

Recent advancements and challenges in flexible low temperature dye sensitised solar cells

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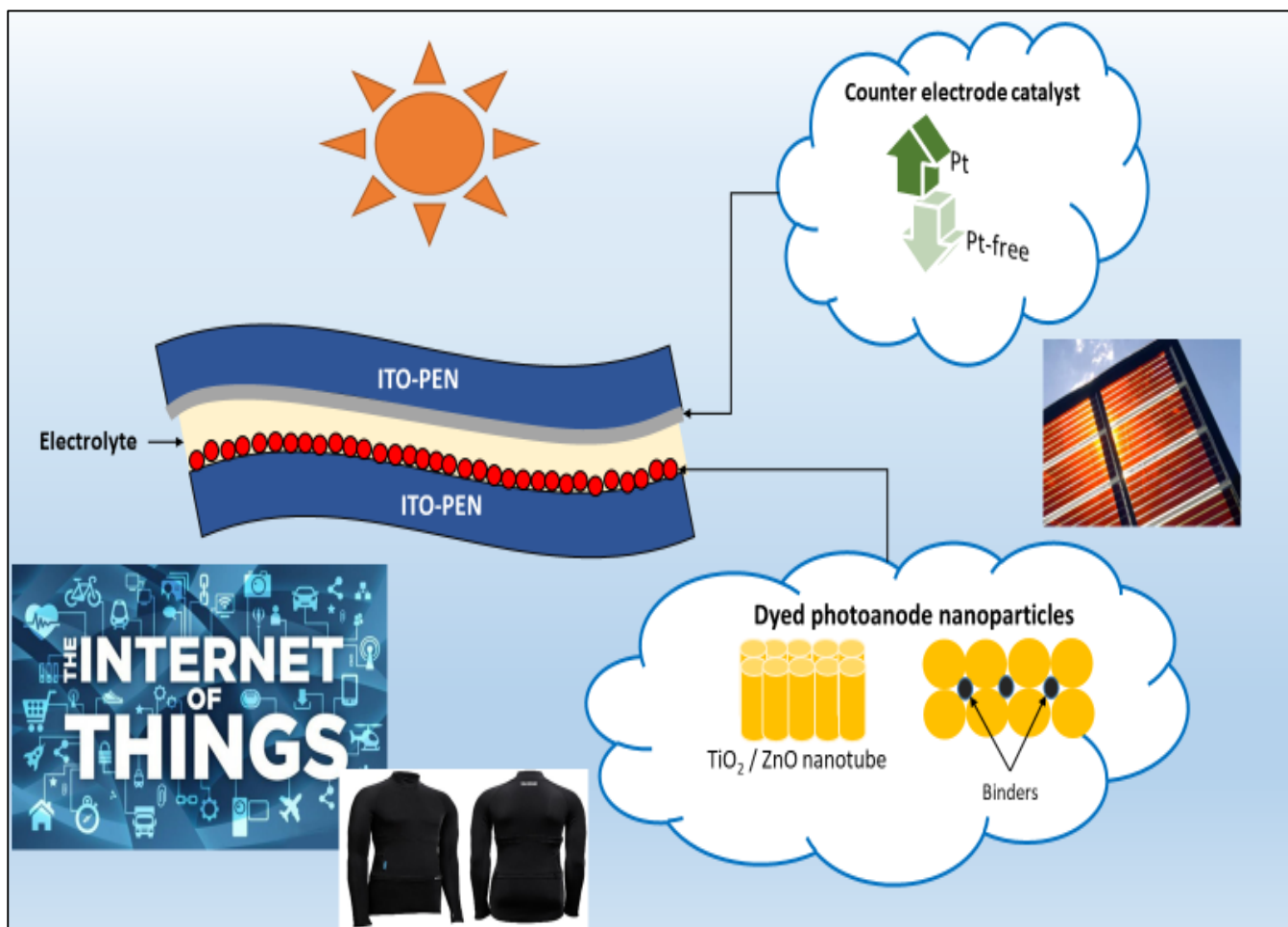
Abstract

Dye sensitised solar cells (DSSCs) have been in extensive development in recent years in the field of solar energy due to its cost-effectiveness, ease in fabrication, flexibility, and being able to be transparent and coloured as well. Two broad categories of DSSCs based on their fabrication temperature are (1) high-temperature DSSCs and (2) low-temperature DSSCs. Although the low-temperature DSSCs (sintered at less than 150°C) can be flexible and printed on a plastic roll, however, their power conversion efficiency (PCE) is way less compared to their high-temperature counterpart. Research is underway to improve the PCE of low-temperature DSSCs and modules to optimum levels. In this review, an attempt has been made to evaluate different materials and fabrication methods for improved performance of flexible low-temperature DSSCs while also comparing them with the usual rigid high temperature device. Another objective of this study is to critically discuss the progress being made in flexible module development. This review paper would be able to provide comprehensive summary of the recent developments of flexible low-temperature dye sensitised solar cells and modules for reference and also serve as guide for further research in this area.

Keywords: Flexible DSSCs; Low temperature; Photoanode; Counter electrode; Plastic substrate

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Graphical Abstract



Highlights

- Comprehensive summary of the recent developments of flexible low-temperature DSSCs
- **The highest reported conversion efficiency for flexible DSSC is around 10.28%**
- Evaluation of materials and fabrication methods for flexible low-temperature DSSCs
- Technical challenges of flexible low-temperature DSSCs are discussed thoroughly
- Up-scaling of flexible DSSC module is still a challenge due to the low PCE

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Nomenclature

Abbreviations:

CIGS	Copper indium gallium selenide
DC	Direct current
DSSC	Dye sensitised solar cell
EMTE	Embedded metal-mesh transparent electrode
EY	Eosin Y
FESEM	Field emission scanning electron microscope
FF	Fill factor
FTO	Fluorine tin oxide
FTO-PET	Fluorine tin oxide-polyethylene terephthalate
GQD	Graphene quantum dot
IOT	Internet of things
IPCE	Incident photon-to-current conversion efficiency [%]
ITO	Indium tin oxide
ITO-PEI	Indium tin oxide-polyetherimide
ITO-PEN	Indium tin oxide-polyethylene naphthalate
ITO-PET	Indium tin oxide-polyethylene terephthalate

LHE	Light harvesting efficiency [%]
MWCNT	Multi-walled carbon nanotube
PCE	Power conversion efficiency [%]
PDDA	Poly (diallyl dimethylammonium chloride)
PEDOT	Poly(3,4-ethylenedioxythiophene)
PEDOT:PSS	Poly(3,4-ethylenedioxythiophene) polystyrene sulfonate
PEI	Polyetherimide
PEO	Polyethylene oxide
RF	Radio frequency
SEM	Scanning electron microscope
TTIP	Titanium isopropoxide
USD	United States Dollar

Symbols:

J_{\max}	Maximum current [A/cm^2]
J_{SC}	Short circuit current [A/cm^2]
η_{col}	Charge collection efficiency at external circuit [%]
η_{inj}	Quantum efficiency of electron injection [%]
N_e	Number of electron flow through external circuit
N_p	Number of incident photons
P_{in}	Incident power on an area of the cell under standard air mass [W/m^2]
P_{\max}	Maximum power output
V_{\max}	Maximum voltage [V]
V_{OC}	Open circuit voltage [V]

1. Introduction

Fossil fuels have been essential for industrial activity and even day-to-day activities however, they are associated with multiple hazards towards environment, human health and wildlife. Fossil fuels such as coals are the major source of greenhouse gas emissions, particularly carbon dioxide (CO_2), accounted to be 36.3 billion tonnes in 2021 [1]. Several human health complications caused by air pollution include respiratory disease such as tuberculosis, lung cancer, bronchitis, asthma and heart disease [2]. Thus, there have been multiple efforts taken to combat this issue by introducing the use of renewable energy producing minimal carbon emissions. Solar

energy is one of the prominent renewable energy with no harm to the environment and helps in reducing the dependence on fossil fuels.

1.1 Background of solar cell technology

Solar energy has been the subject of extensive research from several years with development of many solar powered technologies such as solar cells, solar thermal collectors, solar cooker, solar dryer, solar water heaters etc., [3]. Solar cell directly converts sunlight into electricity via photoelectric effect, and they are seen to be a promising solar technology due to their materials abundance while also being clean, safe and affordable to operate [4]. The technology has evolved over the years and can be classified into three generations. The first-generation crystalline silicon (Si) based solar cells has dominated the photovoltaic (PV) market and managed to yield high power conversion efficiency (PCE) of around 26% [5]. Moreover, their lifespan can reach to more than 20 years. However, Si-based solar cells have several drawbacks which include expensive, complicated and energy intensive production process, high environmental impact of manufacturing, poor aesthetics and low performance in low light conditions, limiting their presence in terrestrial photovoltaic market [6,7]. Second generation solar cells are based on thin film technologies using thin layer of semiconducting materials such as amorphous silicon, cadmium telluride, copper indium selenide and copper indium gallium diselenide [8]. Despite the good flexibility and lightweight design with low cost, the technology suffers from several drawbacks. These are mainly from the materials such as indium that are becoming rarer and more expensive while cadmium is highly toxic, causing issues in manufacturing [9]. Therefore, third generation solar cells, known as emerging photovoltaic, have been touted as an alternative to Si-based solar cells and thin film solar cells. Organic solar cells, perovskite solar cells and particularly dye sensitised solar cells (DSSCs) are examples of third generation solar cells that has been developed with low cost from an abundance of materials with little environmental impact [4].

1.2 Dye sensitised solar cell as an alternative for solar cell technology

Dye sensitised solar cell (DSSC) is a sandwich structure made up of several components such as conductive substrate, photoanode, counter electrode, electrolyte and sensitiser [10]. The device has been under intensive investigation with considerable amount of work have been done since their first discovery by O'Regan and Gratzel in 1991 [7]. The data from ScienceDirect database showed around 22,500 published

papers, meanwhile Google Scholar database showed 27,500 published papers from 1999 by using “dye sensitised/sensitized solar cell” as key word. DSSCs possess several advantages such as easy fabrication process, low cost with easily available materials for construction and ability to operate well under partial light conditions [11,12]. A comparison between several solar cell technology was conducted by Giannouli [13], highlighting their conversion efficiency and benefits as in Table 1. Despite having lower PCE, DSSC are still under extensive research mainly due to their low cost and simple method. Comparative studies between performance of DSSC and Si solar cells were made by Hamed et al. [14] and Olulope et al. [15] both acknowledging that DSSC was able to react effectively even under low level light condition as long as there was a source of light. Although producing lower conversion efficiency, their good performance under such conditions and in particular their low cost, made DSSC an attractive alternative for solar cell.

Table 1 Type of third generation solar cells and their advantages

Type of solar cells	Conversion efficiency (%)	Reference	Advantages
Dye sensitised	12.3	[16]	<ul style="list-style-type: none"> • Low cost to performance ratio • Simple assembly
Perovskite	21.1	[17]	<ul style="list-style-type: none"> • High electron and hole mobility • Bandgap can be tuned • Low exciton binding energy
Organic	17.0	[18]	<ul style="list-style-type: none"> • Lightweight • Large surface area • Can be processed on flexible substrate

DSSC usually has a rigid structure due to the use of glass materials as the conductive substrate to withstand high manufacturing temperatures [19]. However, these structures have several drawbacks in regards to their weight, rigidity and lack of flexibility particularly these days, since there have been growing demands for

lightweight, flexible and mobile solar chargers [20]. Therefore, different type of DSSCs has been developed in recent years called a flexible DSSC utilising flexible substrate to further enhance the application of DSSC and keeping them up-to-date in the evolution of solar cell technology.

1.3 Flexible and low temperature DSSCs

Flexible DSSCs are usually constructed with plastic substrate and the device is lightweight, flexible while also compatible with high throughput processes such as roll-to-roll forming, screen printing etc. [21]. The flexibility of the plastic substrate allowed the device to have more freedom in shaping the device and combined in a curved shape to be applied for mobile and wearable applications [22]. Flexible DSSC is not only limited to plastic substrate, as metal substrate has also been used as conducting substrate [23]. However, metal substrates have several drawbacks where metal mesh has low effective area to accept sunlight, while metal foil have no transmittance, meaning these two are more suitable for large scale commercial flexible DSSC [24]. Hence, plastic substrate using indium tin oxide-polyethylene terephthalate (ITO-PET) and indium tin oxide-polyethylene naphthalate (ITO-PEN) polymer is more commonly applied for flexible DSSC even though they also suffer from some drawbacks themselves. The low conversion efficiency of flexible DSSC has been the main issue for the device with highest reported so far is at around 10.28% [25]. This is due to the low temperature used when sintering the photoanode nanoparticles coated on the plastic substrate or for other treatment process as the polymers can only withstand low temperature of below 150°C [26]. Typically, for a rigid glass-based DSSC, the photoanode-coated glass substrate is sintered at high temperature of 450°C for titanium dioxide (TiO₂) nanoparticles to remove organic surfactant in the photoanode paste or suspension, improve connection between the photoanode particles and better adhesion between the photoanode and the substrate [20]. However, the same process is not applicable for flexible plastic-based DSSC as the substrate would deform at such high temperature. Thus, low temperature of 150°C would be applied when sintering the flexible plastic-based DSSC, causing it to have worse inter-particle connection between the photoanode particles that eventually leads to lower efficiency [20]. There have been efforts to make use of different materials for DSSC components such as binder-added TiO₂ photoanode [27] and polyetherimide polymer (PEI) substrate [28] as well as utilising different

fabrication methods such as electrochemical deposition [29] and pulsed laser deposition [30].

1.4 Objectives, novelty and structure of review paper

The review paper is written by taking into consideration the problems faced by flexible DSSC and studies conducted by researchers in recent years. An attempt has been made to review different materials and module designs used in developing flexible and low temperature DSSC and comparing them with the usual rigid and high temperature DSSC. Moreover, different fabrication methods that are suitable to develop flexible and low temperature DSSC have been discussed in detail. Furthermore, the potentials and challenges for flexible and low temperature DSSC has also been critically discussed. Several relevant review papers have been reviewed to find the research gaps and novelties of this review paper.

Several review papers on the topic of DSSC have been published such as reviews by Sharma et al. [19], Sharma et al. [31] and Kumar et al. [32], discussing the developments of DSSC components, materials and fabrication methods without focusing on flexible DSSC as the main topic. Saeed et al. [33] meanwhile reviewed the developments of DSSC under ambient illumination condition, a key attribute separating the use of DSSC over other solar cell technology, while also including several studies on flexible DSSC. Although these reviews do not emphasise on flexible or low temperature DSSC, they all recommended on the application of flexible DSSC as the future of DSSC development. Thus, indicating the potential of such device and the discussion on the topic can be further seen in some other reviews.

One of the common discussions in review papers on DSSC is to evaluate the different materials and fabrication methods to construct the DSSC component and the effects on efficiency. Such example can be seen in reviews by Li et al. [24], Zhao et al. [34] and Zhu et al. [35] that reviewed materials and components such as indium tin oxide-polyethylene terephthalate (ITO-PET) and indium tin oxide-polyethylene naphthalate (ITO-PEN) substrates, TiO₂ and zinc oxide (ZnO) photoanode, platinum (Pt) counter electrode, polyethylene oxide (PEO) and polymer electrolyte as well as ruthenium (Ru)-based sensitisers that are suitable for flexible DSSC. These reviews discovered that the highest conversion efficiency for flexible DSSC is still lower than 11% compared to other flexible solar cell especially perovskite solar cells that produced around 12-14.78% conversion efficiency. Thus, indicating there are still much work to

be done in improving the performance of flexible DSSC that can compete with other available solar cells. A review by Lund et al. [36] also discusses the application of flexible textile DSSC in literature as well as mechanical stability of flexible DSSC which was found to produce good results in literature despite the low cell stability of these devices. Common drawbacks and research gaps found in these reviews are the lack of emphasis on the advancements of rigid glass-based DSSC which should go hand-in-hand with the development of flexible DSSC. Hence, allowing comparisons between the two types of devices to be made that will give a clearer idea on whether flexible DSSC is able to compete with rigid glass-based DSSC. This issue is addressed in this review paper and is also one of the novelties for the paper.

