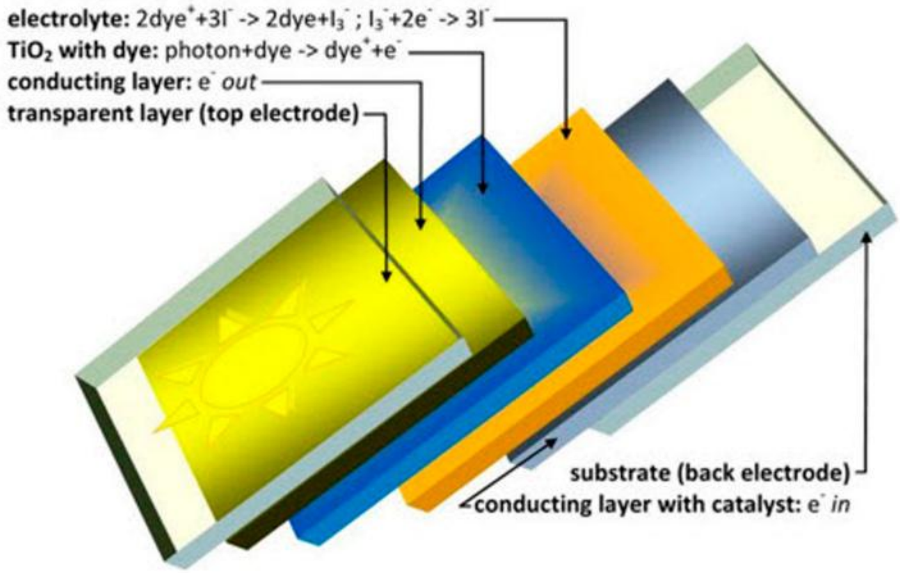

Figure 1. DSSC principle of operation and basic technological parts (Ehrmann and Blachowicz 2019).

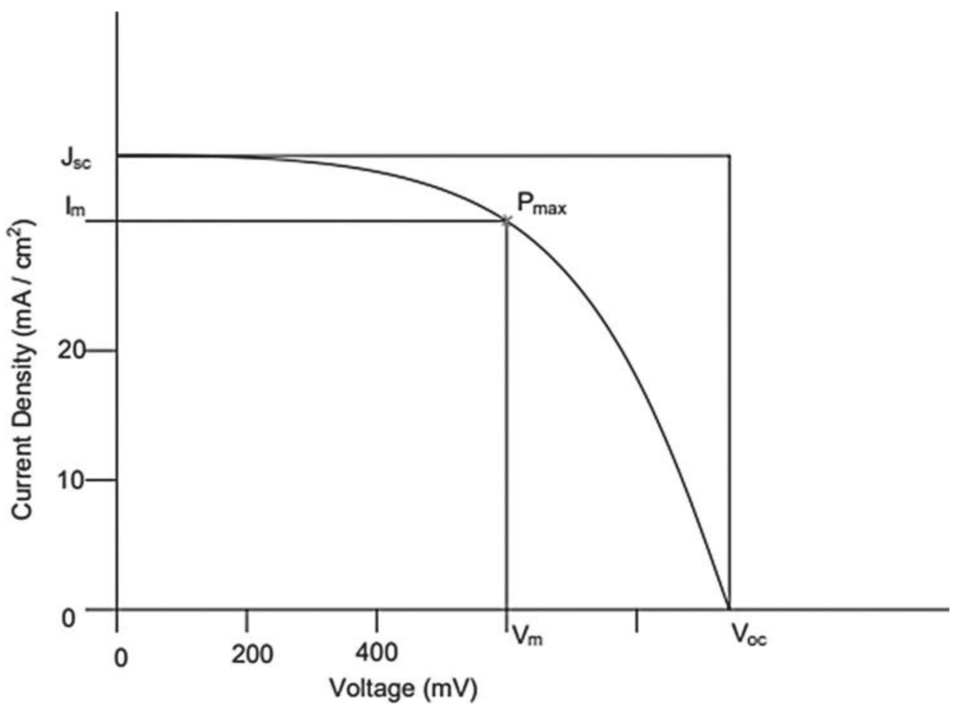

Figure 2. J–V curve of DSSC.

efficiency of DSSCs depends on the bandgap and morphology of photoanode material. Its function is to adsorb dye molecules and transfer electrons to the external circuit (Karim et al. 2019; Pallikkara and Ramakrishnan 2021). An efficient photoanode material must acquire the following characteristics: (i) high surface area for maximum dye absorption, (ii) fast electron injection from the dye and fast electron transfer to an external circuit, (iii) pore size should be precisely altered to assist optimum diffusion of dye and electrolyte, (iv) high resistance to photo-corrosion, (v) potential in scattering or absorbing the sunlight for efficient dye functioning, (vi) a good electron acceptor, and (vii) optimal interface linkage with a conductive layer on the substrate and with the dye molecules.

The photoanode design improvements and photon–electron conversion process is vital for constructing high-performance DSSCs. Generally, while developing DSSC, the mentioned properties of the photoanodes are considered.

The photoanode comprises of Transparent conducting oxides (TCOs) substrates, namely Indium tin oxide (ITO) and Fluorine tin oxide (FTO) that exhibit adequate transparency and electron collection rate. However, the cost of TCOs is comparatively high. The electronic properties of sensitisers are accountable for

Table 1. Effect of different photoanode materials on efficiency of DSSC.

