Engineering flexible dye-sensitized solar cells for portable electronics.

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Abstract

With growing demand for economic, eco-friendly, easy-manufacturing and renewable portable power source, flexible dye-sensitized solar cells (FDSSCs) exhibit a promising potential in the field of portable power sources. However, FDSSCs surfer from low conversion efficiency and poor stability, which hinder their applications. In this work, we have summarized recent development and challenges according to respective functions of their vital components (substrate, semiconductor film, sensitizer, electrolytes and counter electrode) as well as their effects on photoelectric conversion efficiency. Meanwhile, typical fabrication approaches have been analyzed for increasing the incident photon to current efficiency (IPCE). The approaches in rigid dye-sensitized solar cells (DSSCs) for improving performance are also introduced to provide the favorable direction for accelerating FDSSCs' application in field of potable power source.

Keywords: flexible, dye-sensitized solar cell, photoelectric conversion, portable electronic devices.

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1. Introduction

With the world's ever increasing demand for energy, fossil fuels as the major resource of electric energy, have been mined and burnt to supply the electric power, causing serious environmental problems, such as greenhouse effect, atmospheric pollution, water resource pollution and so on [1]. Meanwhile, human beings have to face with the fact that fossil fuels are limited and will be used out in recent future [2]. Given this emergency situation mentioned above, energy and environment problem are the top challenges in next 50 years [3], and developing clean, renewable and sustainable energy sources has been the indispensable strategy to continue civilization of our society. Among the renewable energies, solar energy is the most promising energy sources to change current energy consumption structure and support the low-carbon development due to its duration, abundance, pollution free and security [4, 5]. Actually, solar energy arrived in the earth's surface amount up to 3.8 million EJ/year, which can meet the annual energy demand of the world in short time [6, 7]. Therefore, developing excellent solar cells has been prioritized in the developing strategies in many countries.

Nowadays, when various mobile electronic devices bring convenience to us, short serving time is always worrisome, so the demand for portable power source increases sharply [8, 9].

Among current portable powers, the flexible film solar cells (FFSCs) are a promising type power source due to their excellent portability, sustainable output, autogenic transformation and

remarkable conversion efficiency [10-14]. The FFSCs, such as flexible amorphous silicon (α-Si, 13.6% [15]), flexible copper indium gallium selenide (CIGS, 21.7% [16]), flexible cadmium telluride (CdTe, 16.5% [17]), flexible gallium arsenide (GaAs, 26.7% [18]), flexible perovskite

solar cells (PSC, 17.3% [19]) and flexible dye-sensitized solar cells (FDSSCs), all have been

investigated widely and achieved remarkable conversion efficiency. However, highly technical

requirements for fabrication (α-Si, CIGS, CdTe, GaAs), toxicity of materials (CdTe, GaAs, PSC) and high cost limit their applications [20-24].

Among flexible devices above, the flexible dye-sensitized solar cells (FDSSCs) show enormous potential for the portable power due to easy manufacture, abundant materials, low

product cost, potential conversion efficiency (up to 32% in theory) and excellent flexibility. In order to understand the status of FDSSCs research and development, Figure 1 presents the publications survey between 2008-2017 for DSSCs and FDSSCs.

Figure 1 reveals that thousands of publications about dye-sensitized solar cells (DSSCs) have been reported each year in past decade, but the publications with titles including "flexible" or "plastic dye-sensitized solar cells" is 2-5% of the publications. The number of annual publications increases from 2008 to 2014, and start to decrease after 2014. This phenomenon is likely induced by the following factors:

DSSCs possess enormous potential to be commercialized with cost-efficiency, which attracts a mass of researcher's attention. So a series of attempts had been made to exploit its potential before 2014. However, there is no radical improvement achieved in conversion efficiency, which weakens researchers' enthusiasm to DSSCs. Meanwhile, an important progress had been achieved in different type solar cells, such as CdTe solar cells, CIGS solar cells and perovskite solar cells. Thus, the number of annual publications reached to the highest in 2014. After that, this number declines gradually. Comparing to the effort on DSSCs research and development, the input amount for FDSSCs is rather small due to its relatively low conversion efficiency, and insufficient cognition on the potential in portable power source [25]. This phenomenon hints that researches involving in FDSSCs are hysteretic by contrasting with promising prospect.