A recent review by Noorasid et al. [22] emphasised on the development of photoanode component for both flexible and glass-based DSSC, including the addition of precursors to the photoanode materials as well as fabrication techniques such as doctor blade and electrophoretic deposition for flexible DSSC. Meanwhile Wu et al. [4] reviewed various type of counter electrodes for DSSC, including a section specifically for flexible counter electrodes for flexible DSSC application. Despite these reviews discussing both flexible and non-flexible DSSC, they are mainly focused on certain DSSC component which will be addressed in this review paper by reviewing the advancements of conductive substrate, photoanode and counter electrode for both types of DSSC. Another novelty aspect of this review paper is by including the explanation on several fabrication techniques that can be conducted at low temperature, suitable for the construction of flexible DSSC. The final novelty aspect of this review paper lies on the discussion of flexible DSSC module by exploring some literatures that developed such structures to evaluate their methods, effectiveness and stability which still remains a less explored topic in DSSC. A summary of several review papers published on this topic in recent years can be seen in Figure 1.

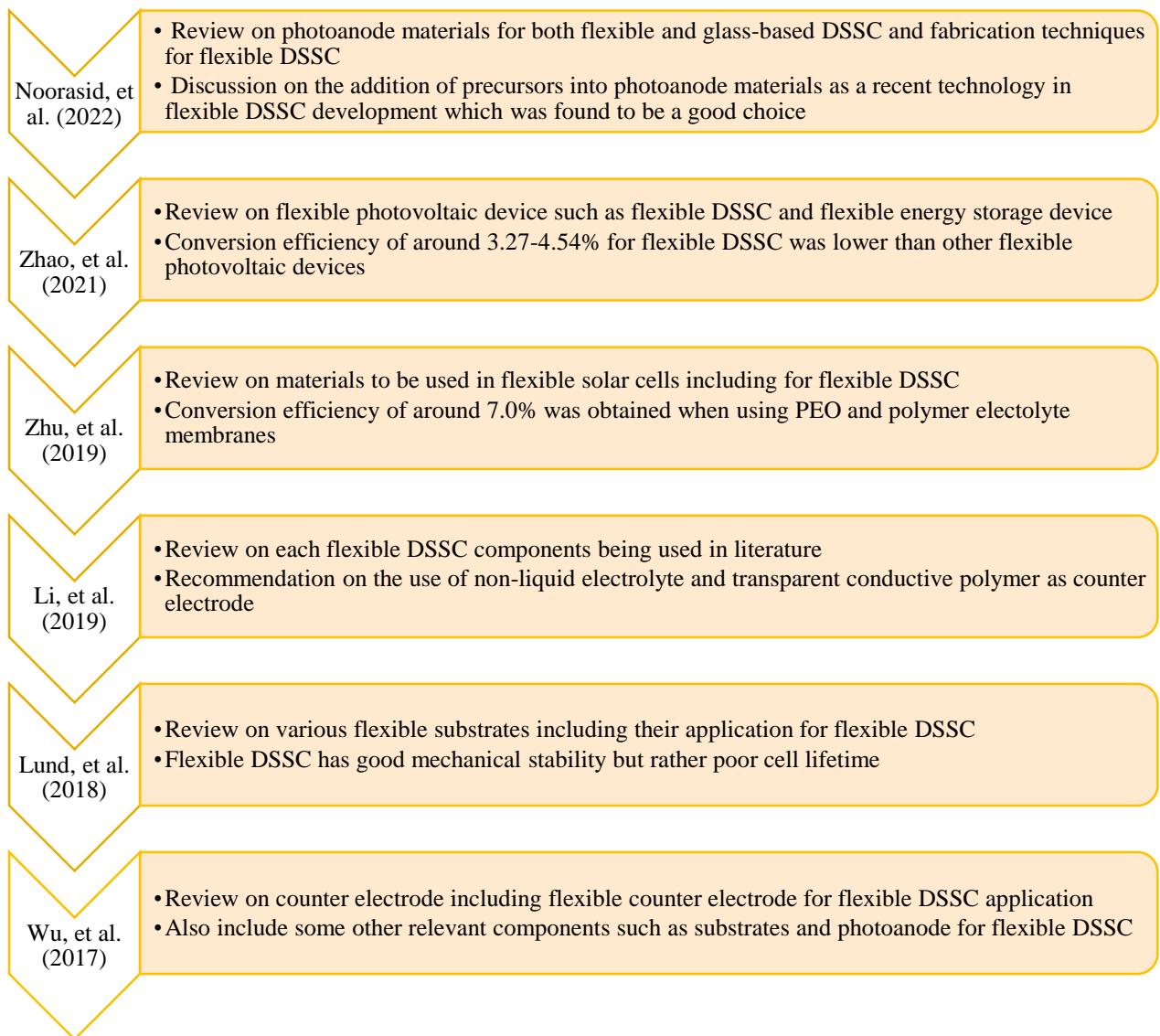


Figure 1 Summary of review paper published on flexible DSSC and flexible solar cells in recent years

In literature, it can be seen that the amount of publication for flexible DSSC review paper is much less compared to a more generalised review on DSSC. This was probably due to the lack of drastic improvements in the conversion efficiency for flexible DSSC as well as the developments of other flexible solar cells such as Cu(In,Ga)Se (CIGS) solar cells, hydrogenated amorphous silicon (a-Si:H) solar cells and perovskite solar cells [24,37]. Thus, making this review paper titled “Recent advancements and challenges in flexible low temperature dye sensitised solar cell” a significant addition to not only in the field of flexible DSSC, but also in solar cell technology in general.

This review paper is divided into six different sections starting with a brief explanation on the working principle of DSSC and basic mathematical equations to give the readers a basic idea on how the device works. After that, a section is dedicated to explain and discuss several suitable fabrication methods for flexible and low temperature DSSC. The next section covers the main focus of the review paper, which is the advancements of flexible and low temperature DSSC where it is classified into several sub-sections based on the DSSC components as well as flexible DSSC module. The section also covers the recent developments of rigid and high temperature DSSC as a comparison to give more insights for readers learning this topic. The subsequent section is an overview on the potential and challenges in the matter to help set up the final section which is the conclusion and recommendations for further studies to be made on this subject.

2. Working principle and performance evaluation of DSSC

2.1 DSSC structure and working principle

Dye sensitised solar cell (DSSC) is made up of several main components including the conductive substrate, photoanode, counter electrode, electrolyte and sensitizer. The structure is usually formed of two conductive substrates sandwiched together with one side coated with a layer of photoanode nanoparticles soaked with sensitizer/dye while the other side is coated with a layer of counter electrode catalyst. Some electrolyte, usually in the form of liquid is deposited in between the two substrates before they are sealed together, completing the structure of the solar device. The structure of DSSC and the basic working principle of the device can be seen in the schematic diagram shown in Figure 2.

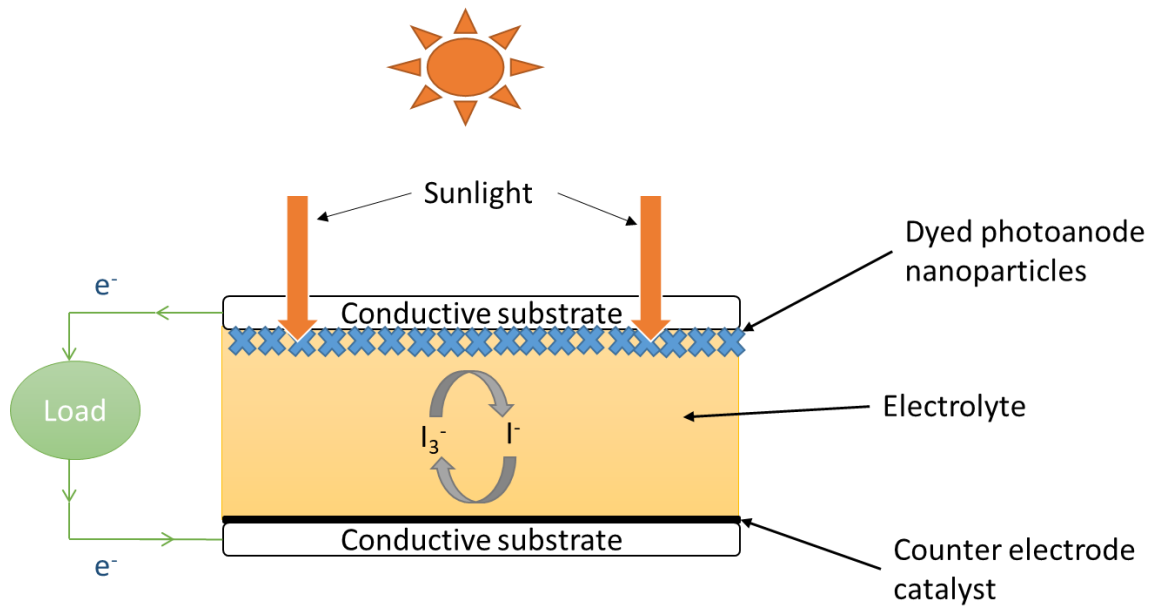


Figure 2 Schematic diagram of dye sensitised solar cell

As shown in Figure 2, a basic working principle of the DSSC device can be explained into several steps. First, the DSSC is exposed and illuminated by sunlight allowing the dye on the surface of the photoanode layer to absorb the light, causing the electrons in the dye molecules to get excited. The electrons will then get injected into the conduction band of the photoanode causing the sensitizer to be oxidised [38]. The injected electrons are then diffused through the structure of the photoanode film before being transferred to the counter electrode via the external circuit of the device. Then, these electrons are collected by the electrolyte found in the counter electrode before they interact and reduce the redox mediator in the electrolyte. Dye regeneration process then occur as the reduced redox mediator reduces the oxidised sensitizer back to their original ground state. Thus, completing the circuit and the process keeps on repeating to run the solar device.

2.2 Evaluating the performance of DSSC

The performance of developed DSSC can be evaluated by calculating several key parameters that include the short circuit current (J_{SC}), open circuit voltage (V_{OC}), fill factor (FF), power conversion efficiency (PCE) and incident photon-to-current conversion efficiency (IPCE). These parameters can be obtained from the current-voltage (I-V) curve generated by using solar simulator at constant irradiation as compared to using outdoor solar radiation [39]. A typical I-V curve for DSSC devices is shown in Figure 3 with several of the aforementioned parameters included.

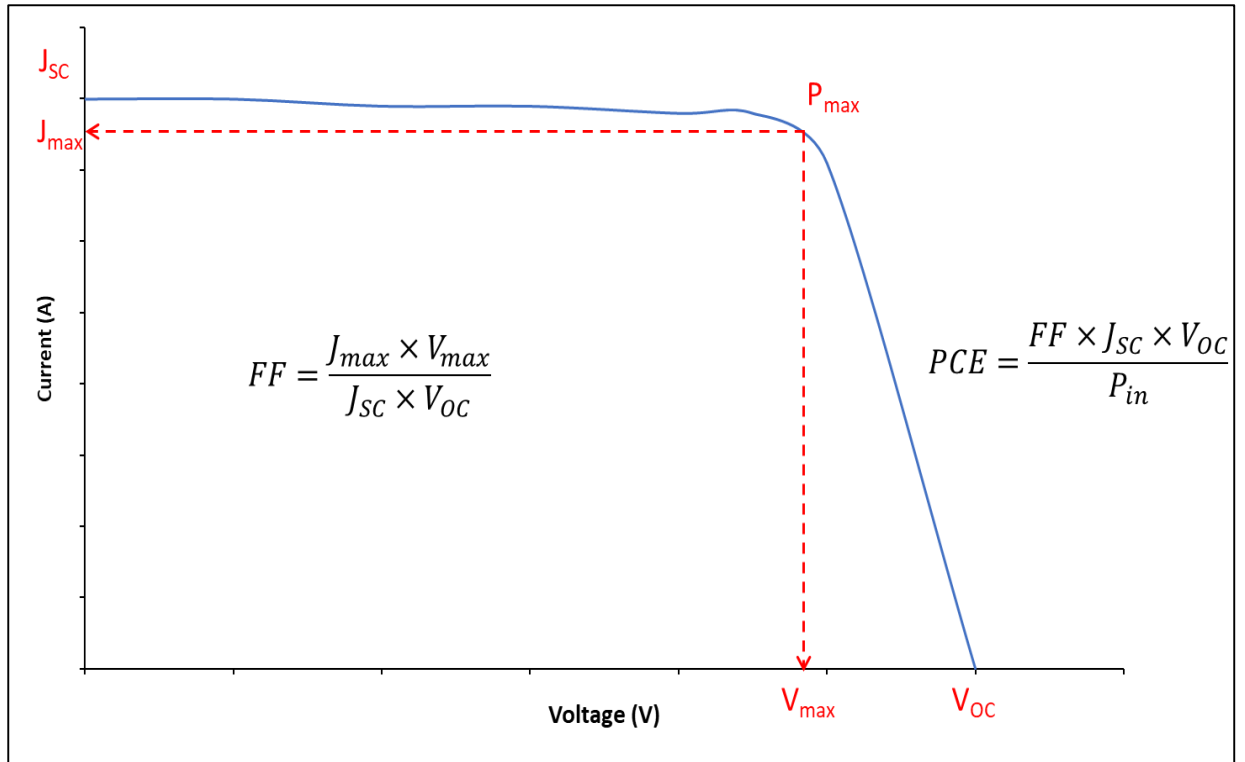


Figure 3 A typical I-V curve for DSSC devices

Short circuit current (J_{sc}) is the maximum output current of DSSC and they are measured under light irradiation and when the voltage across the cell is zero as shown in Figure 3. Meanwhile, open circuit voltage (V_{oc}) is the maximum voltage provided to the external circuit by the solar cell and the parameter is measured when no current is flowing as shown in Figure 3. From the I-V curve, the maximum power output (P_{max}) can be obtained based on the product of maximum current (J_{max}) and maximum voltage (V_{max}). Fill factor (FF) is defined as the ratio of P_{max} of the solar cell to the product of J_{sc} and V_{oc} [39]. The equation to calculate FF can be seen in Equation (1) as follows:

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

From the fill factor, the power conversion efficiency (PCE) can be calculated. PCE is the percentage of solar energy shined on the DSSC device converted into electrical energy which can be calculated as shown in Equation (2) [19].

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

Where, P_{in} is the incident power on an area of the cell under standard air mass 1.5 irradiation or 1000 W/m^2 [39]. Besides PCE, the incident photon-to-current conversion

efficiency (IPCE) can also be calculated. IPCE is defined as the ratio of the number of electrons that flow through the external circuit (N_e) to the number of incident photons (N_p) as shown in Equation (3) [40].

$$IPCE(\lambda) = \frac{N_e(\lambda)}{N_p(\lambda)} \quad (3)$$

IPCE is also known as the external quantum efficiency that can be expressed with three efficiency parameters as shown in Equation (4) [40].

$$IPCE(\lambda) = LHE(\lambda) \times \eta_{inj}(\lambda) \times \eta_{col}(\lambda) \quad (4)$$

Where LHE is the light harvesting efficiency, η_{inj} is the quantum efficiency of electron injection and η_{col} is the efficiency of charge collection at the external circuit. Based on the I-V curve and equations (1) – (4), the performance of DSSC can be evaluated where typically highly efficient DSSC would yield high PCE and IPCE values.