Metal oxides	Band gap (eV)	Best efficiency (%)	Characteristics	Limitations	Refs.
TiO_2	3.2	14.3	(i) It is less expensive, non-toxic, Chemical stability and biocompatible. (ii) Higher surface to volume ratio. (iii) Improved photo stability. (iv) Wide range of UV absorption (v) A high degree of charge transfer.	(i) Limited electron mobility.	Gnida et al. (2021), Kakiage et al. (2015)
ZnO	3.3	7.5	(i) Efficient, Heterogeneous, Reusable, and Ecofriendly (ii) High electron mobility.	(i) Dye-dependent performance. (ii) Complexation with dyes.	Vittal and Ho (2017)
SnO_2	3.6	8.23	(i) High exciton binding energy. (ii) N-type semiconducting metal oxide with a wide band gap.	(i) Electron recombination occurs at a faster rate. (ii) Less adsorption of dye	Banik, Ansari, and Qureshi (2018), Li et al. (2014)
Nb_2O_5	3.2-5	$> 6\%$	(i) A larger bandgap. (ii) Excellent electron injection efficiency and chemical stability.	(i) Dye loading sites are reduced.	Panetta et al. (2017), Nunes et al. (2019)
WO_3	2.6-3	1.46	(i) A high degree of stability. (ii) Excellent carrier mobility.	(i) The surface is acidic. (ii) A stronger conduction band edge.	Patil et al. (2021), Zheng, Tachibana, and Kalantar-Zadeh (2010)
In_2O_3	3.6	< 2	(i) Higher electron lifetime. (ii) Broad bandgap with large surface area. (iii) Hollow porous structure improves dye loading.	(i) More positive potentials of bands.	Mahalingam and Abdullah (2016)
$SrTiO_3$	4.15	0.58	(i) Active under UV region (ii) Wide bandgap (iii) Restrains the photogenerated charge carriers recombination	(i) Low electrical conductivity	Gholamrezaei et al. (2016)
Zn_2SnO_4	3.6	3.8	(i) High electron mobility (ii) Wide bandgap	(i) Short electron diffusion length	Tan et al. (2007)
$BaSnO_3$	2.9-4	5.2	(i) Predominant electron collection (ii) Wide bandgap (iii) High electron mobility	(i) Threading dislocation limits electron mobility	Najafabadi, Ahmadi, and Ghanaatshoar (2021), Kim et al. (2013)
$CoTiO_3$	2.25	7.67	(i) Increased light scattering (ii) Reduced inner surface area and dye loading (iii) Bandgap constriction	(i) Uncontrolled sizes and morphologies	Aynehband et al. (2019)

Photo-electron generation. Thus, augmenting the band structure of sensitisers can magnify the photon–electron conversion process and intensify the extinction coefficients in the near-IR region (Hagfeldt et al. 2010).

Since electron mobility is closely linked to the semiconducting layer, small electron injection barriers between the sensitiser and the semiconducting layer, a long diffusion distance, and quick transferability are desired (Hagfeldt et al. 2010). The total kinetics of electron removal and recovery determine the rate of charge carrier recombination (Hagfeldt et al. 2010; Halme et al. 2010). During these processes, a large amount of energy is generated, and efficiency is lost. As a result, photoanode research is still based on exploring solutions to existing challenges.

Various Semiconducting metal oxides such as TiO_2 (Gnida et al. 2021; Shakeel Ahmad, Pandey, and Rahim 2017), ZnO (Anta and Guille 2012; Efa and Imae 2019; Marlinda et al. 2019), SnO_2 (Z. Li et al. 2014), Nb_2O_5 (Ou et al. 2012; Panetta et al. 2017; Nunes et al. 2019), WO_3 (Zheng, Tachibana, and Kalantar-Zadeh 2010; Patil et al. 2021), In_2O_3 (Mahalingam and Abdullah 2016), $SrTiO_3$ (Gholamrezaei et al. 2016), Zn_2SnO_4 (Tan et al. 2007), $BaSnO_3$ (Kim et al. 2013; Najafabadi, Ahmadi, and Ghanaatshoar 2021; Bhojanaa and Pandikumar 2021) and $CoTiO_3$ (Aynehband et al. 2019) have been investigated as photoanode materials with improved photo-corrosion resistance and DSSC's efficiency. Table 1 shows the summary of different photoanode materials and their performance in DSSCs.

DSSCs using metal oxides as photoanodes, such as In_2O_3, WO_3, Nb_2O_5, and $SrTiO_3$, have shown lower quality performance than TiO_2-based DSSCs, as revealed in the research papers discussed above. They have, however, contributed to a solid and fundamental understanding of the process in DSSC systems. At present, TiO_2 nanoparticle recorded the highest efficiency (14.3%) and has been the best photoanode material in DSSCs (Kakiage et al. 2015). Moreover, TiO_2 is a cheap, non-toxic, widely available, biocompatible material and has a wide energy bandgap (Frank and Brudvig 2004; Green et al. 2005). TiO_2 exists in three forms: rutile with an energy bandgap of 3.05 eV, anatase with an energy bandgap of 3.20 eV, and brookite with an energy bandgap of 3.28 eV (Lau et al. 2014; Valencia, Marín, and Restrepo 2010). Anatase forms have a large surface area and a large electron coefficient. As a result, it is recognised as an efficient candidate compared to rutile and is strongly recommended for DSSCs. Brookite is not recommended due to its complicated synthesis process (N. G. Park, Van De Lagemaat, and Frank 2000). The major drawback of the TiO_2 based photoanode in DSSC is its random electron transport, which leads to the electron–hole recombination process, affecting the overall performance. This deficiency of the TiO_2-based DSSCs can be overcome by doping with metals, non-metals, semiconductors and carbon materials (Saravanan et al. 2017).

To date, various techniques for improving photoanode efficiency have been attempted, including doping with metal atoms, nanocomposites (Lim et al. 2015; Pandikumar, Saranya,

Figure 3. Schematic representation of modification techniques for metal oxide photoanode.

and Ramaraj 2012), light scattering layer (Chou et al. 2012; H. H. Wang et al. 2013; Mustafa and Sulaiman 2021), surface treatment with additional metal oxides (Ako et al. 2016; Basu et al. 2016; Sun et al. 2016; Xuhui et al. 2014), hierarchical structures or $TiCl_4$ post-treatment (Sommeling et al. 2006; Huang et al. 2012) and photosensitiser modifications (Carella, Borbone, and Centore 2018; Maddah, Berry, and Behura 2020) (Figure 3). The best efficiency recorded for various metal oxides based on the energy bandgap, their characteristics, and limitations have been summarised in Table 1.