So far, a series of outstanding reviews have been reported toward DSSCs [26-29]. Among these reviews, various advanced technologies and basic theories have been discussed in detail, but most of them are focused on one of five major factors (namely substrate, semiconductor film, sensitizers, electrolyte and counter electrode). These reviews ignored the major characteristics of FDSSCs because of the difference in substrates. Although Weerasinghe *et al*. presented a review on FDSSCs, their focus was on the fabrication processes on plastic-substrates [30].

Moreover, FDSSCs has no breakthrough in practical conversion efficiency compared with

high theoretic conversion efficiency, and surfer from difficult sealing and poor mechanical stability. Therefore, optimizing the properties of substrate, semiconductor film, sensitizers, electrolyte and counter electrode is an alternative route to accelerate FDSSCs' applications [31]. In this review these factors are comprehensively discussed to facilitate all-around understanding its mechanism, improving its conversion efficiency and lifetime. Particularly, the achievements verified in rigid DSSCs are also introduced for facilitating FDSSCs' evolution.

2. Definition of FDSSCs and rigid DSSCs

2.1. Inheritance and distinction

Deriving from rigid DSSCs, many elements of FDSSCs are able to inherit directly from rigid DSSCs, such as sensitizers, materials for counter electrode (CE), redox couples, sandwich structure etc. However, its requirement for flexibility resulted in some differences from rigid DSSCs in terms of substrate, fabricating methods of the photoanode, post-treatment, the highest IPCE.

Table 1. The brief contrast between rigid and flexible DSSCs

Types	Substrate	Past-annealed	Semiconductors	Highest IPCE
Rigid DSSCs	ITO[a]/FTO[b] glass	Yes	TiO_2, ZnO, SnO_2, Nb_2O_5, WO_3, $SrTiO_3$, In_2O_3, Zn_2SnO_4 etc.	14.3% [32]
Flexible DSSCs Flexible	Metal mesh/foil/wire	Yes	TiO_2, ZnO, SnO_2, Zn_2SnO_4.	9.1% [33]
DSSCs	TPCO[c] film	No	TiO_2, ZnO.	7.6% [34]

[a]ITO: indium tin oxide, [b]FTO: fluorine doped tin oxide, [c]TPCO: transparent conductive oxide on polymer substrate.

As shown in the Table 1, the fluorine doped tin oxide (FTO) glass and indium tin oxide (ITO) glass are usually employed as substrates in rigid DSSCs, due to their excellent transmittance, conductivity, acid resistance, alkali resistance and high-temperature resistance. Therefore, the inorganic photoanode can be synthesized on conductive glass substrate at high-temperature environment as well as post-annealed at high temperature to improve charge transfer ratio [35].

The FDSSCs normally employ transparent conductive plastic film (denoted as TPCO, indium tin oxide/polyethylene naphthalate—ITO/PEN, the fluorine doped tin oxide/polyethylene terephthalate—FTO/PET), or flexible metal material (such as titanium foil, titanium wires, titanium or stainless steel wire mesh etc.) as substrates [36, 37]. The photoanode with TPCO substrate is unable to be annealed at high temperature (>450℃) for removing organic residuals in nanostructured film and forming chemical bonding between conductive layer and semiconductor film [30], which leads to low charge transport and charge collection. On the contrary, the flexible photoanode based on metallic foil substrates can be treated with annealing at high temperature to

improve charge transfer and collection.

In preparation of FDSSCs, plentiful metal oxides have been investigated as the photoanode material in the rigid DSSCs, such as TiO_2, ZnO, SnO_2, Nb_2O_5, WO_3, $SrTiO_3$, In_2O_3, and Zn_2SnO_4 [38-42]. However, the major photoanode materials applied in the FDSSCs are limited on TiO_2, ZnO, SnO_2, Zn_2SnO_4 [43-46], plus a few reports on SnO_2 and Zn_2SnO_4.