3. Fabrication techniques for low temperature DSSC

Fabrication techniques used to coat or apply the photoanode materials onto the conductive substrate play a role in determining the performance of the device. The technique chosen should depend on the size, shape, flexibility of the device as well as other factors such as energy demand, production cost and volume [41]. The fabrication techniques can be classified as physical and chemical method of deposition. Physical method relies on the physical phenomenon to prepare and deposit the materials, while chemical method is based on the chemical reaction of substrate, precursor and chemical medium [42]. Some examples of fabrication techniques include doctor blade, spin coating, screen printing, electro spray deposition, electrochemical deposition, sol-gel and spray pyrolysis method. However, not all of the methods listed are suitable for flexible plastic-based DSSC that require low manufacturing temperature. Thus, suitable fabrication techniques based on low operating temperature needed to be selected to perform such task as illustrated in Figure 4.

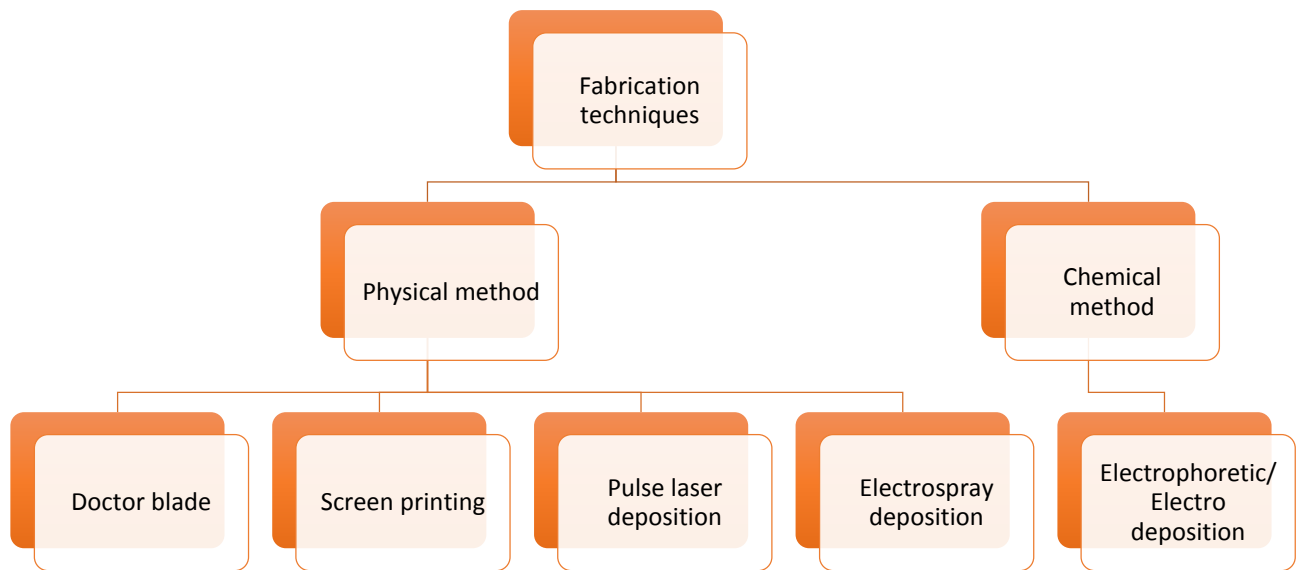


Figure 4 Classification of several fabrication techniques for low temperature DSSC

A common fabrication technique for flexible and low temperature DSSC is the doctor blade technique. **Doctor blade** is a physical method of deposition that is cost-effective, flexible and simple, making them a popular choice for DSSC fabrication [42]. The method is performed by covering parts of the conductive substrate with tape before applying some amount of photoanode paste on the available space on the substrate. A blade or other suitable item such as glass rod is used to spread the paste on the substrate and moved back and forth to spread it uniformly. The tape is removed once the paste dried up, leaving some parts of the substrate coated with the photoanode nanoparticles as shown in Figure 5. The thickness of the coating layer depends on the thickness of the tape and it can even be increased by repeating the spreading process multiple times.

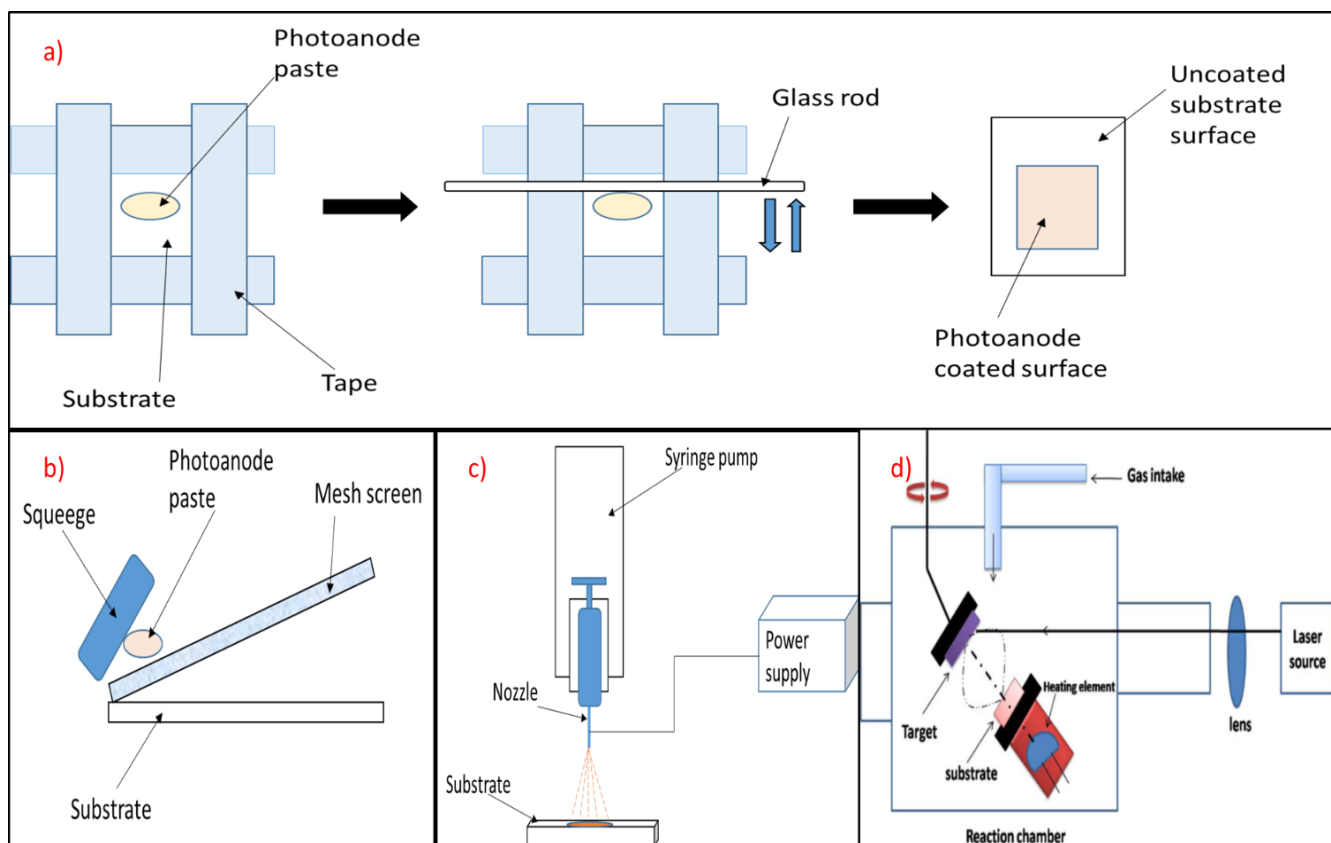


Figure 5 Schematic diagram of multiple low temperature fabrication techniques a) Doctor blade b) Screen printing c) Electro spray deposition d) Pulse laser deposition [42]

Another fabrication method with low temperature for flexible DSSC is screen printing. **Screen printing** is a commonly used printing technique due to abundance of materials for printable paste that can be scaled up for volume manufacturing [43]. Screen printing is also suitable for making and depositing thin films as it is easy to control the thickness by simply managing the thickness of fabric being used [44]. According to a study by Liu et al. [43], screen printing was deemed to be a good fabrication technique by using high viscosity TiO_2 paste for the printing process to attach more dye molecules. Screen printing uses a mesh placed in between the photoanode paste and the conductive substrate. A squeegee is then used to press the paste onto the substrate, forming a pattern based on the openings of the mesh.

Pulsed laser deposition and electro spray deposition are another physical method of deposition with low temperature process. **Pulsed laser deposition** is a relatively simple and straight forward technique by making use of laser energy to evaporate a small surface layer material that will be deposited on a substrate [45]. The fabrication method is also feasible as they are able to modify the optical properties of the photoanode materials simply by varying the number of pulses [46]. A study by Han et

al. [30] demonstrated that pulsed laser deposition method was able to tailor the morphology and thickness of nanostructured ZnO films. This was done by controlling the deposition time or number of laser pulses that influenced the dye absorption and electrolyte diffusion of the nanostructure. **Electrospray deposition** is a simple method by depositing one droplet of sample solution at a time that enables the formation of sub-structure within a film and to prepare nano-sized spheres of photoanode nanoparticles [47]. The fabrication method was also deemed to be better than conventional methods such as screen printing and doctor blade, as the morphology of fabricated TiO₂ showed a semi-self-assembled structure for better performance. Electrospray deposition is able to attain uniform deposition even at large surface, has the capability to remove excess solvent before nanostructures arrived to the substrate for quick deposition of multi-layered films and is able to operate at room-temperature [48].

Fabrication method based on chemical deposition method such as electro or electrophoretic deposition can also be applied at low temperature. Both these methods are a form of electrochemical deposition where electro deposition provided electric current to the electrodes that are then dipped in electrolyte consisting of ions of the deposited species [42]. **Electro deposition** has been one of the methods of choice due to their versatility, accurate controllability, low cost, ability to operate at room temperature with quick rate of deposition while still maintaining good uniformity [49]. **Electrophoretic deposition** meanwhile, deposits charged particles onto a substrate by applying direct current (DC) voltage after the movement of charged particles in a suspension medium is completed [50]. The deposition method possessed several advantages including simple equipment, low cost, high rate of deposition, no requirement of binder, ability to prepare any shape of conductive substrate desired and high reproducibility [50,51]. A summary of several fabrication techniques applied for low temperature DSSC can be seen in Table 2.

Table 2 Fabrication techniques suitable for low temperature DSSC

Fabrication technique	Photoanode	Reference
Doctor blade	TiO ₂ + Titanium isopropoxide	[27]
	(TTIP)	[52]
	TiO ₂ + H ₂ TiF ₆	[53]
	GQD-TiO ₂	[54]
	TiO ₂ nanoparticles	[29]

	TiO ₂ nanoparticles	[55]
	Thin film TiO ₂	[56]
	ZnO + TiO ₂ binder	
Screen printing	TiO ₂ nanoparticles	[43]
	Pre-dyed ZnO	[21]
	TiO ₂ -ZnO	[57]
Pulsed laser deposition	Nanoporous ZnO film	[30]
Electrospray deposition	TiO ₂ nanoparticles	[58]
Electrophoretic deposition	ZnO + graphene oxide	[59]
	ZnO nanoparticles	[60]
Electro deposition	TiO ₂ nanoparticles	[61]

From Table 2 and in literature, it can be observed that physical methods of deposition particularly doctor blade method, has been the preferred choice of technique. The simplicity, ease of operation and low cost of the technique has made them a popular choice amongst researchers. Physical method especially doctor blade method is expected to continue being widely used in studies at various levels as the method also uses minimal and simple equipment. However, the main issue is the efficiency of DSSC prepared by low temperature fabrication technique is lower than the ones at high temperature. Thus, adding some post-treatment methods such as hot-compress treatment [60] and laser sintering [54] can help to increase the inter-particle connectivity of the photoanode for better efficiency. But this will then take away the simplicity, low cost and quick overall process from using just one of the fabrication techniques mentioned. Hence, researchers have been working on developing better DSSC components as to avoid adding more process and cost of production.

4. Recent advancements in flexible low temperature DSSC

There have been numerous research and development conducted in the field of solar cell technology especially for dye sensitised solar cells. Various different type of DSSCs have been developed over the years either by introducing new materials to construct DSSC component or studying the parameters and operating conditions when constructing the solar device. This chapter will look into some of the recent advancements of DSSC development including for flexible and low temperature DSSC

to give an insight on the research trend on this ever-growing topic by going over the DSSC components and modules.

4.1 Conductive substrate

Conductive substrate usually makes up the main bulk of the DSSC structure and there are usually two of them with the rest of the components located in between them. They are used to prepare the photoanode and counter electrode for the assembly of the device by acting as support for the photoanode materials and counter electrode catalyst [32]. A substrate that is suitable for DSSC require two main characteristics: i) to have 80% transparency for the passage of optimum sunlight into the cell ii) to have high electrical conductivity for efficient charge transfer and minimising energy loss [19]. Thus, fluorine tin oxide (FTO) and indium tin oxide (ITO) glass substrate has been the commonly applied substrate for DSSC.

The use of glass substrate is normally associated with the construction of the rigid and high temperature DSSC which can be seen in a lot of studies such as Razamin et al. [62], Shashanka et al. [63], Oh et al. [64] and Sharif et al. [65] where FTO glass substrates were coated by various materials such as TiO_2 and ZnO photoanode as well as Pt, nickel selenide and iron selenide counter electrode catalyst. The FTO glass was sintered at high temperature of around 450-500°C as part of the preparation process of the electrodes, producing conversion efficiency in the range of 1.01-8.03%. Meanwhile, the application of ITO glass can be seen in research by Murugadoss et al. [66] and Mahmoud et al. [67] where both ITO glass was sintered at high temperature of 450°C for the preparation of TiO_2 photoanode, yielding efficiency of 7.41% and 1.59% respectively. The thermal stability of the glass substrates at high temperature is one of the reasons they are commonly used for DSSC construction, but they can also be effective at low temperature. This is demonstrated in a study by Mustafa et al. [68] where ITO glass was sintered at relatively low temperature of 200°C to prepare TiO_2 photoanode, producing efficiency of around 3.72-4.07%. ZnO and ZnO @carbon dot was coated on ITO glass before being annealed at 100°C by Gopal et al. [69] to be formed as photoanode that resulted in efficiency of 8% and 7% respectively. Hence, indicating the capability of glass substrate to operate at various temperature. Among these two however, FTO glass is the more commonly used substrate, as FTO glass show higher efficiency compared to ITO glass due to their quick charge transportation and also the relatively lower price of fluorine and tin for FTO glass compared to indium for

ITO glass [70,71]. Even though the glass substrates are widely used in DSSC, the rigidity of these glass substrates causes them to have poor ductility and limits their flexibility, making them less suitable for flexible DSSC applications that have been growing in popularity [37]. Therefore, other materials have been applied as substrate for the construction of flexible DSSC.