As shown in Figure 3, researchers worldwide are working on modifying the properties of semiconducting materials used in photoanodes (such as TiO_2, ZnO, SnO_2, etc.) (Pandikumar, Saranya, and Ramaraj 2012). The most effective technique to modify the property of semiconductors is bandgap tuning. The bandgap is altered by doping with metal atoms, resulting in changes in properties such as band positions, electrical conductance, charge recombination rates, and trap or defect level distribution (Zhao et al. 2012; Wang and Teng 2009). The performance of DSSCs is determined by the metal atoms used to dope them. Several research studies on TiO_2 nanostructures have been conducted using doping atoms such as Au, V, Sc, Sn, Fe, Cu, Mn, Co, Ta, Zn, Cr, Nb, La etc. (Du et al. 2012; Liu et al. 2011; Seo et al. 2013; Latini et al. 2013; Ni et al. 2019; Ako et al. 2015; Patle, Huse, and Chaudhari 2017; Shalan and Rashad 2013; Ghosh et al. 2012; Hara et al. 2013; Wang and Teng 2009; Kim et al. 2008; Chandiran et al. 2010; Zhang et al. 2010). As per the literature, TiO_2 photoanode doped with gold nanoparticles (Au NPs) attained a maximum efficiency of 8.13% (Du et al. 2012; Nahm et al. 2011; Sahu, Gordon, and Tarr 2012a; Sahu et al. 2012b). It has been reported that the doping of metals into the semiconducting metal oxides increases the solar light absorption capacity and the capacity to inject electrons. The optimum efficiency for DCCS with V-doped TiO_2 was found to be 6.81%. (Liu et al. 2011; Seo et al. 2013). The electron mobility and the flat band potential of TiO_2 were found to improve upon doping with Sn, leading to improved DSSCs performance with 8.75% efficiency (Ni et al. 2019). Latini et al. researched the role of Scandium (Sc) doping on titania beads for its application as a photoanode material in DSSC. As the size of Sc^{3+} is similar to that of the Ti^{4+}, substituting Sc^{3+} for Ti^{4+} produces holes in the valance band of TiO_2, making Sc^{3+} an excellent dopant in titania beads, improving the efficiency of DSSC up to 9.6% (Latini et al. 2013). In another report, Ga doping resulted in a negative shift of the CB and repressed charge recombination, improving photovoltaic performances (Duan et al. 2015). Ako et al. investigated three metals as dopants in TiO_2 nanocrystalline powders: tin, iron, and copper. According to their findings, $SnTiO_2$ > pure TiO_2 > Cu-TiO_2 > > Fe- TiO_2 is the order of power conversion efficiency. Better efficiency of Sn-TiO_2's was due to a modest increase in defect concentrations, which resulted in less recombination at trapped sites (Ako et al. 2015). Another study based on transition metals such as Cu^{2+}, Fe^{3+}, and Mn^{2+} doped TiO_2 by Patle et al. showed that the photoconversion efficiency is higher for Mn^{2+} doped TiO_2 based photoanode as compared to the undoped and Cu^{2+} and Fe^{3+} doped TiO_2 (Patle, Huse, and Chaudhari 2017). Materials derived from carbon (such as graphene and carbon nanotubes), noble metals (gold and silver) with plasmon effect, and transition metal oxides are frequently used to form nanocomposites of TiO_2 photoanodes for DSSCs (Alagarsamy Pandikumar et al. 2016). The most widely used carbon-based materials are graphene and carbon nanotubes due to excellent electron mobility, a large surface area, and excellent mechanical properties (Singh et al. 2011; Kama and Prashant 2007). Graphene can lower resistance and charge recombination rates at the electrolyte–electrode interface. In addition to this, it also enhances the electron transfer from the semiconductor films to the conductive substrates. Integrating graphene in photoanodes also improves light-harvesting, thus increasing the number of photo-induced electrons (Wu and Ting 2015; Fan, Yu, and Ho 2017). Fan and co-workers invented a TiO_2 nanosheet/graphene composite film photoanode for DSSCs with 0.75 wt% graphene. The conversion efficiency of the composite film was increased by 25% compared with the bare TiO_2 nanosheet photoanode (Fan, Liu, and Yu 2012). Carbon nanotubes reduce the charge recombination and boost electron transport from the films to the FTO substrates and result in enhanced conversion efficiency (MacDonald et al. 2015; T. Y. Lee, Alegaonkar, and Yoo 2007). Hence, Carbon-based materials can be used to increase DSSC performance significantly.

Because of the two-dimensional structure of the former, graphene performs better than carbon nanotubes in terms of durability when used for DSSCs. The weight per cent of graphene or carbon nanotubes in the photoanode must be kept to less than 1 wt% in the film. High loadings of graphene or carbon nanotubes will protect the dyes from visible light adsorption, resulting in a decrease in the number of photogenerated electrons and the efficiency of the DSSCs.

The surface modification of semiconductor photoanode by mono or multication metal oxides is considered an effective strategy to reduce electron recombination and improve DSSC performance. The surface modification of metal oxide with $TiCl_4$ is popularly being used among several other surface modification techniques. It results in the formation of the TiO_2 layer on the semiconductor photoanode, improving the electronic properties of semiconductor electrodes.

The TiO_2 based DSSCs showed improved photovoltaics because there is an increased active surface area resulting in increased dye adsorption and thus the enhancement in the light-harvesting property. Secondly, there is a downward shift in the conduction band of $TiO_{2,}$ which results in the amplified electron injection and declines charge recombination (Sommeling et al. 2006). Numerous methods have been applied in surface treatments to improve all properties of metal oxide photoanodes. For thin film deposition of photoanodes, physical and chemical methods are available. Among various chemical methods, chemical vapour deposition (CVD) (Murakami et al. 2004), plasma-enhanced CVD (Li et al. 2016), sol-gel method (Lee, Chae, and Kang 2010), and atomic layer deposition (Park et al. 2010) are significantly used. While, physical thin film deposition methods include sputtering (Zatirostami 2020), pulsed laser deposition (PLD) (Lee et al. 2009), cathodic arc deposition (Aramwit et al. 2014), thermal evaporation (Yan et al. 2012), and others. Among these, atomic layer deposition (ALD) is the most advanced method for surface treatment and is widely used today.

The treatment of TiO_2 with large bandgap metal oxides such as Ga_2O_3, Al_2O_3, $SrCO_3$, La_2O_3, Y_2O_3, NiO, MgO, and Nb_2O_5 was very effective to enhance the efficiency of DSSCs (Chandiran et al. 2012; X. Gao et al. 2013; S. Wang et al. 2012; Yu et al. 2012; Kim and Moon 2012; Chen et al. 2001; Lin et al. 2014; Yao et al. 2016; Wu et al. 2008). These metal oxides created a barrier layer that reduces electron recombination and leads to increased efficiency. This add-on layer may affect dye loading and electron injection, resulting in decreased power conversion efficiency. However, these flaws are minor, and the benefits outweigh them; hence its research study has greater significance.

The anatase TiO_2 scattering layer of larger particles is more effective at harvesting light, resulting in improved Sn doped TiO_2 up to 9.43% (Ni et al. 2019). Apart from TiO_2 nanosheet/graphene composite film photoanode, Li et al. reported the preparation of novel multishell SiO_2@Au@TiO_2 (SAT) microspheres embedded with Au NPs and its application as photoanode material for DSSC. The excellent light reflection/ scattering property of the SiO_2@Au@ TiO_2 (SAT), along with the surface plasma resonance (SPR) properties of Au NPs, enhanced the efficiency of DCCS as high as 7.75%, remarkably higher than those of the conventional DSSC by 23.7% and 28.0%, respectively (Li et al. 2009).