It can be found from Table 1 that the highest IPCE of rigid DSSCs is nearly twice over that of TPCO-substrate FDSSCs, and is as nearly 1.5 times as that of metal-substrate FDSSCs. The conversion efficiency of the rigid DSSCs is considerably higher than that of the FDSSCs, which result from three reasons, as follows:

- Conductive glass possesses better transmittance and conductivity than that of conductive plastic film, which makes rigid DSSCs own better light-absorbing capacity and electron-collecting ability.
- Plastic substrate limits extreme methods to fabricate and improve photoanode for high performance, such as the preparation of semiconductor film on strongly alkaline as well as high-temperature synthesis or post-annealing etc.
- In addition, some improving strategies applied in rigid DSSCs weren't employed in FDSSCs, such as the utilization of high efficiency dyes (i.e. SM371 and SM315 [47]), some low-corrosion electrolyte (i.e. cobalt-base redox couple, copper-based redox couple etc.).

Although rigid DSSCs possess high IPCE compared with FDSSCs, rigid DSSCs shown poor portability since conductive glass substrate has heavy weight, low removability, and inferior ability to adapt complicated space or irregular surface. Instead, the flexible substrate offers remarkable removability and good adaptability to complicated space or irregular surface, which makes the FDSSCs more competitive in portable power source. In addition, flexible substrates enable FDSSCs to be produced in large scale via roll to roll for reducing product cost [48]. Thus, large-scale producing methods were also investigated, such as ultrasonic spray-coating technology [49], large area atmospheric-pressure plasmas process [50] and so on.

2.2. Challenges

Conversion efficiency and stability are two indexes representing the property of flexible and rigid DSSCs. Improving conversion efficiency is the first priority for commercial application of

flexible and rigid DSSCs. It depends on reducing the recombination reactions, increasing electron collection efficiency, enhancing light-absorbing capacity and catalytic performance of CE,

strengthening ionic conductivity of electrolyte [31]. The device's stability is an important issue, in particular the device with plastic substrates. In the preparation of plastic substrate-based FDSSCs, the photoanodes are generally fabricated via binder-free methods or without annealing treatment at high temperature. The binder-free semiconductor layer results in the formation of crack in drying process, which increases the risk of the connection between electrolyte and conductive layer. The absence of annealing treatment makes organic residuals remain in semiconductor layer, which weakens electron transport. Obviously, organic residuals and crack both can reduce conversion efficiency and lifetime of the FDSSCs.

In addition, the FDSSCs suffer from internal stretch or compression of semiconductor film under outside force, which accelerates the degeneration of the photoanode due to the interconnection among the nanostructures and between the semiconductor film and conductive layer [51]. The schematic of degeneration can be illustrated by Figure 2(a). Figure 2(b-c) shows clearly microscopic morphology of metal oxide film before and after bending under outside force [52]. The cracks of metal oxide film make more challenges in designing excellent and stable

commercial FDSSCs.

3. Structure and operational principle

Figure 3(a) shows a structure of typical FDSSCs. It consists of the photoanode which composes of semiconductor layer sensitized by dye on flexible conductive substrate, the counter electrode that is made by coating thin catalyst layer on a transparent conductive substrate, as well as the electrolyte frequently contained with iodine-based redox couple(I^-/I_3^-). In the photoanode, the semiconductor film is often made up of single nanostructured or multiple nanostructured materials, such as TiO_2 nanotube array (NTAs), TiO_2 nanowires array (NWAs), TiO_2 nanosheets (NSs), ZnO nanoparticles (NPs), TiO_2 mesoporous film, TiO_2 NPs/ZnO NWAs, ZnO NPs/TiO_2 NTAs and so on [53-56]. The semiconductor film must absorb dye to form a valid photoanode, which presents substrate/semiconductor/dye structure.