Flexible DSSC normally uses plastic substrates such as ITO-polyethylene naphthalate (PEN) and ITO-polyethylene terephthalate (PET). These substrates are chosen due to their good flexibility, conductivity, lightweight, transmittance and ability to be mass produced via roll-to-roll process at lower cost [24,35]. However, these plastic substrates will melt or deform when operated at high temperature ($>150^{\circ}\text{C}$) due to their thermal instability at such conditions [28]. Therefore, the construction of DSSC devices using such substrate would be done at low temperature ($\leq 150^{\circ}\text{C}$) which can be observed from several studies conducted recently. Al-Khafaji et al. [72] conducted a study by coating TiO_2 on both FTO glass and ITO-PET plastic substrates via doctor-blade method. The study discovered that the ITO-PET based DSSC would always yield lower conversion efficiency compared to FTO-based DSSC at any variables with the highest efficiency recorded at 1.8% and 2.3% respectively. ITO-PEN substrate was coated by TiO_2 photoanode and carbon counter electrode in a study by Chuang et al. [73] that resulted in conversion efficiency of around 1.32-2.54%. Different plastic substrate, ITO-polyetherimide (PEI) plastic substrate have higher thermal stability than PET and PEN to produce a more efficient solar cell [74]. By radio frequency (RF) magnetron sputtering, PEI substrates were deposited on ITO surface reported in Wante et al. [28] and Wante et al. [20] where a PCE of 2.8% was produced in both studies.

Low sintering temperature for the construction of DSSC photoanode causes slower electron transport rate which encourages recombination reaction, weak charge collection efficiency and bigger intrinsic resistance that eventually lowers their efficiency [24,26]. This has been an issue for the use of plastic substrate as shown in some of the examples, the conversion efficiency is found to be relatively low especially when compared to DSSC utilising glass substrate sintered at high temperature. However, the application of PEI polymer has shown there are room to improve the performance of plastic-based DSSC by developing more suitable polymers that can possibly withstand higher temperature or by developing sintering methods that allow the plastic substrate to be able withstand higher temperature. Some studies have even

use metal substrates as substitutes for plastic substrate in order to develop their flexible DSSC.

Metal substrates have good electrical conductivity, flexibility, ductility, thermal stability at high temperature, low sheet resistance and low cost that makes them an exciting alternative to ITO plastic substrate [4]. Ti metal foil has been utilised as metal substrates due to their resistance to the corrosive nature of the iodide-triiodide (I/I_3^-) electrolyte and they are usually chemically or physically treated to improve the photovoltaic performance [75]. Such example can be seen in a study by Rui et al. [76] where they compare the performance of flexible DSSC using untreated Ti foil substrate and hydrogen peroxide (H_2O_2) treated Ti foil substrate. As shown in Figure 6, the H_2O_2 treated Ti foil was found to have form well-connected three-dimensional network and continuous pores to produce better efficiency than untreated Ti foil with values of 4.98% and 3.74% respectively. However, Ti metal foil usually suffers from light opacity, limiting their application in flexible DSSC as compared to Ti metal mesh or wires that can allow the passing of sunlight [4,77]. The structure of Ti mesh can be seen in Figure 6 from a study by Luo et al. [77] where they managed to develop flexible DSSC based on mesh-vertically oriented TiO_2 nanotubes substrates via anodization method. The mesh and nanotube layers were found to have good light scattering ability with conversion efficiency of 1.67-2.66%. However, the high price of Ti limits their wider application, allowing substrates such as stainless steel with good corrosion resistance and low price to be utilised in flexible DSSC even if their performance is not as good as Ti substrate [24]. By using stainless steel for both photoanode and counter electrode substrates, Yang et al. [78] managed to develop a flexible DSSC with PCE of 0.83%. Treatment or modification of the substrate could help to enhance their performance as demonstrated by Sheng et al. [23]. ZnO nanorod was added to the stainless-steel mesh, and they are treated with ammonium hexafluorotitanate ($(NH_4)_2TiF_6$). They discovered that the treated substrate yielded higher conversion efficiency of 1.93-2.70% compared to untreated substrate (0.98%) with lower recombination reaction and better electron injection.

Another alternative for flexible DSSC substrate also includes the application of textile or fabric materials via two different configuration methods, namely layer stacking [79] that are simple and effective or yard intersection [80], suitable for a large-scale and flexible textile DSSC development [81]. Layer stacking involved transferring as-prepared solar cell onto the textile substrates as demonstrated by Liu et al. [79] where

TiO₂ film was screen printed onto woven polyester cotton fabric and glass fibre textile. The research yielded a relatively decent conversion efficiency of 3.24% and 4.04% for woven polyester cotton fabric and glass fibre textile respectively. However, after three months, the DSSCs were found to have lower conversion efficiency due to the films cracking and peeling off before eventually dissolving in the electrolyte. Thus, highlighting the lack of durability and is an issue for such DSSC device. In yarn intersection, the photoanode yarn and counter electrode yarn are usually woven into a textile form as shown in Figure 6. By interlacing the sensitised ZnO nanoarrays photoanode yarns with the Cu-based counter electrode yarn, a flexible, lightweight and low-cost textile DSSC with efficiency of 1.3% was developed [82]. Meanwhile, Gao et al. [80] was able to develop a large-scale textile DSSC with conversion efficiency of 1.92% by intersecting the TiO₂ photoanode yarn with the silver (Ag)-plated nylon counter electrode yarns that are also compatible with conventional clothes. Kohn et al. [83] took a different approach by preparing a full nanofiber DSSC via electrospinning both the TiO₂ photoanode and graphite counter electrode catalyst coated with poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS), serving as the conductive substrate. Textile DSSC with conversion efficiency of 0.0014% was developed without short-circuit issues despite the low efficiency compared to glass-based DSSC (0.037%). Similar to plastic-based flexible DSSC, textile DSSC still suffers some limitations, mainly the inability to sinter the TiO₂ layer at 500°C to form a porous structure and fix them to the fabric surface [84]. Thus, making the development of low temperature textile DSSC a major challenge to researchers.

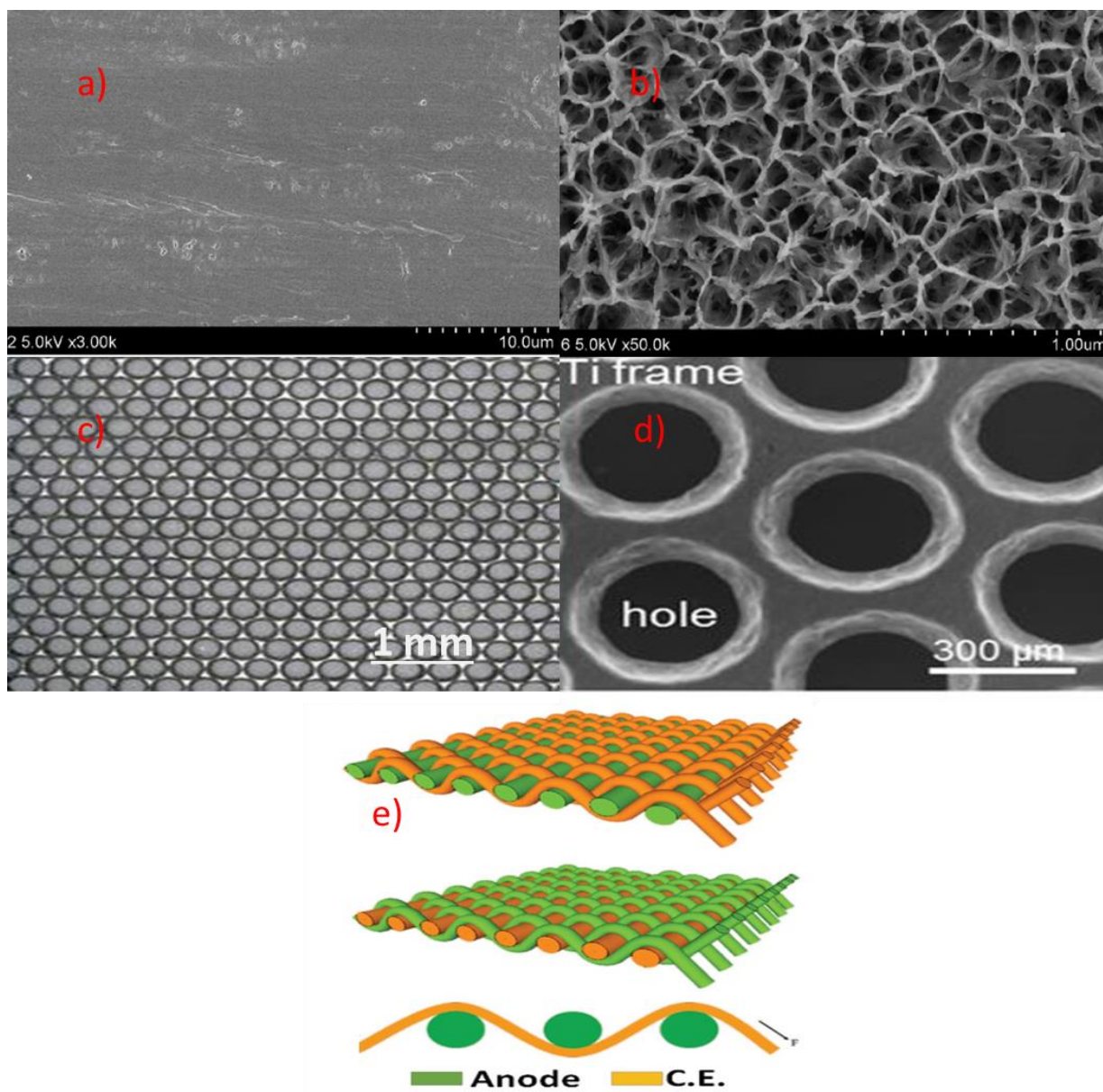


Figure 6 a) & b) FESEM image of untreated Ti foil and H_2O_2 treated Ti foil [76] c) & d) FESEM images on the surface of Ti mesh at *different magnification* [77] e) Schematic diagram of textile DSSC based on yarn intersection of photoanode and counter electrode yarn woven together [82]

From some of the examples in literature, it is obvious that the glass-based DSSC has produced better conversion efficiency than flexible DSSC. The low thermal stability of the plastic and textile substrate have remained a major stumbling block in developing a more efficient flexible DSSC. Developing polymer-based substrate with high thermal stability could be an approach to address the low sintering temperature issue for flexible DSSC that will ultimately increase the efficiency of the solar device. Apart from that, applying different sintering approaches such as laser sintering [54] and

plate fin heat sink assisted elevated temperature sintering [85] could be done without damaging the plastic substrate, particularly for the latter, where the ITO-PET substrate was still fine even at 250°C sintering. Therefore, there are still more potential and research gaps that can be explored in enhancing the sintering process in DSSC development. Although metal substrates have shown good efficiency and are flexible, their flexibility are still limited when compared to plastic or textile-based DSSC which would limit their further application. The stability issues for flexible DSSC based on plastic and textile substrate due to the leakage of electrolyte can be addressed with the use of gel electrolyte from non-toxic polymer such as Polyethylene oxide (PEO) [86]. Developing plastic-based DSSC should be the higher focus as compared to other flexible substrates due to their cost while having more upsides and wider potential application.

4.2 Photoanode

Photoanode are usually prepared by coating one of the glass or plastic substrate with a semiconducting material layer. The function of the photoanode is supporting the sensitiser loading and transporting the photoexcited electrons from the sensitiser to the external circuit [87]. Material that possesses large surface area and quick charge transport rate is a candidate to become an ideal photoanode material as they allow higher dye loading for light harvesting purposes and higher efficiency for electron collection. Apart from that, having good structural arrangement and chemical stability while still being low cost and environmentally friendly are some other desirable characteristics for ideal photoanode materials [88]. Hence, wide-band gap semiconducting metal oxides such as zinc oxide (ZnO) and titanium dioxide (TiO₂) are some of the most commonly used photoanode materials in DSSC. These semiconducting metal oxides are usually used as photoanodes in their nanocrystalline form to obtain a large surface area for better light absorption [89].

4.2.1 Titanium dioxide (TiO₂)

Titanium dioxide is commonly used as photoanode materials as they cost less, easily available, non-toxic and has a large surface area, maximising dye loading and light absorption [90]. TiO₂ is classified into different crystalline forms: i) brookite ii) rutile, the most thermodynamically stable and iii) anatase, the most preferred in DSSC with better conversion efficiency [40]. TiO₂ has been a subject of interest for a lot of DSSC research and development with

multiple efforts conducted to improve their performance via various factors including their nanostructures and morphologies.

The nanostructures of TiO_2 are important in determining their performance as DSSC photoanode due to each nanostructures possessing different properties, especially with regards to the surface area [91]. Mesoporous TiO_2 nanoparticles are usually used in DSSC, but several other nanostructures such as nanowires, nanotubes, nanorods and nanofibers have also been studied in literature [40]. DSSC assembled with TiO_2 nanowires was found to have better performance than DSSC assembled with commercial TiO_2 nanoparticles (P25) according to a study by Hu et al. [92]. Higher conversion efficiency was obtained when TiO_2 nanowires was used with a value of 4.04% compared to P25 (3.14%) with higher short-circuit current density, fill factor and electron lifetime. The result is attributed to the fast interfacial electron transport in the one-dimensional nanowires that made them a promising TiO_2 nanostructure.

TiO_2 nanotubes has been reported to exhibit better electron transport efficiency with high potential to be used as DSSC photoanode [93]. TiO_2 nanotubes was also found to have better dye loading and dye absorption capacity than TiO_2 nanowires due to the high active surface area present in the structure as reported by Cai et al. [94] and Gnida et al. [95]. TiO_2 nanotubes have also been utilised as photoanode in flexible DSSC development by using them on Ti metal substrates due to their ability endure stress under external bending and can be used in complex environments [96].

These one-dimensional nanostructure however, still suffers from low specific surface area which can be overcome by the development of nanoarrays. Xiao et al. [97] managed to synthesise mesoporous nanowire arrays with a large specific area and good crystalline structure. An increase in efficiency can be observed for DSSC with TiO_2 mesoporous nanowire (3.20%) compared to DSSC with the usual TiO_2 nanowires (0.28%). Hierarchical TiO_2 nanowire arrays were found to have increased the internal surface area and light scattering properties according to Liu et al. [98]. The flexible DSSC based on the developed TiO_2 nanostructure produced better efficiency than regular nanowire arrays with values of 3.85% and 3.31% respectively. By growing the TiO_2 nanotube arrays on all sides of the metal grids of Ti mesh as shown in Figure 7,

more sunlight was able to be harvested from multiple angles resulting in conversion efficiency of more than 5%, which was rather high for a flexible DSSC [99]. Another study that managed to produce a flexible DSSC with high PCE (5.3%) was done by Lu et al. [100] using TiO₂ nanorod arrays photoanode on ITO-PEN plastic prepared via RF reactive magnetron sputtering technique.

TiO₂ nanostructures have shown to be an important parameter in determining their performance as DSSC photoanode and should be investigated farther. Another approach that can be taken is by combining several nanostructures to form hybrid nanostructure that can further enlarge the surface area and improve the light scattering capacity for better efficiency. However, careful consideration needed to be made when choosing which nanostructures to combine as unsuitable combination would cause difficulties when absorbing the dye molecules, causing lower efficiency. Apart from that, another factor to consider when developing these nanostructures are that the process should be easy and simple to both create as well as recreate, otherwise it will not be suitable for wider commercial application.