In addition to this, light-scattering materials are added to the photoanode film. It is an effective and widely used method for enhancing the photoanode's light absorption and thus improving the performance of DSSCs. When incident light travels through the photoanode film, scattering material particles have a larger size and can reflect and/or scatter light. It produces a longer light path, increased optical absorption, and improved light-harvesting properties. Light-scattering materials are typically applied in photoanodes using a 'double-layer structure' or a 'mixture structure to improve light scattering and dye loading'. The conventional TiO_2 with large particles scattering layer has been suggested to replace by bilayer structure with one-dimensional material overlayer with low dye loading ability and TiO_2 nanoparticles as the under-layer (Bakhshayesh et al. 2013; Qiu, Chen, and Yang 2010). The utmost conversion efficiency of 9.43% was achieved by introducing the bifunctional hollow spheres of TiO_2 as the scattering layer (Koo et al. 2008).

In recent years, hierarchical materials of primary nanostructures, such as ZnO or TiO_2 nanocrystallites have been increasingly used in DSSC photoanodes. These primary structures form a secondary structure and are either spherical in shape, low dimensional, or three-dimensional (Liu et al. 2016). Dou et al. (2012) reported the synthesis of Zn doped SnO_2 nanocrystals for photoanode applications with PCE of 2.07% with V_{oc} of 0.67 V. The modification of photoanode with $TiCL_4$ resulted in the decrease dye adsorption; however, the efficiency of the DSSCs had been found to increase. Hierarchical materials can supply larger specific surface areas to load sufficient dyes by the primary nanostructure compared with conventional nanoparticles and one-dimensional nanoarrays and exhibit greater light-harvesting efficiency, reduced electron recombination, and facilitated diffusion.

4. Green synthesised nanomaterials for photoanode

Green synthesised nanomaterials have gained significant importance in diversified fields such as biodiesel manufacturing (Varghese, Joy, and Johnson 2017), improving photocatalysis effect (Manjari et al. 2017), water purification (Peternela et al. 2018), solar cells efficiency improvement (Sharma et al. 2015), supercapacitors conductivity (Pendashteh, Mousavi, and Rahmanifar 2013), improving electrocatalysis (Ramachandran, Arya Nair, and Yesodha 2019) etc.

Two distinct essential standards of combination, namely the top-down and bottom-up approaches, have been scrutinised to obtain nanomaterials with the desired dimensions, shape, and functionalities. Earlier, nanomaterials/nanoparticles were set up through assorted scope of novel syntheses like ball milling, lithographic techniques, etching, and sputtering (Cao and Wang 2004). A bottom-up approach includes several methods like Hydrothermal (Santhi et al. 2020), modified Solvothermal (Ramakrishnan et al. 2018), Chemical vapour deposition (Huynh et al. 2020), Sol-gel processes (Uribe-López et al. 2021), Spray pyrolysis (Tiyyagura et al. 2020), Laser synthesis (Amendola et al. 2020), and Microwave-assisted synthesis (Phuruangrat et al. 2019), which produce nanoparticles from simpler molecules. Excitingly, the morphological constraints of nanoparticles (e.g. size and shape) can be moderated by changing chemical concentrations and reaction conditions (e.g. pH and temperature). However, if these amalgamated nanomaterials are exposed to real-world applications or conditions, then they can overcome challenges such as (i) stability in different environmental conditions, (ii) lack of understanding of modelling factors, (iii) bioaccumulation of nanoparticles and associated toxicity (iv) extensive analysis demands, (v) need for specialised operators, (vi) issues with system assembly and structures, and (vii) recycling/reuse/regeneration (Singh et al. 2018).

In the current scenario, the prudent properties of nanomaterials should be subsequently enhanced to meet the foregoing research. On the other hand, these constraints are creating new and exciting opportunities in this emerging field of research. To overcome these limitations, modern materials science and technology research and development are focusing on a new age of 'green synthesis' methods. The combination of metal and metal oxide nanoparticles using a novel green synthesis approach is favourable and less toxic than the physical and chemical processes that are toxic to the environment, expensive, require more energy, and exertive and time-consuming process (Mbonyiryivuze et al. 2015).

Various resources are available for the green synthesis of NPs, such as plants and their products, bacteria, fungi, algae and viruses. There is an increase in research publications every year in this area, as indicated in Scopus keyword search for metal nanoparticles and plant extract. There are approximately 468 publications reported in the year 2020 (Dikshit et al. 2021). Thus, increased interest is seen in this area, and significant growth is also observed.

It is observed that various biomolecules perform an important role as reducing capping agents in metal reduction. Variation in the stability and reducing potential of biomolecules leads to variation in size, shape and properties of NPs. There are multiple biomolecules present in plant extracts like amino acids, alkaloids, aldehydes, flavones, ketones, proteins, phenolics, polysaccharides, saponins, tannins, terpenoids, and vitamins. Complete reduction time for NPs from plant extract ranges from a few hours to 48 hrs which is quite faster as compared to microorganism based reduction, which normally takes 24–120 h (Makarov 2014). As per past studies, plant-based biosynthesis of Ag NPs shows better production rate, size and morphological properties compared with other biological methods (Makarov 2014). Hence, the plant-based synthesis of NPs can be seen as a better and sustainable alternative among the biological techniques for physical and chemical synthesis of NPs.

DSSC is made possible by optimising the key components: counter electrodes, iodine electrolyte, photoanode (TiO_2, ZnO, and SnO_2) and dye as sensitisers. Various authors and researchers have focused on improving every component of the DSSC to expand power conversion efficiency (PCE). The photoanode is important in determining the performance of DSSCs (Kakiage et al. 2015). A semiconductor or photoanode can effectively increase the performance of DSSCs if it has a large surface area and thickness, which can enhance the rate and impact of dye loading onto it (Rothenberger, Comte, and Gra 1999). The use of nanomaterials as photoanodes has been reported as the best choice in harvesting light and extracting charges, thereby resulting in enhanced photovoltage, photocurrent, and FF (Panchal, Shah, and Padharia 2015). Apart from TiO_2 as a photoanodes, zinc oxide has also been adopted as a photoanode having the second-best performance after TiO_2 (Keis et al. 2002; El-Agez et al. 2012). For better DSSCs performance, overall light absorption across the solar spectrum is difficult to achieve. To increase the light-absorbing capacity of the photoanode, dye molecules are adsorbed on the surface of the nanostructured oxide films (Kakiage et al. 2015). The performance of DSSC also depends on the morphology, phase compositions, and other properties (Alivov and Fan 2009; Li et al. 2009). Improving the performance of TiO_2 has been achieved by mixing TiO_2 with different sizes, phase composition, and morphology (Zhang et al. 2008; Liu, Enache-Pommer, and Aydil 2010).