Table 3 Non-flexible and flexible DSSC based on TiO₂ nanostructure photoanode

TiO₂ nanostructure	Conversion efficiency (%)	Reference
<i>Non-flexible DSSC</i>		
Nanowires	4.04	[92]
Mesoporous nanowire arrays	3.20	[97]
Rough-surface nanowire arrays	8.28	[101]
Nanotubes	5.33	[94]
Nanowires	4.36	
Nanoparticles	6.69	[95]
Nanoparticles + nanotubes	6.97	
Nanoparticles + nanowires	5.44	
Nanorods	2.01	[102]
<i>Flexible DSSC</i>		
Hierarchical nanowire arrays	3.85	[98]
Nanotube arrays	1.67-2.66	[77]
Nanotube arrays	5.00	[99]

Nanotubes	5.71	[103]
Nanotube arrays	6.25	[104]
Nanorod arrays	5.30	[100]

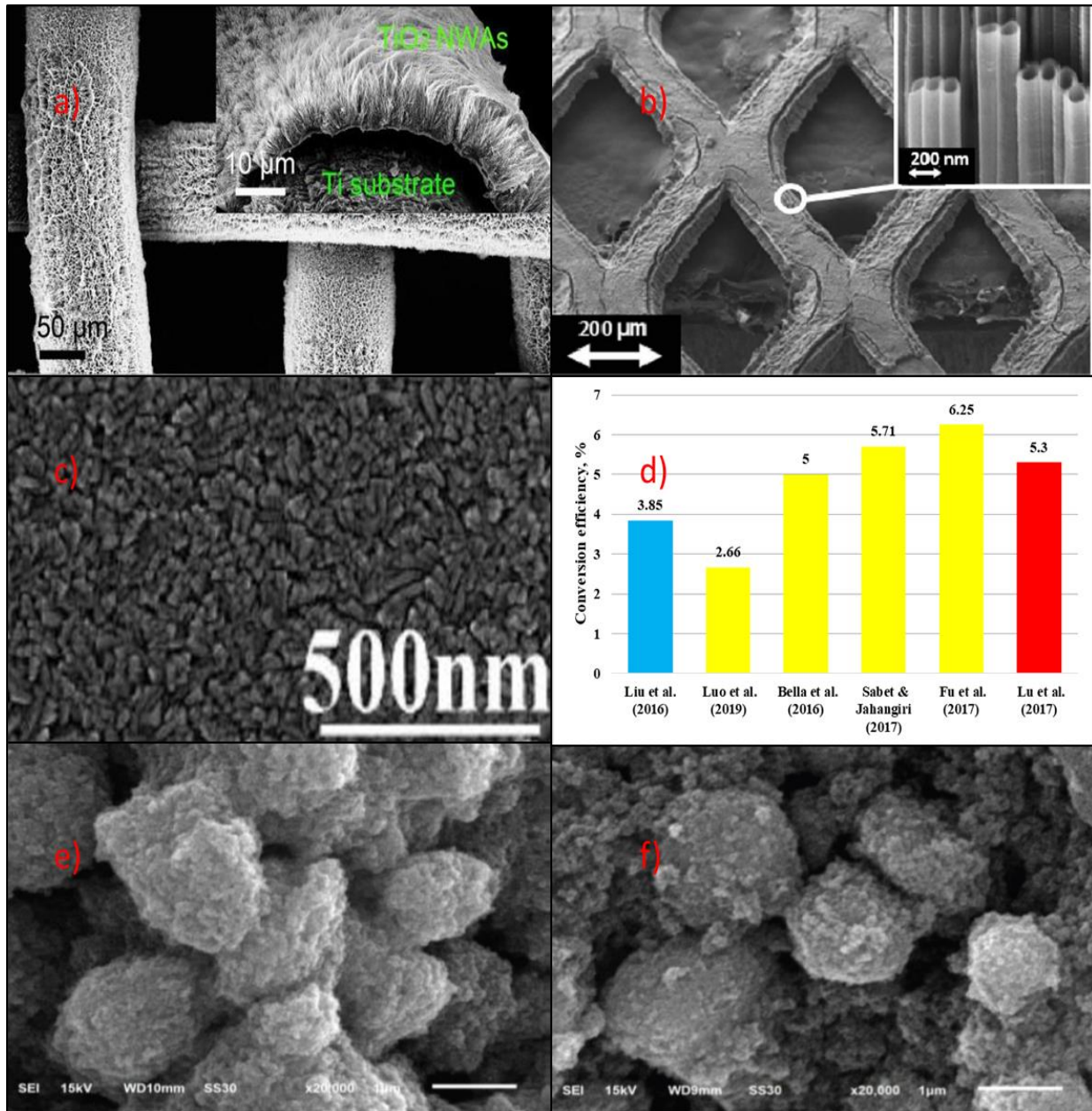


Figure 7 a) FESEM image of TiO_2 nanowire arrays on Ti mesh [105] b) FESEM image of TiO_2 nanotubes on Ti mesh [99] c) SEM image of TiO_2 nanorod arrays on ITO-PEN plastic [100] d) Chart comparing the conversion efficiency of several flexible DSSC using different TiO_2 nanostructures e) & f) SEM images of binder-free TiO_2 photoanode and binder-added TiO_2 photoanode showing small particles, TTIP filling in the cavities between TiO_2 particles [27]

Another approach to enhance the performance of TiO₂ photoanode is by doping them with metal cations, precious metals and non-metallic anions [24]. Doping TiO₂ with metallic cations allow the change in band gap, increase electrical properties, reduce recombination and also effect the PCE of DSSC [106]. The doping of TiO₂ nanowire arrays photoanode with metal cation, tin (Sn), was found to have boosted the electron mobility and increases the flat band potential according to Ni et al. [107]. This can be seen with the high conversion efficiency of 8.75% produced by the DSSC which can even be enhanced to 9.43% when added with anatase TiO₂ light scattering layer. Meanwhile, metal cations from magnesium (Mg²⁺) ions were discovered to change conduction band and state of the surface of TiO₂, limiting the electron recombination according to Song et al. [108]. This was proven as the flexible DSSC based on the Mg²⁺ doped photoanode yielded higher efficiency compared to undoped ones with values of 2.2% and 1.7% respectively.

Noble metals such as silver and gold has also been utilised as dopant material for TiO₂ photoanode by making use of their plasmonic effect. Silver (Ag) doped with TiO₂ photoanode reduced the charge transfer resistance, increase charge transport and charge carrier lifetime that eventually reduces recombination as demonstrated by Saravanan et al. [109]. The DSSC produced a conversion efficiency of 3.62%, a 28% increase from DSSC with undoped TiO₂ photoanode. The plasmonic effect of the silver nanoparticles was concluded as the deciding factor in enhancing the conversion efficiency of DSSC. Gold nanoparticles has better stability in iodide-based electrolyte compared to silver nanoparticles and the plasmonic effect of the gold nanoparticles managed to enhance the conversion efficiency of the DSSC by almost 50% with a value of 3.12% reported by Jun et al. [110].

Co-doping erbium (Er³⁺) ions and ytterbium (Yb³⁺) ions into TiO₂ nanowires/nanoparticles composite on Ti mesh managed to produce quick transfer of electron, good light-up conversion and lowers the electron recombination losses according to a study by Liu et al. [105]. A 68% improvement in conversion efficiency was recorded in the study as the flexible DSSC with the co-doped photoanode produced a value of 8.10% compared to undoped flexible DSSC (4.82%). Co-doping could be a promising approach for

DSSC as they combined the benefits of both suitable dopants to produce superior performance.

All in all, the doping of TiO₂ with wide variety of metallic and non-metallic dopants have generally managed to enhance the efficiency of both flexible and non-flexible DSSC by tuning the band structure of TiO₂. Although a wide variety of dopants have been studied, there is still the question on what is the most suitable dopant for TiO₂ photoanode as the efficiencies obtained from literature for each dopant are still scattered. Therefore, opportunities to find the best dopant for TiO₂ as well as determining the optimum dopant concentration still exist.

Table 4 Dopants used with TiO₂ photoanode for both flexible and non-flexible DSSC

Dopant	Conversion efficiency (%)	Conversion efficiency with undoped TiO₂ (%)	Reference
<i>Non-flexible DSSC</i>			
Cr	3.89	1.05	[111]
Sn	8.75	7.46	[107]
Sn	4.01	0.87	[112]
Vanadium (V)	3.33	1.31	[113]
Iron (Fe)	4.85		[113]
Germanium (Ge)	5.65	4.03	[114]
Ag	3.62	2.83	[109]
Au	3.12	2.09	[110]
Au	6.80	5.66	[115]
Nanodiamonds	0.74	0.26	[116]
<i>Flexible DSSC</i>			
Nb	7.20	4.96	[117]
Mg ²⁺	2.20	1.70	[108]
Er ³⁺ - Yb ³⁺	8.10	4.82	[105]

Several other approaches have been taken by researchers to improve the performance of TiO₂ photoanode at low temperature operation that are suitable

for flexible DSSC. Binder addition is a good approach to improve the performance of TiO₂ photoanode by improving the connection of TiO₂ particles, increasing the surface area and porosity as demonstrated by Muliani et al. [27]. Titanium isopropoxide (TTIP) was used as binder added to TiO₂ coated on ITO-PET plastic and sintered at low temperature of 120°C. The binder-added photoanode was found to have small particles of TTIP filling in the spaces between the TiO₂ particles, creating connection between them for better electron pathway as shown in Figure 7. Thus, producing a higher conversion efficiency than binder-free photoanode flexible DSSC with values of 0.31% and 0.18% respectively.

Another approach that can be taken is by adding sintering agent or additives during the low temperature sintering process to improve the inter-particle connectivity of the photoanode. An example can be seen in a study by Holliman et al. [52] that investigated the use of TiO₂ with chemical sintering agent, hexafluorotitanic acid (H₂TiF₆), sintered at low temperature of 120°C. Conversion efficiency of 4.2% was obtained when using ITO-PET substrate while the highest efficiency was obtained when FTO glass substrate was used with co-sensitiser at the same temperature. The use of the acid as sintering agent and co-sensitisation showed promise for low temperature processed DSSC and probably it could be applied for plastic substrate in future development. Ahmad et al. [118] meanwhile used zinc (Zn) nanoparticles as sintering additive to the TiO₂ nanoparticles photoanode that managed to increase the efficiency to 4.92% compared to bare TiO₂ (4.27%). The lower melting temperature of Zn nanoparticles improved the inter-particle connectivity due to the formation of necks at the TiO₂-Zn interface even at relatively low sintering temperature (200°C).

Graphene is another material that has been applied in DSSC as an electron acceptor that can be converted into graphene quantum dot (GQD) by tuning the band-gap [53]. GQD has also been used to form composite photoanode with TiO₂ for flexible DSSC [53]. Binder-free TiO₂ was prepared with GQD on ITO-PEN film, where a conversion efficiency of 4.43% was exhibited in the study. Although there was limited improvement when compared to binder-free TiO₂ photoanode (4.2%), the improvement of the overall performance such as longer life-time, reduced recombination, improved

charge transport from the use GQD-TiO₂ composite made them an exciting prospect for flexible DSSC application. From literature, it can be seen that the efficiency of flexible low temperature DSSC has not improved significantly with the addition of either binder and sintering agents as shown in Table 5. Indicating there are still more research needed to be conducted in this topic by studying the effects of several parameters that might affect their performance such as the amount or composition of the additional material, sintering temperature and duration. Apart from that, using other materials at smaller sized particles could also be done to further improve the inter-particle connection of TiO₂ photoanode.

Table 5 Studies on flexible DSSC and low temperature non-flexible DSSC using TiO₂ photoanode

Photoanode	Conversion efficiency (%)	Reference
TiO ₂	0.18	[27]
TiO ₂ + TTIP	0.31	
TiO ₂		[43]
Screen printing	1.04-4.30	
Spray coating	1.06-1.29	
TiO ₂ + Triton X-100 via		
Screen printing	0.41-0.77	
Spray coating	1.89-2.58	
TiO ₂ + H ₂ TiF ₆	4.20	[52]
TiO ₂	4.27	[118]
TiO ₂ + Zn	4.92	
TiO ₂	4.20	[53]
TiO ₂ + GQD	4.43	

Several conclusions can be made on the application of TiO₂ photoanode for both flexible and non-flexible DSSC. 1) The nanostructures of TiO₂ play an important part in determining the surface area, dye and light absorption that eventually led to the efficiency of DSSC device. 2) Modifications of TiO₂ via the doping process has managed to enhance the efficiency of DSSC and should be further investigated to find how different dopants effects the band structure

and surface modifications of TiO₂ photoanode. This is especially since the most suitable dopant for TiO₂ are still up for debate with a lot of different available materials to test from. 3) Binder or sintering agent addition have yet to significantly enhance the efficiency of flexible DSSC. However, the use of sintering agents for TiO₂ photoanode showed promise in improving efficiency by proper sintering. Using materials with low melting point as sintering agent allowed the formation of necks at the TiO₂ interface for better inter-particle connection and efficiency even at lower sintering temperature. Despite the advantages and good performance of TiO₂, there have also been demands in using different material as an alternative to improve the charge collection and reduce the energy losses during electron injection [119].

4.2.2 Zinc oxide (ZnO)

Zinc oxide (ZnO) has been widely explored as an alternative to TiO₂ photoanode than other metal oxides due to their similar bandgap of around 3.37 eV, high electron mobility and better electron diffusivity [120]. Although DSSC based on ZnO photoanode was not able to produce conversion efficiency as high as TiO₂-based DSSC, they have better mobility and carrier lifetime of electrons with easy synthesis that encourages their application in DSSC [32]. Similar to TiO₂, most conventional methods in fabricating ZnO nanoparticles involved sintering them at high temperature of 450-500°C for better inter-particle connection [121].

One of the characteristics of ZnO allowed them to be fabricated into various different nanostructures such as nanotubes, nanoparticles, nanowires, nanoflowers and nanoclusters [40]. One advantage of using ZnO nanostructure as photoanode can be seen in a study by Jamalullail et al. [122] that compared the performance of conventional TiO₂ with ZnO nanorods annealed at 450°C. DSSC based on ZnO nanorods yielded conversion efficiency of 0.34% compared to DSSC based on conventional TiO₂ with 0.29% due to the bigger particle size of nanorods structure for better dye loading and direct pathways for electrons movement. High electron mobility (155 cm²/Vs) of ZnO was also concluded as another contributing factor for their superiority in performance. Ling & Ahmad [123] increased the concentration of ZnO solution in preparing the photoanode to increase the thickness and diameter of nanorods, preventing

the increase in resistance. The study managed to produce DSSC yielding highest efficiency of 1.73% by conducting the process at low annealing temperature of 100°C, suitable for flexible DSSC.

Pandey et al. [124] discovered the optimal annealing temperature for the fabrication of ZnO nanoparticles was 400°C with conversion efficiency of 3.35% with good dye absorption, high charge transport and lower charge recombination. Such high optimal annealing temperature would pose a problem for the application of ZnO nanoparticles in flexible low temperature DSSC. However, by applying post-deposition treatment such as hot-compress, the ZnO nanoparticles film managed to form a homogenous compact layer with good adhesion to the flexible substrate [60]. A decent conversion efficiency of 2.63% was achieved when the ZnO nanoparticles coated on ITO-PET plastic was compressed at 130 MPa and 70°C, producing a homogenous and smooth surface for better electron transport as shown in Figure 8.

The properties of ZnO have also allowed them to be synthesised into several different morphologies such as nanoprism, hollow nanoprism and nanorice [125]. By changing the polarity of solvent, different ZnO nanostructures was able to be synthesised with the nanorice structure producing the highest conversion efficiency of 2.11% compared to nanoprism (1.11%) and hollow nanoprism (0.18%). The superiority of ZnO nanorice was attributed to the larger surface area as shown in Figure 8 for better conductivity and dye absorption. The immersion of ZnO films in mixed solvents could improve their microstructures for better inter-particle connections according to Ohashi et al. [126]. The study managed to fabricate two different ZnO morphologies: flower-like particles and densely-packed nanoparticles on plastic substrate as shown in Figure 8 for flexible DSSC with conversion efficiency of 3.9% and 4.1% respectively.