From prior exploration, it is well established that the TiO_2, *n*-type semiconductor, to be one of the promising photoanode materials with very distinct properties; it has excellent bandgap characteristics ($E_g - 3.2$ eV) with dye, phenomenal charge transfer capability, is recyclable, and higher refraction indices (Cormier et al. 2018; Francis et al. 2011). Different synthetic procedures are available to acquire TiO_2 nanoparticles, e.g. green synthesis approach, chemical extraction, physical processing, etc. (Taheriniya and Behboodi 2016; Sundrarajan and Gowri 2011). The physical processing involves excessive pressure and temperature treatment to get the required accuracy and nanomaterial size.

In chemical processing-based synthesis, the use of toxic chemicals is considered hazardous for the environment and the person handling it. Besides, green synthesis is socially accepted as the best eco-friendly, economical as it doesn't require high temperature and pressure processing, critical chemical treatment (Taheriniya and Behboodi 2016; Mohan Yedurkar, Maurya, and Mahanwar 2016).

Various stabilising extracts and solvents are utilised to combine TiO_2 nanoparticles, preventing them from agglomerating and to bringing down particle size to increase the performance characteristic of DSSC. A simple sol-gel technique for green synthesis was utilised for *Bixa orellana* seed substrate as the capping agent regulates the nanoparticle size. It offers several advantages over the conventional sol-gel approach, such as low-cost synthesis, formation of pure anatase phase, enhanced surface area, and high energy conversion efficiency that make it a potential approach in advanced DSSCs. The comparison of lab-processed TiO_2 (TNP) was compared with the plant seed developed TiO_2 nanoparticles (G-TNP). It is discovered that the photocurrent transfer tendency of the DSSC is more prominent with mesoporous G-TNP film as compared to TNP films which bring about a conversion efficiency of around 3% (Maurya et al. 2019).

Contrarily, an attempt was made to synthesise the mixed anatase and rutile TiO_2 by using the extract of naturally occurred *Phellinus-mushroom* and photoelectrical energy conversion efficiency of 3.8% is achieved (Ekar et al. 2017). This signifies a potentiality of green TiO_2 microbial synthesis, and efforts are in progress to fabricate perovskite solar cells using the obtained mixed TiO_2 nanostructures. Green synthesis of TiO_2 nanostructures was carried out using fruit extracts of *pineapple, grape, and orange* as reducing agents. A DSSC was fabricated using TiO_2 nanostructures as a photoanode and *Murraya koenigii* fruit (MKF) dye and the commercial N719 dye as a photosensitiser. This study reveals that G- TiO_2 nanostructure exhibits the maximum efficiency of 1.78% for MKF dye and 4.33% for N719 dye (Senthamarai et al. 2020).

This green synthesis approach also includes the TiO_2 coated with metals as photoanode to amplify DSSC performance. The challenge encountered using this metal-coated photoanode was thermal instability (Mcevoy 2007; Maçaira, Andrade, and Mendes 2013). The use of biosynthesis mediated nanoparticles as co-photoanode material to enhance the performance of DSSCs has been rarely documented. Rapid synthesis of silver nanoparticles (Ag NPs) from Henna extract has been reported for their application to improve the photoabsorption ability of photoanode (Kiruba Daniel et al. 2013). Since Henna is cheaper and readily available biological source rich in phenolic compounds, it can potentially be used to synthesise Ag NPs to improve photoelectron capture, photoabsorption, and thus enhanced photocurrent generation.

On the other hand, uniform Ag NPs were synthesised at room temperature by using *Peltophorum pterocarpum* flower extract, which showed a remarkable increase in power conversion efficiency of dye-sensitised solar cells from 2.83% to 3.62% with an augmentation of around 28% by incorporation of 2 wt% green synthesised Ag NPs into TiO_2 photoanodes (Saravanan et al. 2017). Thereafter in 2019, Ag NPs were synthesised using leaf extract of guava and were incorporated by dip coating method on the surface of TiO_2. The resulting Ag- TiO_2 photoanode with 30 min dip-coating effectively enhanced the solar cell efficiency from 4.85% to 6.69% (Ramanarayanan et al. 2019). However, very few reports have been found until now on device fabrication, including DSSCs using green synthesised Ag NPs. The green synthesis method produces stable Ag NPs. The Ag NPs enhance the power/efficiency of the TiO_2 nanoparticles to adsorb the dye and consequently enhance the light absorption (Saravanan et al. 2017).

Green synthesis of Au NPs, platinum nanoparticles (Pt NPs), and TiO_2 doped with Au and Pt NPs were carried out using *T. arjuna* bark extract. The TiO_2 NPs doped with the Au NPs and Pt NPs were characterised using Raman spectroscopy and photoluminescence spectroscopy. The appearance of strongest Eg mode at 144 cm^{-1} was attributed to the Ti-O stretching vibrations ascertaining the presence of anatase phase in the TiO_2 NPs. The photoluminescence studies showed the quenching of the fluorescence of the $TiO_{2,}$ indicating the strong interaction between the TiO_2 and the metal NPs. XRD analysis confirmed the formation of anatase TiO_2 with no phase transformation with the doping of Au and Pt NPs. The formation of spherical particles with homogenous particle distribution was evinced by SEM analysis. The energy conversion efficiency and the V_{oc} were found to be improved with Au doped TiO_2 as compared to the pure anatase TiO_2 and the Pt doped TiO_2 (Gopinath et al. 2016). It is observed that this is an eco-friendly synthesis method that provides nontoxic and environmental friendly nanomaterials for solar energy device application. The maximum energy conversion efficiency of 3.44% is obtained from Au doped TiO_2 compared to pure and Pt doped TiO_2 nanoparticles. The green synthesised Au NPs, Au@TiO_2 centred shell nanostructures and Au:TiO_2 nanocomposite was used to fabricate photoelectrodes DSSC applications (Solaiyammal and Murugakoothan 2018; Solaiyammal and Murugakoothan 2019). This study revealed that the efficiency of DSSC is significantly increased from 5.2% to 8.6% with Au: TiO_2 based nanocomposite. Thus, it is concluded that the Au NPs in the TiO_2 matrix improve the DSSC device performance. In addition to this, the formation of Schottky barrier at the Au@TiO_2 interface could effectively minimise the charge recombination as a result of fast charge transfer process.

In recent studies, to improve the stability and efficiency of DSSC, polymer-based electrolytes such as poly(ethylene oxide) (PEO), polyurethane (PU), poly(vinylidene fluoride) (PVDF), polyacrylonitrile (PAN), and poly(vinyl chloride) (PVC) (Prabakaran et al. 2018; Arof et al. 2014) were used. Hence, the study was done on a green synthesised TiO_2 nanoparticle using *Averrhoa bilimbi* extract with 4-Diamino-6-Phenyl-1-3-5-Triazineto (DPT), Poly(ethylene glycol)/KI/I2/2 electrolyte, which improved the long term stability of the polymer electrolyte, ionic conductivity, and also enhanced the photoelectric energy conversion efficiency (Abisharani et al. 2020). Thus, it opened a door for utilising natural TiO_2 in DSSC applications to improve the stability of polymer electrolytes with added advantages of green synthesised NPs such as their eco-friendliness, low cost etc.