Table 6 Non-flexible and flexible DSSC based on ZnO nanostructure photoanode

Nanostructure	Annealing/ sintering temperature (°C)	Conversion efficiency (%)	Reference
<i>Non-flexible DSSC</i>			
Nanorods	450	0.34	[122]

Nanorods	100	1.73	[123]
Nanorods	500	2.35	[127]
Nanoparticles	400	3.35	[124]
Nanoparticles	400	6.20	[128]
Nanowire arrays	300	0.63	
Nanoprism	350	1.11	[125]
Hollow nanoprism		0.18	
Nanorice		2.11	
<i>Flexible DSSC</i>			
Nanoparticles	100	2.63	[60]
Nanowire arrays	350	1.78	[129]
Nanosheet		2.71	
Flower-like particles	120	3.90	[126]
Densely packed nanoparticles		4.10	

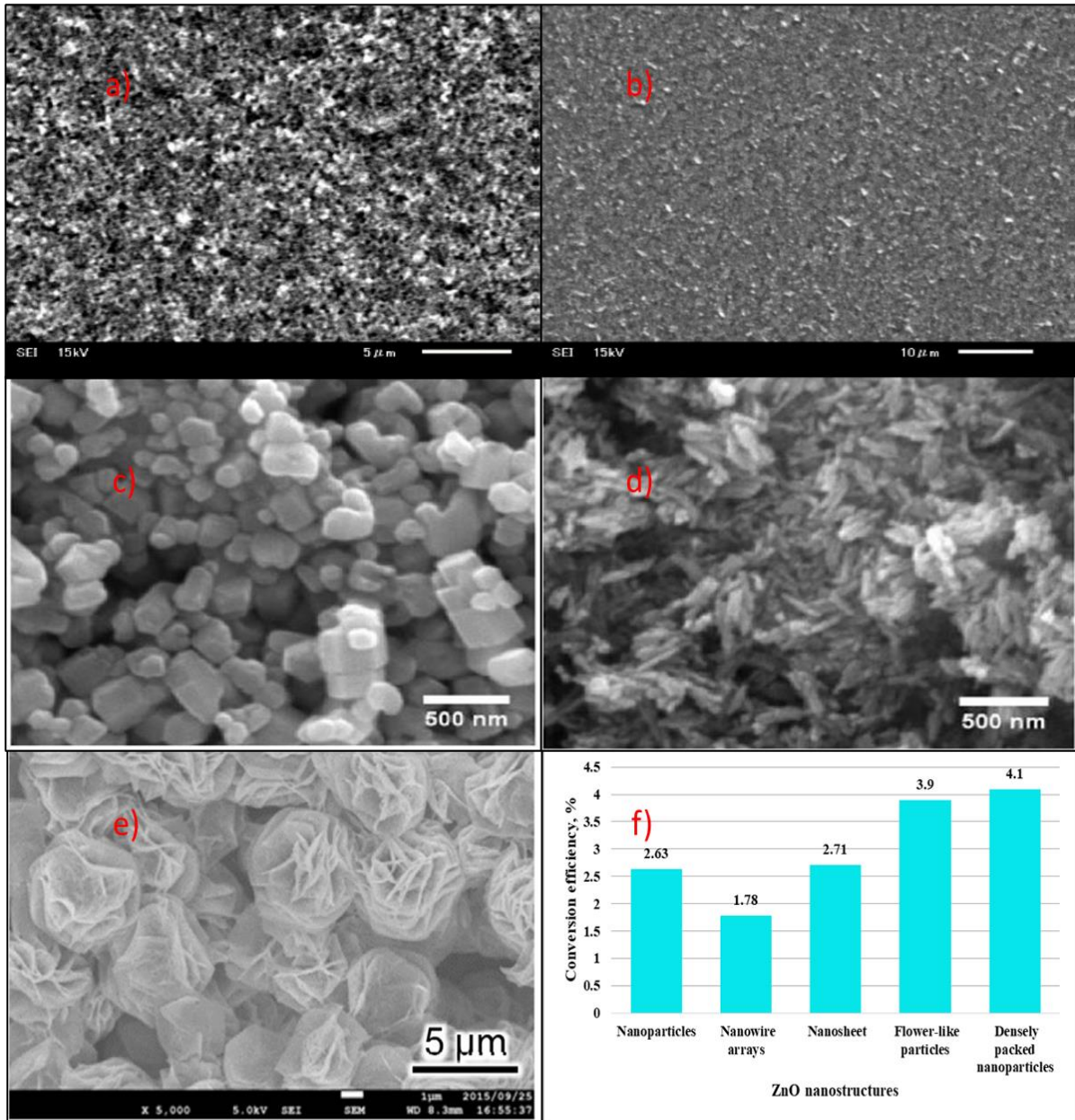


Figure 8 a) & b) SEM images of ZnO nanoparticle without hot-compress treatment & smooth ZnO nanoparticle surface after hot-compress treatment [60] c) & d) SEM image of ZnO nanoprism & SEM image of ZnO nanorice with larger surface area [125] e) FESEM image of ZnO with flower-like particles nanostructure [126] f) Chart comparing efficiency of several ZnO nanostructures for flexible DSSC

The versatility of ZnO to be fabricated into various morphologies with varying degree of success in increasing the surface area and efficiency of DSSC photoanode have made them a good alternative to TiO₂. However, similar to TiO₂, the complexity in creating some of the ZnO nanostructures might be a stumbling block for wider commercial application. Also, forming hybrid nanostructure is another alternative to enhance the performance of ZnO

photoanode as long as the combined nanostructures are suitable and work well together.

ZnO doping have also been applied in order to tune the bandgap and surface modification, enhancing the efficiency of DSSC by using metal cations, non-metal anions and noble metals. The low toxicity and source abundance of copper (Cu) has made them a desirable dopant to ZnO photoanode by being cost-effective, prevent recombination and improve light harvesting efficiency [130,131]. Esgin et al. [130] discovered that the incorporation of Cu^{2+} ions formed relevant oxides on the surface of ZnO to increase light dispersion and inhibits the agglomeration of Zn^{2+} /dye. The study reported higher conversion efficiency of 2.03% by doping Cu into ZnO nanopowder which was a 20% increase from undoped ZnO nanopowder.

Doping ZnO nanoparticles with Nickel tetraphenylporphyrin (NiTPP) was found to have improved the electron transfer, light scattering and generation of excitons that eventually improve the PCE of flexible DSSC [132]. Highest conversion efficiency of 2.7% was reported, which was a 42% increase from flexible DSSC using undoped ZnO photoanode. However, the doping of ZnO nanoparticles is to be conducted at low NiTPP concentration, as ZnO nanoparticles doped at high concentration of NiTPP causes structural defects, aggregation and low electron transfer.

Table 7 Dopants used with ZnO photoanode for flexible and non-flexible DSSC

Dopants	Annealing/ sintering temperature (°C)	Conversion efficiency (%)	Conversion efficiency with undoped ZnO (%)	Reference
<i>Non-flexible DSSC</i>				
Cu	450	2.03	1.41	[130]
Ag	550	0.44	0.42	[133]
Magnesium (Mg)	500	3.53	n/a	[134]
Lithium (Li)	400	1.23	0.50	[135]
Cobalt (Co)	400	1.67	n/a	[136]

<i>Flexible DSSC</i>				
NiTPP	100	2.70	1.86	[132]

Similar to TiO₂, doping ZnO with metallic and non-metallic dopants have brought positive effects in the performance of the photoanode materials as shown in Table 7. However, the addition of dopants to ZnO photoanode have not brought a significance improvement to the efficiency of DSSC as well as there have not been as many studies conducted on ZnO dopants compared to TiO₂. Hence, an opportunity is present to investigate on suitable ZnO dopants as well as reasons as to why this approach is not as desirable when compared to TiO₂ doping.

One of the measures taken to enhance the performance of ZnO photoanode for flexible DSSC include the addition of binder. Adding binder such as TiO₂ to ZnO paste enhances the stability, adhesion and surface morphology of the ZnO film as reported by Choudhury et al. [56]. Better photovoltaic performance was produced with the highest conversion efficiency of 1.75%, higher than the flexible DSSC with binder-free ZnO film (0.63%). The study heat compressed the photoanode samples at 70°C as post-treatment process, indicating the suitability for flexible DSSC construction. Other than being used as binder, TiO₂ can also be used as buffer layer on the ZnO films to weaken the acidic corrosion and enhance performance [137]. A simplified method of ammonium hexafluorotitanate ((NH₄)₂TiF₆) treatment was used to prepare a TiO₂ buffer layer, coating the ZnO nanorods on stainless-steel mesh substrate [23]. A conversion efficiency of 3.12% was obtained in the study as the buffer layer helped weakened the recombination reaction, reduce the dye molecule aggregation while improving the photoexcited electron injection and conversion efficiency.

A derivate of graphene named graphene oxide, have also been incorporated into ZnO nanoparticles as reported by Atanacio-Sanchez et al. [59]. The flexible DSSC that uses graphene oxide-modified ZnO nanoparticles produced a better conversion efficiency than bare ZnO nanoparticles with values of 0.4992% and 0.4499% respectively. The increase was attributed to the improvement in the distribution of ZnO nanoparticles that enhances dye

absorption for better photovoltaic performance. Pre-dyeing ZnO nanoparticles would form a coloured nanoporous ZnO photoanode to remove the time-consuming dyeing process while the pre-dyeing process is simple and easy to perform [21]. A drawback in applying this method is the limited connection between the ZnO nanoparticles as well as the electrical contact between the ITO-PEN substrate and ZnO nanoparticles as the dye was covering the ZnO nanoparticles. Hence, a hot-press treatment was required afterwards with the highest flexible DSSC efficiency produced was 4.56%.

Similar to TiO₂, the addition of sintering agents and binder to ZnO photoanode have slightly increased the efficiency of flexible DSSC as shown in Table 8. There have to be more studies conducted in finding suitable materials to be incorporated with ZnO photoanode for flexible DSSC in the future as the semiconducting material have shown some promise and potential to be a good alternative to TiO₂ photoanode.

Table 8 Studies on flexible DSSC using ZnO photoanode

Photoanode	Annealing/ sintering temperature (°C)	Conversion efficiency (%)	Reference
ZnO	70	0.63	[56]
ZnO + TiO ₂ binder		1.75	
ZnO	300	2.84	[23]
ZnO + TiO ₂ buffer layer		3.12	
ZnO	120	0.45	[59]
ZnO + graphene oxide		0.50	
Pre-dyed ZnO	120	4.56	[21]

From literature, it can be seen that ZnO has showed a lot of promise to be applied as good DSSC photoanode for both flexible and non-flexible device and be comparable to TiO₂. However, ZnO still suffers from several drawbacks as their efficiency is still lower than TiO₂ photoanode due to slower electron injection kinetics as well as dissolving in iodide/triiodide electrolyte and instable in acidic dye [32,138]. Similar conclusions on the application of this photoanode can be drawn from TiO₂ with regards to the nanostructures, doping and addition of sintering agents and binders.

Both TiO₂ and ZnO are still commonly used as DSSC photoanode due to their superiority to other materials. Adding other materials for surface modifications, tuning bandgap and improving the inter-particle connection have shown to be good approach in improving the performance of both photoanodes even more. Another good alternative to TiO₂ and ZnO photoanode is by combining them together to form a composite to get the best of both worlds. Besides these two, there are always room for other semiconducting metal oxides such as tin oxide (SnO₂) due to their high bandgap value and electron mobility [119]. All in all, the study in DSSC photoanode component is expected to continue to develop in years to come with the introduction of other materials and binders, dopants and sintering agent to be added in addressing some of the ongoing issues found in this component.

4.3 Counter electrode

Counter electrode is placed on the opposite side of the photoanode by coating the conducting substrate with catalytic materials. The counter electrode collects the electrons from the external circuit in order to regenerate the redox couple. The catalyst materials used for the counter electrode are required to have high catalytic activity with high conductivity and low charge transfer resistance for charge transport to yield high efficiencies [139]. Apart from that, an optimal counter electrode catalyst should also have low cost, large surface area, chemical corrosion resistance and good adhesive with the conductive substrate [4]. Taking accounts most of these considerations, the precious noble metal platinum (Pt), has been the most commonly used catalyst material for the counter electrode in DSSC, producing high efficiency. Pt have also been used as catalyst for counter electrode for flexible DSSC and they can be found utilised in most of the literature mentioned in the previous sections. Several examples of flexible DSSC based on Pt as counter electrode catalyst can be observed in Table 9.

Table 9 Flexible DSSC based on Pt as counter electrode catalyst

Substrate	Photoanode	Manufacturing techniques	Conversion efficiency (%)	Reference
Ti foil	TiO ₂	Sputtering	2.66	[77]
ITO-PET			4.98	[76]
			2.20	[108]

ITO-PEN			3.85	[117]
Nickel EMTE		Pulse-electro deposition	5.67	[140]
Stainless- steel mesh	ZnO	Cyclic voltammetry	2.84	[23]
ITO-PET			2.02	
ITO-PET		Electrodeposition	0.50	[59]

Pt have been a commonly used counter electrode catalyst for flexible DSSC despite their high manufacturing temperature of around 400-500°C. This is mainly attributed to their superiority as counter electrode catalyst as reported by Sabet & Jahangiri [103], comparing the performance of flexible DSSC using several different counter electrode catalysts. The high catalytic activity, nobility and stability of Pt-based counter electrode was displayed as they produce the highest conversion efficiency for the flexible DSSC compared to graphene and multi-walled carbon nanotube (MWCNT) with values of 5.71%, 1.96% and 2.75% respectively. Similar results were obtained in another study, where the Pt-based flexible DSSC showed the highest conversion efficiency (4.4%) compared to graphene and MWCNT, even when both counter electrode and TiO₂ photoanode were deposited on ITO-PET substrate [141]. The performance of Pt-based counter electrode can be further enhanced by using different substrates as demonstrated by Khan et al. [140]. The study reported that the Pt nanoparticles coated on nickel (Ni) embedded metal-mesh transparent electrode (EMTE) produced higher conversion efficiency than Pt nanoparticles coated on the usual ITO-PEN plastic with values of 5.67% and 4.31% respectively. With excellent sheet resistance, good optical transparency and conductivity of Ni EMTE, a flexible DSSC with improved efficiency was obtained. The combination of other materials with Pt was found to have boosted the performance of flexible DSSC as demonstrated by Yue et al. [142]. The study combined Pt with nickel sulphide (NiS) and titanium (Ti) foil to form NiS/Pt/Ti and Pt/Ti as counter electrode catalyst for flexible DSSC with impressive conversion efficiency of 7.20% and 6.07% respectively. The addition of Pt was found to have formed a bump-like surface for the NiS/Pt/Ti counter electrode as shown in Figure 9 that provide a high effective contact area between the counter electrode with electrolyte, eventually leading to better electrocatalytic activity and

efficiency. The developed counter electrode showed promise for large scale flexible DSSC application due to their low cost, large surface area, good efficiency, excellent conductivity and low charge transfer resistance.

Table 10 Flexible DSSC based on Pt as counter electrode catalyst at low manufacturing temperature

Counter electrode catalyst	Temperature (°C)	Conversion efficiency (%)	Reference
NiS/Pt/Ti	100	7.20	[142]
Pt/Ti		6.07	
Pt nanofibers	70	3.82	[143]
Pt	70	7.29 for front illumination 5.85 for rear illumination	[144]
	80	5.71	[103]
	80	4.40	[141]

From Table 9 and Table 10, it can be seen that Pt has remained a popular choice as counter electrode catalyst in flexible DSSC with good PCE produced. However, there have been constant efforts being made to find an alternative to Pt due to their high cost, with Pt being a precious noble metal and are less abundant [19]. Thus, Pt-free counter electrode based on materials such as carbon, polymer and others have been developed with hope that they will be able to compete with the performance shown by Pt-based counter electrode [145].