Among many metal oxide semiconductors, TiO_2 is considered as an ideal and ZnO, ZrO_2, CdS/CdSe and SnO_2 are considered as promising alternative materials for photoanode (Mehmood et al. 2015; Li et al. 2013). ZnO is cheap, non-toxic and benign to the ecosystem with a bandgap of 3.37 eV and an exciton restricting energy of 60 meV at room temperature

(Kaidashev et al. 2003). The ZnO has enormous utilisation in applications of sensors, photovoltaic and cosmetic products than other metal oxides NP's (Özgür et al. 2005; Rositza 2012). ZnO is considered as desired raw material for photoanodes in DSSCs because of its various advantages like flexibility in synthesis, the richest family of nanostructures, high electron mobility and similar energy band structure as TiO_2 (Ko et al. 2011; Grätzel 2001). Recently there have been several reports on ZnO nanoparticle synthesis using *Vernonia amygdalina* (Ossai, Ezike, and Dikko 2020), *Tilia aomentosa* (Shashanka et al. 2020), *Colocasia esculenta* (Kumar, Sakthivel, and Balasubramanian 2017), *Amorphophallus konjac* (Kumar, Sakthivel, and Balasubramanian 2017), *Carica papaya* (Rathnasamy et al. 2017), *Lycopersicon esculentum* (Sutradhar and Saha 2016) extract and its application in DSSC. It is found that thin films of ZnO/GO/ TiO_2 nanocomposite enhanced power conversion efficiency by 6.18% (Sutradhar and Saha 2016). ZrO nanoparticles were prepared via green synthesis using *Glorisa superba* tuber methanolic extract and used as photoanode in DSSC.

The previous research findings observed that the photoanode rectification could improve the electron infusion and smother the electron rejoining while forming an energy barrier (Vennila et al. 2018). Despite these advancements, the bacterial synthesis of more unpredictable semiconductor nanostructures, such as Core/Shell Quantum Dots, has yet to be explained. The biosynthesis of metal nanoparticles is more difficult than the natural union of quantum dots. The generation of heterogeneous nanostructures such as CdSe, CdTe, CdS, and Ag_2S involves the association of elements in an oxidation state, for example, Ag, Se, Cu, and Au. The use of microorganisms for the supervised processing of centre shell hetero-nanostructures has become an unexplored challenge in this situation (Gallardo et al. 2014; Plaza et al. 2016).

Based on this, the very first attempt was made to develop a biological method to synthesise CdS/CdSe Core/Shell QDs by using *bacteria cells* and an investigation on their potential application in QDSSCs is done. It has been proven that the biologically synthesised Core/Shell QDs are more photo-stable than CdS QDs and is a key characteristic for their technological application in solar cells and bioimaging. The result reveals that when compared to CdS QDs solar cells, the efficiency of solar cells sensitised with CdS/CdSe QDs increased nearly 2.5 times (Órdenes-Aenishanslins et al. 2019). For the first time, mass-scale *sugar-mediated* green synthesis of tin oxide (SnO_2) is reported and the results obtained with SnO_2-ZnS NC-photoanode showed improvement in dye attachment, (V_{oc}), electron life-time, excess light-harvesting capability, and reduced recombination rate (Shaikh, Mane, and Joo 2014).

Apart from this, titanium oxide and zinc oxide are two examples of metal oxides and – are preferred for photoanode because of their wide bandgap semiconductor characteristics. Among these materials, anatase TiO_2 has been extensively studied for DSSC applications, and the use of anatase TiO_2 in DSSCs has yielded a photovoltaic power conversion efficiency of over 10% to date (Liao et al. 2011; Sauvage et al. 2010).

To enhance the efficiency and performance characteristic of DSSC, the thickness of metal oxide, different composition of metal oxides, morphological orientation and the most important bandgap of the photoanode can be optimised (Sengupta et al. 2016). Colossal research has already been carried out by varying the above mentioned parameters. Table 2 shows the morphological and photoelectrical properties of DSSC.

Morphological features greatly impact the performance of photoelectrodes in dye-sensitised solar cells, determined mainly by changes in the electron transport effectiveness, surface area open for dye loading, and pore diameter for electrolyte diffusion (Maçaira, Andrade, and Mendes 2013). The performance of DSSC based on green sensitised nanoparticles, nanostructures, and nanocomposites as photoanode is shown in Figure 4(a,b).

Figure 4(a) shows the performance of DSSCs fabricated using the green synthesised ZrO_2, TiO_2, ZnO, and SnO_2 nanostructures as photoanodes. It is observed that the green TiO_2 obtained from *Averrhoa bilimbi* plant extract has a bandgap of 3.2 eV and a particle size of 15 nm. The DSSC is very economical, nontoxic, and performs better efficiency of 5.2% when using cis dithiocynato-N, N-bis (2,2- bipyridyl-4,4-dicarboxylic acid) ruthenium (II) (N3 dye). It also generates 0.845 V an open-circuit voltage and 11.6 mA/cm^2 high J_{sc}, and 0.53 FF.

Figure 4(b) expresses the DSSC performance based on TiO_2 nanostructures and nanocomposites with green sensitised co-photoanode material. The highest power conversion efficiency of 8.2% is attained by Au- TiO_2 nanocomposite. Remarkably the aqueous extract derived from leaves of *Phyllanthus embilica* was used to synthesise (Au NPs) without any stabiliser and surfactant. The AuNP's morphology is spherical and 21.05 nm in size. The peak short circuit current density (J_{sc}) is 15.88 mA/cm^2, with a V_{oc} of 0.76 V and a FF of 0.71. As a result, without using any harmful chemicals, the green synthesis of metal and metal oxide NPs could be a promising, low-cost, and environmentally friendly process. Properties like size, morphology, crystalline structure can be controlled and attained with this technique (Nabi et al. 2018). Moreover, in DSSC, very few plants are leveraged and many more plants need to be explored for phytosynthesis of NPs. There are various areas where more research on the green synthesis of NPs is required, mainly focusing on optimising the process to gain the required shape and size and physicochemical properties. Mechanistic features, stability of NPs needs to be examined and explored as well for practical use. Multiple parameters like pH, temperature, and salt concentration in the biological extract are required to be studied for their effect on NPs properties. There is more scope for research in terms of large scale production of NPs and it's usage in various applications based on UV light absorption.

5. Progress in DSSC device efficiency

In the DSSC, the most crucial mechanism is to transfer and collect the free electrons excited by the photo anodic reactions from dye to the power circuit. The bandgap, composition, morphology, and thickness of metal oxide layer are the key parameters significantly influencing the performance of photoanode (Marlinda et al. 2019). Amongst various metal oxides, TiO_2 and ZnO nanoparticles have broad bandgap semiconducting characteristics and the best photoanodic reaction showing superior photoconversion efficiencies. Most of the researchers have used the conventional synthesis approaches, namely solvothermal, sol-gel, hydrothermal etc., to synthesise photoanode nanomaterials.

Table 2. Photoelectrical and morphological properties of DSSC.

Nanoparticles	Extract source	Particle size (nm)	Morphology of NPs	J_{sc} (mA/cm^2)	V_{oc} (V)	FF (%)	η (%)	Refs.
TiO_2	*Phellinus linteus* mushroom	Diameter 25, Length of 150	Nanorods	8.18	0.69	67	3.80	Ekar et al. (2017)
TiO_2	*Bixa orellana*	13 ± 2	Spherical	9	0.506	0.65	3	Maurya et al. (2019)
TiO_2	Coelastrellasp MG257917	21–105	Spherical	3.8	0.98	0.4	1.09	Adenigba et al. (2020)
TiO_2	*Averrhoa bilimbi*	15	Aggregated and Irregular	11.6	0.845	0.53	5.2	Abisharani et al. (2020)
G-TiO_2 + N719	Grape		Nanorods	9.95	0.663	0.65	4.33	Senthamarai et al. (2020)
Au-TiO_2	*Terminalia arjuna*	5–10	Spherical	9.43	0.77	0.47	3.44	Gopinath et al. (2016)
Pt-TiO_2	*Terminalia arjuna*	5–10	spherical	8.64	0.71	0.508	3.14	Gopinath et al. (2016)
Au@TiO_2	*Phyllanthus emblica*	60–80	Spherical	15.28	0.73	0.68	7.6	Solaiyammal and Murugakoothan (2018)
Au-TiO_2	*Phyllanthus embilica*	21.05	Spherical	15.88	0.76	0.71	8.6	Solaiyammal and Murugakoothan (2019)
Ag-TiO_2 (2 wt%)	*Peltophorum pterocarpum*	20–50	Spherical	8	0.71	0.642	3.62	Saravanan et al. (2017)
Ag-TiO_2-30	Guava (*Psidium guajava*)	15–25	Spherical	13.33	0.737	0.666	6.69	Ramanarayanan et al. (2019)
ZrO_2 (zirconia)	Glorisa superba	11.625	Hexagonal	1	–	0.122	1.6	Vennila et al. (2018)
Ag	Henna (*Lawsonia inermis*)	28.84	Spherical	44	0.15	–	–	Kiruba Daniel et al. (2013)
ZnO	Tomato *(Lycopersicon esculentum)*	50–90	spherical	14.4	0.612	0.52	4.61	Sutradhar and Saha (2016)
ZnO/GO	tomato (*Lycopersicon esculentum*)	50–90	spherical	15.8	0.606	0.55	5.26	Sutradhar and Saha (2016)
ZnO/GO/TiO_2	Tomato (*Lycopersicon esculentum*)	50–90	Spherical	17.4	0.512	0.69	6.18	Sutradhar and Saha (2016)
ZnO	*Carica papaya*	50	Spherical	8.1	0.521	0.38	1.6	Rathnasamy et al. (2017)
ZnO	*Amorphophallus konjac*	length of 237, diameter of 76	Rice Shaped	6.8	0.55	0.446	1.66 ± 2	Kumar, Sakthivel, and Balasubramanian (2017)
ZnO	*Colocasia esculenta*	length 179, width 63	Nanorods	4.9	0.51	0.639	1.6	Naresh Kumar et al. (2018)
ZnO	*Vernonia amygdalina* (bitter leaf)	9.5	Spherical	18.05	0.39	0.36	0.63	Ossai, Ezike, and Dikko (2020)
ZnO	*Tilia tomentosa* (Ihlamur)	80	Spherical	6.26	0.65	0.485	1.97	Shashanka et al. (2020)
CdS/CdSe	*Escherichia coli* BW25114	17	Spherical	$2.03 \times 10^{-1} \pm 9.73 \times 10^{-3}$	0.209 ± 0.011	0.422 ± 0.147	$2.00 \times 10^{-2} \pm 1.90 \times 10^{-3}$	Órdenes-Aenishanslins et al. (2019)
SnO_2-4L ZnS	Market sugar	40–60	spherical	9.47	0.61	0.51	3	Shaikh, Mane, and Joo (2014)
SnO_7-SL ZnS	Market sugar	40–60	Spherical	7.72	0.57	0.53	2.4	Shaikh, Mane, and Joo (2014)

To overcome the adverse effects of these conventional methods, green synthesis has opened a new field of research in the modification of photoanodes. To date, the maximum photoconversion efficiency using green sensitised TiO_2 composite based photoanodes is more than 10%. The research report shows that the bandgap modification of TiO_2 and ZnO using appropriate dopants effectively improves photovoltaic performances. In recent studies, the polymer electrolytes like polyethylene oxide (PEO), polyvinylidene fluoride (PVDF), poly acrylonitrile (PAN) and polyvinyl chloride (PVC), etc. have been used to improve the stability and to increase the efficiency of DSSC. The DSSC with PEG polymer electrolyte doped with Bio synthesised TiO_2 to the photoanode have evinced an efficiency of 5.2%. Thus, it explored a novel way of employing the ecofriendly approach to generate TiO_2, which is nontoxic and economical in preparation. DPT doped Polymer electrolytes (PEG/KI/I2) with nanocrystalline

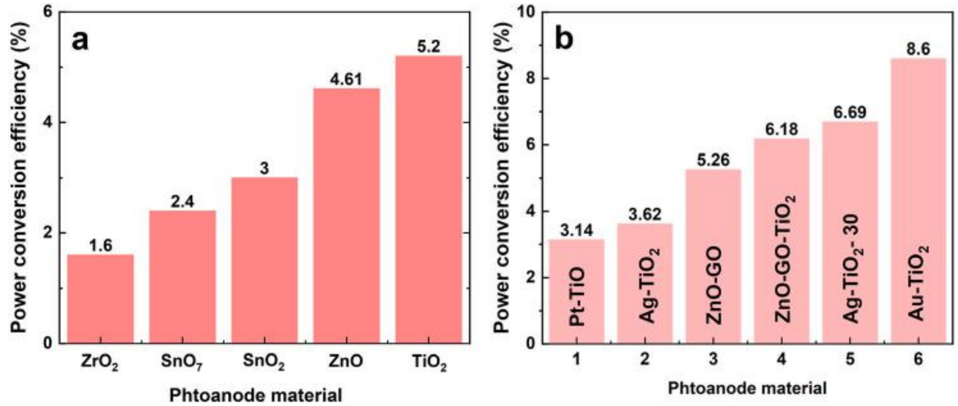

Figure 4. DSSC performance based on (a) green sensitised photoanodes and (b) composite photoanode materials.