Carbon is one of the most abundant materials on earth that can be found everywhere and has been deemed as an attractive alternative to Pt as counter electrode. This is due to several advantages such as low cost, high surface area, good thermal stability, electrical conductivity and catalytic activity with corrosion resistance towards iodine [4]. Several carbon materials have been used as counter electrode including graphite, carbon nanotubes, activated carbon, graphene and carbon black. A simple example of an application of carbon was demonstrated by Kharkwal et al. [146], using an inexpensive pencil lead graphite material as counter electrode. The experiment showed an impressive conversion efficiency of 5.57% and low series resistance, indicating good interface quality with low recombination sites due to the graphite's

good charge transport properties. Graphene's good flexibility and electrical conduction made them an interesting choice as counter electrode in flexible DSSC as reported by Lukaszewicz et al. [147]. The study found that the flexible DSSC based on graphene counter electrode yielded PCE of 3.95%, lower than Pt counter electrode (4.39%). However, the study suggests that graphene counter electrode might be further suited for flexible DSSC application as they were able to yield better efficiency after 100 bending cycles than Pt that suffered cracks and broken away from substrates after the bending tests as shown in Figure 9.

The performance of carbon counter electrode in DSSC can be further enhanced by mixing them with different carbon materials as shown by Zhang et al. [148] that combined graphite, conductive carbon black, graphene and carbon nanotubes at different mass ratio. The highest conversion efficiency yielded was 6.29%, with the mixture found to have improved the surface morphology, increase the charge transfer carrier at the electrolyte and counter electrode interface. The study also fabricated a flexible counter electrode by depositing the carbon mixture on ITO-PET substrate, producing a flexible DSSC with an efficiency of 4.32%. By mixing carbon nanotubes with Poly (diallyl dimethylammonium chloride) (PDDA) that have the ability to withdraw electrons as counter electrode, Kurokawa et al. [149] managed to develop a DSSC with PCE of 4.12% without even undergoing annealing process. The value was even higher than when carbon nanotubes was annealed at 400°C (3.49%). Thus, making them a suitable counter electrode for flexible DSSC construction. Another approach taken by researchers was to combine Pt with carbon to form an excellent catalyst for counter electrode by increasing the catalytic activity and lowering the cost of DSSC by reducing the dosage of Pt required [150]. Pt/carbon sphere composite counter electrode managed to yield high conversion efficiency of 9.02% in a small flexible DSSC and 6.26% in a largescale flexible DSSC, which was higher than flexible DSSC with bare Pt and bare carbon sphere counter electrode.

Table 11 Carbon counter electrodes used in flexible and non-flexible DSSC

Carbon counter electrode	Temperature (°C)	Conversion efficiency (%)	Reference
<i>Non-flexible DSSC</i>			
Graphite	25	5.57	[146]

Carbon nanotubes + graphite + conductive carbon black + graphene	25	6.29	[148]
Activated carbon	400	0.221	[151]
Carbon nanopowder		0.005	
Carbon nanotubes	25	2.78	[149]
Carbon nanotubes	400	3.49	
Carbon nanotubes + PDDA	25	4.12	
<i>Flexible DSSC</i>			
Carbon nanotubes + graphite + conductive carbon black + graphene	25	4.32	[148]
Ag @ zirconium dioxide/carbon (ZrO ₂ /C) nanofiber	120	4.77	[58]
Pt/carbon sphere for small-scale	100	9.02	[150]
Pt/carbon sphere for largescale		6.26	
Nanodiamonds/Zn	90	8.36	[152]

Apart from platinum and carbon, there are also other materials that have been used as counter electrode in flexible DSSC. Poly(3,4-ethylenedioxythiophene) or PEDOT is a conductive polymer that has received a great deal of attention as counter electrode owing to their good conductivity, electrochemical stability and catalytic activity, diverse structure, lightweight and easy processing with ability to be used as transparent electrode [139,153]. PEDOT and PEDOT/Pt counter electrode was used in flexible DSSC in order to replace the expensive Pt or decrease the amount required to use Pt [153]. The PCE of the flexible DSSC was 4.04% and 5.04% for PEDOT and PEDOT/Pt counter electrode respectively. The flexibility, lightweight and portability of flexible PEDOT and PEDOT/Pt counter electrode made them an exciting alternative, particularly PEDOT, for fabricating flexible DSSC. The excellent catalytic activity as well as large specific surface area, high mobility and high electric conductivity of semiconducting material such as molybdenum disulphide (MoS₂), has made them an option as flexible counter electrode as investigated by Gurulakshmi et al. [154]. The

study reported that the use of MoS₂ counter electrode coated on FTO-PET substrate via electrodeposition, managed to produce PCE of 4.35%.

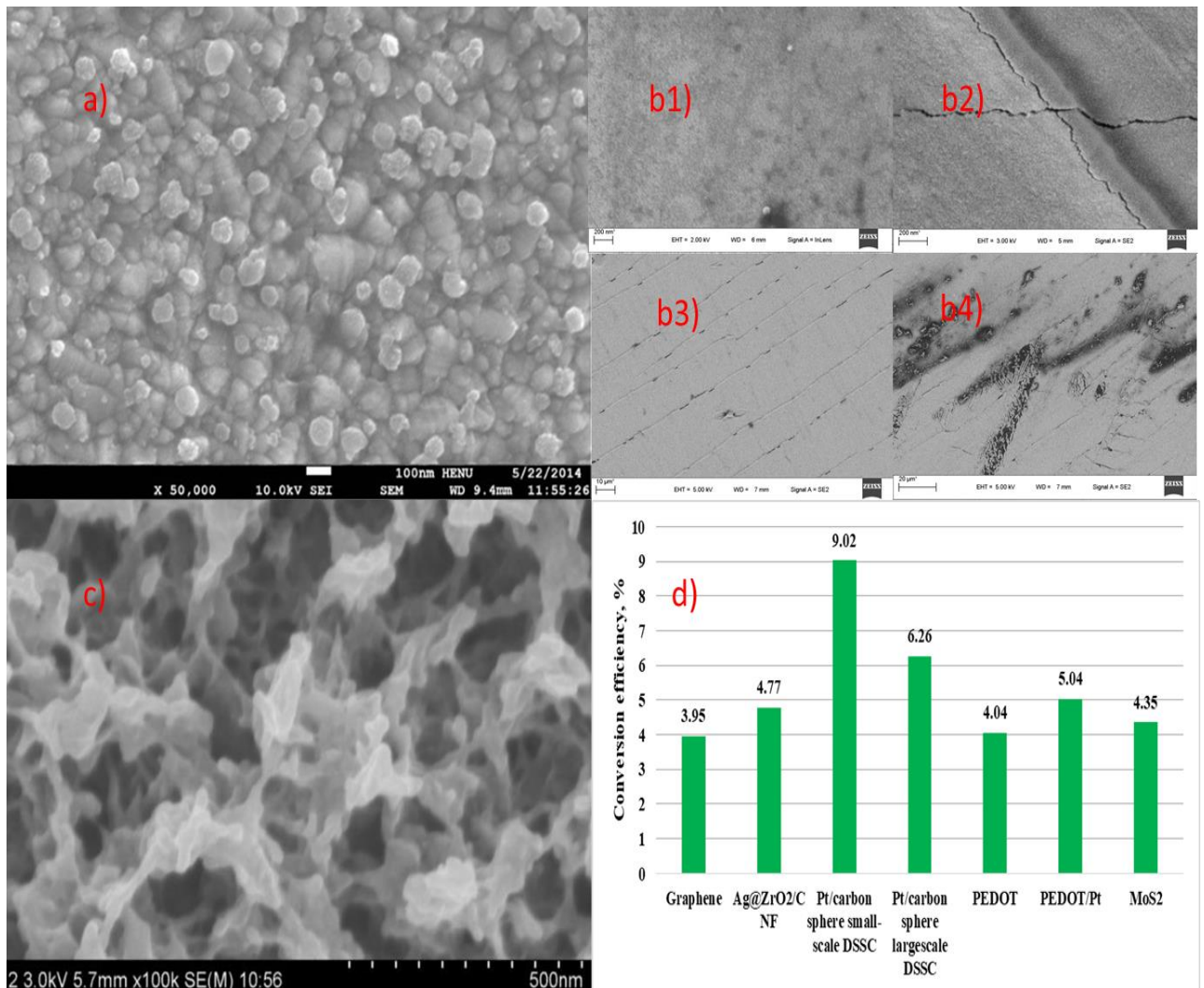


Figure 9 a) SEM image of NiS/Pt/Ti counter electrode with bump-like surface caused by Pt nanoparticles [142] b1) SEM image of Pt film deposited on ITO-PET before bending cycles [147] b2-b4) SEM image of cracked Pt film after 100 bending cycles at different magnifications [147] c) SEM image of PEDOT film on ITO-PEN substrates with typical porous network [153] d) Chart comparing the conversion efficiency of several flexible DSSC with different counter electrodes

From literature, it can be seen that platinum is still a very good option as counter electrode catalyst due to the high performance and conversion efficiency produced despite the high cost of Pt. However, this cost can be minimised by forming Pt composite counter electrodes to reduce the dosage of Pt required, while still maintaining good efficiency. Other simpler and lower cost fabrication methods such as doctor blade, screen printing and spray coating could also be used to deposit the catalyst onto the counter electrode substrate. Using other lower cost materials such as carbon is another

approach to tackle the costing issue of using Pt. However, the inferior PCE produced by these other materials made it difficult to be comparable with Pt. The use of significantly lower cost counter electrode materials would hopefully be enough to compensate, allowing for more research to be conducted using such materials. Developing hybrid materials for counter electrode or other DSSC component seems to generally enhance the performance due to the synergetic effects which should be taken into consideration for future development [4].

4.4 Flexible DSSC modules

The structure of DSSC module is almost similar to typical DSSC except the inclusion of grid lines for making up the ohmic losses from the large electron traveling area. A typical DSSC module consists of components such as rigid/flexible substrate, photoanode, counter electrode, sensitizer, electrolyte and grid lines as shown in Figure 10.

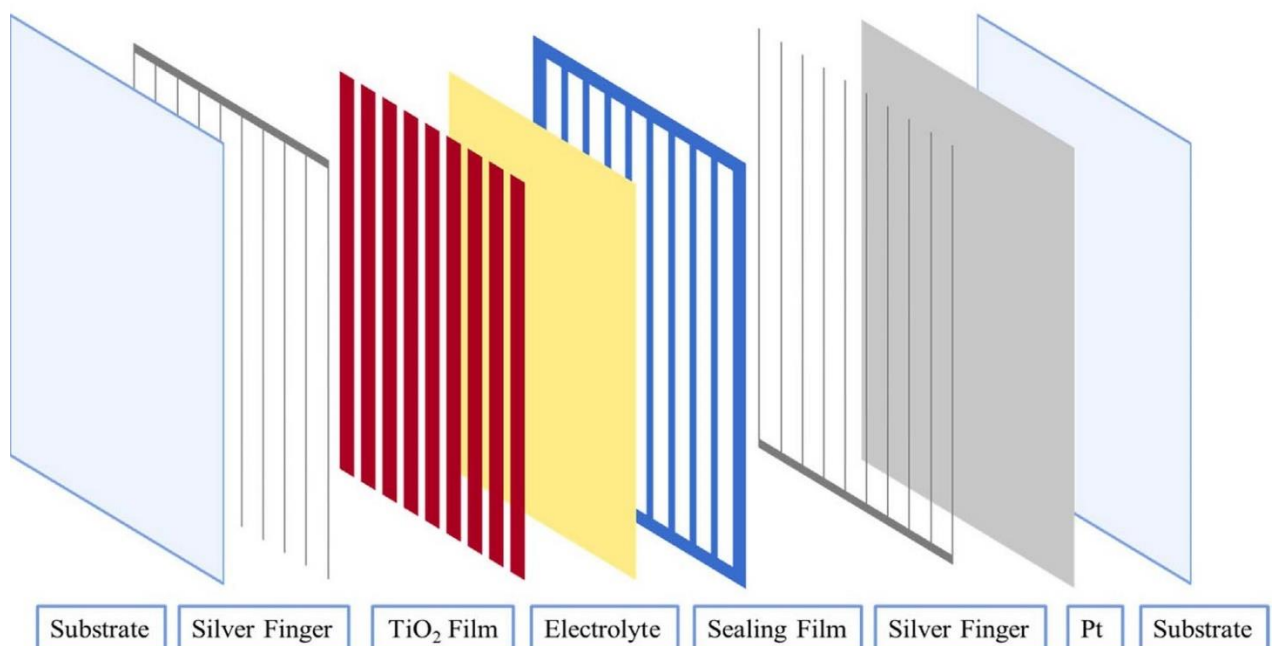


Figure 10 Schematic diagram of a typical flexible DSSC module[155]

Further development for DSSC is to upscale the device from cell to module level for both rigid and flexible DSSC. This however, have remained a challenge as there are issues in upscaling the dimensions of the modules while still keeping a good PCE. Multiple efforts have been done to address the issue either by replacing the materials or using flexible metal foils and plastic as substrates for the modules. Noda

et al. [156] managed to develop a rigid DSSC module based on FTO glass with a large area of 120 mm² and 255 mm², while having high temperature durability. The module was able to maintain 95% of the initial conversion efficiency even after 1000 hours of test due to the protective material of the collecting grid, good sealant and optimised Ru-based dye. Meanwhile, Kwak et al. [157] was able to fabricate a rigid DSSC module consisting of 22-unit cells with dimensions of 30 x 30 cm² and conversion efficiency of 2.33%. The development of these rigid DSSC modules has also encouraged the development for flexible DSSC modules.

Jen et al. [158] was able to develop an upscaled flexible DSSC using TiO₂ nanotube arrays via a parallel module with three single cells. A total active area of 5.4 cm² was fabricated with conversion efficiency of 4.77%, even higher than the single and parallel module of their rigid counter-part. Having a thinner layer of electrolyte was found to have helped the flexible module to have better catalytic activity and electrolyte diffusion. Wu et al. [155] took another approach by developing a facile binder-free TiO₂ paste called PT paste that managed to have strong adhesion with the ITO-PEN plastic substrate at room temperature. By undergoing cold isostatic press, the nanoparticle connection in the photoanode improved. The approach has also allowed them to upscale the flexible module to dimensions of 100 mm x 100 mm with conversion efficiency of 3.27% as shown in Figure 11. The research also demonstrated the flexible DSSC module's ability to charge mobile under indoor light, showing more promises for this module. Bittner et al. [159] was able to scale up a flexible DSSC module by electrodepositing ZnO/eosin Y (EY) hybrid films on ITO-PET plastic substrate. A fully functional flexible DSSC module with an area of 34.56 cm² was developed with a PCE of 2.58% as shown in Figure 11. The increase in the dimensions of flexible DSSC showed that flexible DSSC module can be used for large scale application in the future.

Table 12 Summary of non-flexible and flexible DSSC modules in literature

Photoanode	Counter electrode	Active area (cm ²)	Annealing/sintering temperature (°C)	Conversion efficiency (%)	Reference
<i>Non-flexible DSSC</i>					
TiO ₂ on FTO glass	Pt on Ti plate	12.00 25.50	500	5.80 5.20	[156]
TiO ₂ on transparent conductive oxide glass	Pt on transparent conductive oxide glass	900.00	n/a	2.33	[157]
<i>Flexible DSSC</i>					
TiO ₂ nanotubes on Ti foil	Pt on ITO-PEN	5.40	350	4.70	[158]
ZnO/EY on ITO-PET	Pt on ITO-PET	34.56	100	2.58	[159]
Facile binder-free TiO ₂ paste on ITO-PEN	Pt on ITO-PEN	100.00	120	3.27	[155]

The mechanical stability of a flexible DSSC is another important attribute that sets the module apart from a rigid DSSC module. Bending tests are usually conducted on flexible DSSC and their PCE would be estimated afterwards to determine whether they have good mechanical stability. For example, Xu et al. [160] managed to develop a highly flexible DSSC by using copper sulphide (CuS) transparent conductive film that was fabricated by colloidal crackle pattern as counter electrode. The flexible DSSC showed impressive flexibility as they managed to retain more than 90% PCE even after 500 bending cycles as shown in Figure 11. Baiju et al. [85] reported that sintering the TiO₂ nanoparticles via a plate fin heat sink at 250°C on ITO-PET plastic, did not cause a loss of integrity in the plastic substrate and also produce flexible DSSC with good mechanical stability. The PCE only showed a drop of less than 9% after 30 bending cycles. Thus, highlighting the potential of plastic substrate to withstand higher temperature under the right condition while still maintaining good flexibility. Fu et al. [104] meanwhile, managed to construct a flexible DSSC that was able to maintain over 90% of the initial conversion efficiency after 100 concave and convex bending as shown in Figure 11. A device with excellent mechanical stability was achieved by applying TiO₂ nanotube array prepared by combining hot-water-soaking and TiCl₄ post treatment. Besides bending test, the mechanical stability of the flexible DSSC can also be observed by long term measurement of the device as reported by Chou et al. [161]. The study reported that the PCE of the flexible DSSC only suffered less than 10% loss after 20 days of operation as part of internet of things (IOT) system. Thus, affirming a good mechanical stability and reliability between the Ag nanowires-TiO₂ photoanode with the ITO-PET plastic substrate. The mechanical stability and flexibility of a flexible DSSC module remains one the important factors in upscaling the device to have a longer lasting solar device for commercialisation purposes.

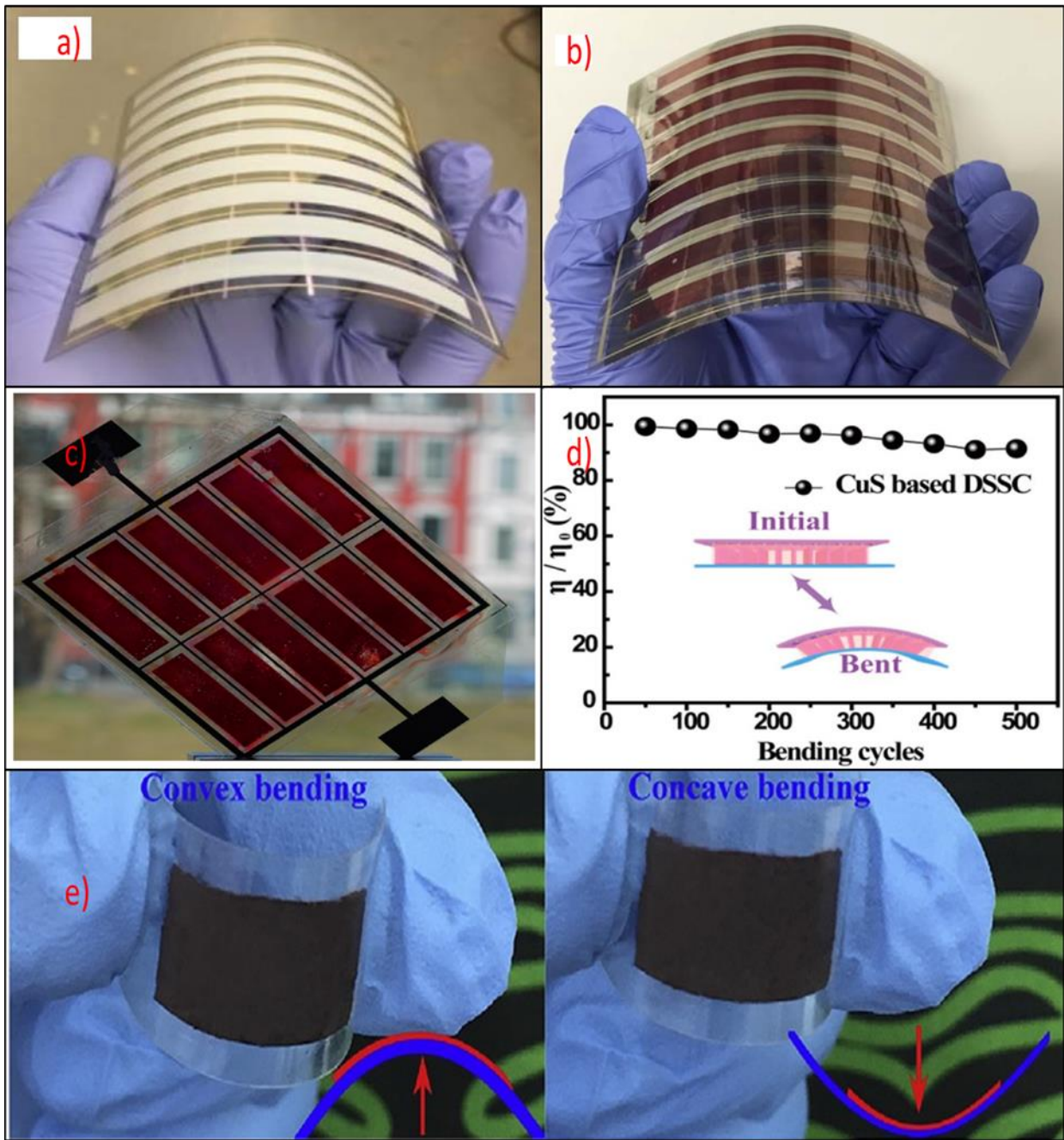


Figure 11 a) & b) Photograph of flexible TiO₂ photoanode & photograph of flexible DSSC module with dimensions of 100 mm x 100 mm [155] c) Photograph of flexible DSSC module with active area of 34.56 cm²[159] d) Stability measurements of flexible DSSC module after bending tests [160] e) Picture of TiO₂ photoanode under convex and concave bending [104]

Apart from the factor mentioned earlier, other factor that can affect the performance of flexible low temperature module is the sealing of the module. The sealing of the solar device is important for the long-term stability of the cells as poor sealing would cause liquid electrolyte to leak and cause poor performance [162]. A common sealing method for flexible DSSC is the use of thermoplastic sealing [27,59,103] on the side of a typical sandwich structure. Luo et al. [77] reported that the

thickness of the Teflon layer, the sealant used in the study, played a part in affecting the efficiency of flexible DSSC. As the thickness of Teflon decreases, the short circuit density was found to increase due to the longer **optical path**. Hence, proper sealing materials and thickness need to be chosen when constructing flexible DSSC.

The advancements on the flexible and low temperature DSSC modules remain an important topic in this field of research particularly regarding the photoanode and substrate. The studies on developing large-scale flexible DSSC modules should be the forefront in advancing flexible DSSC technology to enable them to be deployed on curved space that cover a wider area. Adding binders and performing post-treatment process have shown good results in improving both the efficiency and stability of flexible DSSC modules, but other approach including improving the fabrication techniques such as screen printing can also be taken. With regards to the DSSC sealing, the use of non-liquid electrolyte such as solid and quasi-solid electrolyte would be able to solve leakage issues especially for flexible DSSC. Besides that, another way to improve the DSSC module is by constructing a bifacial structure where both front and back of the module could be illuminated to produce high light absorption and conversion efficiency. The mechanical and flexibility tests need to be conducted for every DSSC module developed as they showed whether the device developed is suitable for long term use and commercialisation purposes.

5. Potential and challenges

Flexible DSSC has shown a lot of benefits as solar devices, but there are still some limitations to the device. Plastic-based flexible DSSC for example, has poor thermal stability that limits the device from undergoing high temperature sintering process to enhance the inter-particle connection of the photoanode and adhesion between the photoanode and plastic substrate. Thus, causing a lower conversion efficiency of the plastic based flexible DSSC when compared to rigid high temperature DSSC. Another challenge is leakage of liquid electrolyte for both flexible and non-flexible DSSC, causing an unstable device over a long period of time with decreasing performance after every operation. The use of metal substrates for flexible DSSC is a good approach, but they suffer from corrosion when in contact with some electrolytes, lowering their overall performance. Some of the main challenges and ways to improve flexible DSSC are listed in Table 13.

Table 13 Challenges and ways to improve flexible DSSC

Challenges	Ways to improve
Poor thermal stability of plastic substrate	<ul style="list-style-type: none"> - Deposition and fabrication of photoanode and counter electrode materials on plastic substrate is done at low temperature - Use flexible metal substrates
Low conversion efficiency compared to rigid DSSC	<ul style="list-style-type: none"> - Addition of binder such as TTIP and sintering aids to the photoanode, doping of photoanode nanoparticles and post treatment of the electrode such as hot press method - Application of different plastic substrates such as PEI for low sintering temperature or metal substrates such as stainless-steel mesh that can withstand high sintering temperature
Leakage of liquid electrolyte	<ul style="list-style-type: none"> - Application of non-liquid electrolyte such as solid or quasi-solid electrolyte in the flexible DSSC
Corrosion of flexible metal substrates	<ul style="list-style-type: none"> - Use different liquid electrolytes such as cobalt complex electrolyte or use non-liquid electrolyte

Despite all the challenges faced, flexible DSSC are still a growing industry and are still showing potential growth in solar technology. The flexibility of the device means that they can be applied to cover bendable and non-planar surfaces, common in electronic devices these days [24]. Thus, allowing them to be integrated with portable electronic devices, energy-harvesting and energy-storage devices. Some of their applications include integrating them into textiles to produce wearable smart clothes as demonstrated by Wen et al. [163]. The study developed a prototype of hybridised self-charging textile system that can harvest energy from solar energy to fabricate smart

clothes and operate wearable electronic devices. The use of flexible DSSC has also made the system inexpensive and easy to fabricate.

The flexibility of DSSC has also opened up their potential to be integrated with other solar devices such as photoelectrosynthetic cell, lithium-ion battery, nanogenerator and supercapacitor to form a unit of integrated systems [164]. One example was reported by Scalia et al. [165], where they formed a flexible energy harvesting and storage system by combining DSSC with an electrical double layer supercapacitor. The device has promising prospects for industrial scale-up due to their ease of fabrication and cost-effectiveness. Besides, flexible DSSC have also found their place in small niche applications for indoor use such as internet of things (IOT) and sensor applications due to their high efficiency under low light intensity, colourful aesthetics and longer life-times while also suitable for portable charging units, buildings and outdoor advertising [22,33,162].

The global market growth of DSSC is projected to grow at a rate of 12.4% from 2020 to 2027, with flexible DSSC as one of the forefronts based on their application as portable charging application accounted for a large share of DSSC market with a value of 33.3% in 2019 and is expected to keep increasing until the year 2027 [166]. The global plastic solar cell market is also projected to reach USD 386.34 million by the year 2026 with flexible DSSC part of them [167]. Besides, continuous development by some prominent players in DSSC technology such as 3GSolar, Fujikura, Solaronix SA, Ricoh, Dyesol Limited, Exeger Operations AB and Sharp Corporation has also encouraged the growth of DSSC in the photovoltaic market. For example, Swedish DSSC company, Exeger has recently raised USD 19 million to support the manufacturing of their flexible solar cell technology, meanwhile RICOH has launched new DSSC modules that improved the power output by 20% from their previous product [168,169]. Hence, with the wide and ever-growing application of flexible DSSC devices, projected increase in DSSC and flexible solar cell market, as well as continuous support by the industry prominent players, there are still a bright future and potential for this solar technology to continue to develop.

6. Conclusions and Recommendations

6.1. Conclusions

Flexible DSSC has shown to be a good alternative to the usual rigid DSSC due to their low cost of production and flexibility, allowing them to be applied in mobile and portable electronic devices. In this review paper, several comparisons can be seen in recent advancements for both rigid and flexible DSSC that still shows the superiority of rigid DSSC with regards to their performance and efficiency. However, the review paper has also managed to look into some methods and materials used in literature to improve the performance of flexible DSSC at low temperature. Based on the previous discussion, some of the important conclusions drawn from the review are given as below;:

1. Some materials such as TiO_2 photoanode and Pt counter electrode are still commonly used for both rigid and flexible DSSC with various modifications such as adding binders, doping process and post-treatments. The use of these materials is still effective as plenty of DSSC based on such materials produced good conversion efficiency compared to when using alternative materials despite their drawbacks. A high conversion efficiency of 8.10% was produced when doped TiO_2 and Pt was used as photoanode and counter electrode respectively. However, highest conversion efficiency of 10.28% was reported for Pt-free fibre shaped flexible DSSC with TiO_2 as photoanode.
2. Metal substrates could be a good alternative to plastic substrates for flexible DSSC application due to their flexibility, thermal stability and good performance. Preparing multiple morphologies of photoanode nanostructure onto the metal substrate could help to improve the performance of the flexible DSSC. High PCE of 8.10% for flexible DSSC was yielded when Ti mesh substrate was used indicating their superiority as flexible substrate.
3. The up-scaling of flexible DSSC module is still a challenge due to the low PCE at large area. A flexible DSSC module with an area of 100 cm^2 with conversion efficiency of 3.27% managed to be developed and multiple efforts still needed to be made to increase the size and efficiency of the device in the future.
4. Physical deposition methods such as doctor blade technique is still a favourable fabrication technique for flexible low temperature DSSC due to the simplicity and relatively good performance of the fabricated device.

5. Sintering temperature of around 100-120°C has been found as the optimal temperature for flexible plastic-based DSSC as they managed to produce relatively high conversion efficiency of around 7-8% while still able to prevent the plastic from deforming.

6.2. Recommendations

Flexible low temperature DSSC could still use with more improvement to further enhance their desirability in solar cell market in the future. The main issue of low conversion efficiency of the solar module still needed to be addressed and some suggestions has been made to be considered in future works:

1. The incorporation of binders, sintering agent or additives are an attractive approach to enhance the performance of photoanode materials. More research needs to be conducted to find suitable materials to be incorporated to the semiconducting metal oxides materials as they have proven to be an effective measure in enhancing the efficiency with simple process.
2. The application of non-liquid electrolyte such as quasi-solid and solid electrolyte need to be further investigated for flexible DSSC in order to prevent the leakage issue faced when using liquid electrolyte. Solving this issue would help improve the stability and lifespan of the flexible DSSC, making them a more attractive prospect in solar cell technology. Hence, developing a quasi-solid electrolyte with good flow under certain condition could be one of the steps taken in the future.
3. There has to be more research done to provide an alternative to PEN and PET plastic substrate such as the PEI plastic substrate. Developing different type of lightweight polymer substrate with possibly good thermal stability would be an ideal way forward. Making use of materials that are renewable and industrially compostable as well as making the substrate thinner are just some approaches that can be taken to minimise their environmental impact.
4. The use of different flexible substrate such as metal and especially textiles should be more focused on in the future due to the potential application flexible DSSC in mobile and wearable electronics. A simple, feasible, durable and efficient approach would be to glue or knit an already functioning DSSC to the textile to form a lightweight, flexible and stretchable DSSC module.

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Declaration-of-interests

The authors declare that they have no known competing financial interests or personal relationships that could have influenced the work reported in this paper.

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Declaration of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:



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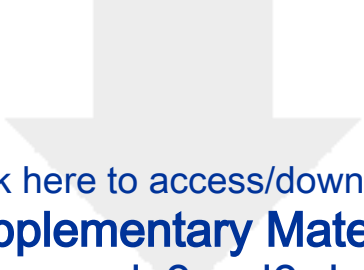
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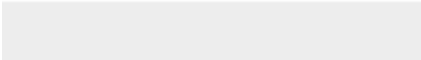

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