TiO_2 increases ionic conductivity, polymer electrolyte long-term stability, and solar to electric energy conversion performance. TiO_2 and ZnO nanostructures with 1D, 2D, and 3D nanostructures have been reported to be beneficial for DSSC applications. More dye adsorption and direct transport are possible with a 1D nanostructure. The highest power conversion efficiency is found when the photoanode metal oxide layer is at its optimal thickness. The addition of the TiO_2 scattering layer on the open-ended TiO_2 nanotube arrays (3D nanostructure) enhanced the power conversion efficiency by 10.30% due to the improved light harvesting (Rho et al. 2015). From the existing literature, it is found that the green sensitised metal nanoparticles like Gold (Ag) and Silver (Au) have been extensively used in photovoltaic applications because of their relative stability and high level of absorbance in the visible region. Also, the amalgamation of a small concentration of Au NPs has resulted in improved efficiency of DSSC in comparison to the pure TiO_2 nanoparticle. Saravanan et al. developed DSSCs with and without different doping concentrations of green synthesised Ag NPs in TiO_2. Because of the plasmonic effect, doping TiO_2 with 2 wt% Ag nanoparticles increased the efficiency of DSSCs by about 28%. In comparison to the non-doped sample, the FF and V_{oc} achieved a maximum efficiency of approximately 7.2% and 12.1% as the doping concentration increased up to 2 wt% Ag NPs. As a result, the silver nanoparticles' plasmonic effect greatly improves the energy conversion performance in DSSCs (Saravanan et al. 2017; Bhojanaa, Ramesh, and Pandikumar 2020). By considering the cost of gold and silver NPs, this review motivates researchers to investigate different materials and structures of photoanode to reduce the cost of DSSC. Among the carbonaceous materials, especially graphene has sparked a lot of interest due to its exclusive optical, thermal, mechanical, and electrical properties. Many researchers have claimed that highly conductive graphene structures can reduce the charge recombination rate and interfacial resistance in combination with TiO_2 photoanode. It also boosts the DSSC performance when used in very low amounts (0.5 wt%). Thus, the influence and interaction of graphene material with different metal oxide particles as photoanodes using green technology can bring DSSC to the next level of development. According to current research progress in photoanode modification, the overall photoconversion efficiency of DSSC remains nearly 13% (Pallikkara and Ramakrishnan 2021).

The DSSCs demonstrated higher dye loading amounts, lower internal resistances, lower electron recombination rates, and faster electron transport rates in the construction of photoanodes with gradient graphene content, resulting in high open circuit voltage and current density. The DSSC had V_{oc} of 0.72 V, J_{sc} of 17.11 mA/cm^2, an FF of 0.63, and an energy conversion efficiency of 7.71% at optimised conditions, implying a 41% improvement in efficiency over a DSSC based on pure TiO_2 photoanode, which had a V_{oc} of 0.69 V, J_{sc} of 12 mA/cm^2, an FF of 0.62, with an efficiency of 5.45%. By increasing the dye loading amount and boosting the electron bridge effect of graphene from TiO_2 to the FTO, which decreased electron recombination rate and enhanced electron transport rate, the TiO_2 photoanode with gradient graphene content could significantly improve the efficiency of DSSCs (Wei et al. 2017).

6. Challenges and prospects

DSSCs are currently rival in market with thin-film solar cells, particularly for cost as well as readily availability. Grand view research has conducted a study on DSSC technology which states expenses in DSSC technology are growing consistently12% each year from 2015, and same is expected till 2022 (Shakeel Ahmad, Pandey, and Rahim 2017). Expenses incurred for DSSC in photovoltaic market in the year 2014 was 49.6 million USD. Apart from expenses, applications of DSSC are also getting increased with the passing time since the last few years. DSSC integration with building-applied photovoltaics and building integrated photovoltaics leads to solar installation growth in commercial and residential complexes. Here are few leading manufactures in DSSC: Konica Minolta Sensing Europe B.V, Merck KGaA, G24 Innovation Ltd, Solaronix, Dyesol Ltd., CSIRO, 3G Solar Photovoltaics Ltd., EXEGER Sweden AB, G24 Power Ltd, and Solaris Nanosciences Corporation (Babar et al. 2020). Significant challenges currently with DSSCs are still with stability and efficiency, which gives scope for improvisation for commercialising DSSCs (Fakharuddin et al. 2014). As of now, the highest efficiency with DSSCs is reported as 13% (Mathew et al. 2014). Following are various recommendations related to this:

- Foremost research should focus on regulating and reducing barriers with the photoanode-electrolyte interface since the transport mechanism of charge plays a critical role in efficiency.

- The recombination rate needs to be reduced to increase conductivity which can be achieved using TiO_2 nanocomposites with carbon allotropes like carbon nanotubes, graphite.
- In a dye-sensitised solar cell, the photovoltaic performance of TiO_2 photoanode is greatly influenced by its crystallinity and morphology. Therefore, different morphologies of TiO_2, namely nanoparticles, nanorods, nanotubes, and nanowires, etc., are being preferred to manufacture the photoanodes of DSSCs. The chemical and physical methods are expensive and use toxic and hazardous materials during nanomaterials synthesis. On the other hand, the biological methods, including bio-sources such as bacteria, fungi, yeast, and plant extract as a reducing agent, are very cost-effective and do not comprise the use of toxic and hazardous materials during the processing. These green methods are low-temperature and energy-saving approaches. Hence, biological or green synthesis is the simple and cost-effective method for preparing nanocrystalline anatase and rutile TiO_2 photoanode with different morphologies.
- The low light dispersing inside TiO_2 photoanode is the fundamental wellspring of low current density and poor optical performance. This is straightforwardly identified with the efficiency of light absorption and charge collection. The TiO_2 morphology can be changed by doping various elements on TiO_2 (nanowire, nanotube, core–shell structure, hollow sphere, core–shell structure, etc.) or by adding TiO_2 nanocomposite based on the generated carbocation materials to improve both.
- The light-harvesting ability must be reinforced to expand the energy conversion efficiency of 3D nanostructure based solar cells. The 0-dimensional nanoparticles can be filled in the cavities of 3-dimensional nanostructure to attain a better light-harvesting ability, which is currently an active field of study.

Disclosure statement

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