Its specific light-to-electricity principle has been demonstrated in the Figure 3(b). When the photons from sunlight inject into the photoanode, dye molecules on the surface of semiconductor assimilate their energy and turn into excited state from base state (turn into Lowest Unoccupied

Molecular Orbital (LUMO) from Highest Occupied Molecular Orbital (HOMO)), then the photo-excited electrons from those dyes at excited state inject into the semiconductor. Meanwhile, dye molecules turn to oxidized state (D^+) due to loss of electron, and then the oxidized dye is

reduced by iodides (I^-) and iodides are oxidized to tri-iodides (I^-_3) which break the balance that the I^-/I^-_3 redox couple keeps a relatively stable state in the electrolyte. In the period,

photo-excited electrons in the semiconductor diffuse to the conductive substrate under the affection rooted in the gap of electron concentration. Next, the photo-excited electron transfers to the counter electrode via the external circuit linked with load and solar cells. In the process, tri-iodides (I^-_3) diffuse to the CE, accepting the electron from the outside circuit, and reduced to iodides with the assist of a catalyst. The procedure is a whole working circle, which is considered as the next Eqs (1-4).

$$2\text{Dye} + \text{hv} \rightarrow 2\text{Dye}^+ + 2e^- \quad (1)$$

$$3I^- \rightarrow I^-_3 + 2e^- \quad (2)$$

$$2\text{Dye}^+ + 2e^- \rightarrow 2\text{Dye} \quad (3)$$

$$I^-_3 + 2e^- \rightarrow 3I^- \quad (4)$$

If there is nothing to be lost in the ideal photo-to-electricity process, DSSCs has long lifetime and remarkable conversion efficiency. But in the practical process, the photo-excited electrons are lost via the recombination that the electrons injected into the semiconductor fail to transfer to external circuit but combine with oxidized dyes or tri-iodides, which reduces the practical current of short circuit. Therefore, inhibiting recombination among photo-excited electrons, tri-iodides and oxidized dyes have been investigated widely to improve conversion efficiency.

4. Major indexes and theoretical calculation

In order to evaluate the property of FDSSCs, four major indexes including voltage of open circuit (V), current intensity of short circuit (J), fill factor (FF) and conversion efficiency (η) are introduced. In FDSSCs, V is defined as the energy difference between the redox potential of redox couple and the quasi-Fermi level of the electron in the semiconductor conduction band. It can be calculated by:

$$V = \frac{F,N}{} - {}_x = \frac{\pm \Delta}{} + \frac{T}{} \ln_N(\quad) - {}^x \quad (5)$$

where, kT is the thermal energy, q is the elementary charge, n corresponds to the electrons

number in the conduction band (CB), N represents the total number of electronic states in the CB, $_x$ refers to the redox potential of the electrolyte, Δ denotes the difference between the conduction band edge of semiconductor before and after dye adsorption [57, 58].

In FDSSCs, J is related to light absorption ability generally, and can be given by:

$$J = \int_\lambda LH() \cdot \cdot d \quad (6)$$

$$LH( ) = 1 - 10^{-\varepsilon(\lambda)\cdot\cdot} \quad (7)$$

where λ is the wavelength of incident light, LHE(λ) refers to the light harvesting efficiency at given wavelength (λ), represents the electron injection efficiency into semiconductor

from dye molecules, and means the charge collection efficiency of photoanode, which can be decreased by recombination. $\varepsilon()$ is the molar absorption coefficient, b and c refer to the thickness of semiconductor film and concentration of dye [58].

Fill factor () is determined by series resistance and shunt resistance mojorly. Namely, FF will increase with the increase of series resistance and the decrease of shunt resistance. It can be

$$= \frac{P_{ax}}{V\cdot J} = \frac{V\cdot J}{V\cdot J} \quad (8)$$

where V and J represents the maximum of voltage and current density responding to max output power (P_x) respectively [59, 60].

Coversion efficiency () is a crucial parameter for its evaluation, and can be determined by:

$$= \frac{P}{P_i} = \frac{V\cdot J\cdot}{P_i} \quad (9)$$

where P is power density of incident light. It is often 100mW/cm^{-2} under common measure standard (AM 1.5G) [61].

5. Factors affecting the performance of FDSSCs

5.1. Substrates

Substrate's functions are mainly in supporting semiconductor film and collecting photo-excited electrons (working as current collector). It can be separated into plastic and metal substrates. Different substrates lead to diverse fabricating process and post treatments.

5.1.1. Plastic substrates

In the preparation of flexible photoanode or CE, the plastic conductive substrate was coated with metal oxides film or catalyst layer. After assembly of FDCCSs, sunlight must go through

plastic to reach at dye layer and be absorbed. Therefore, the performance of FDSSCs relies on the property of the plastic conductive film.

The transparent polymer film with a conductive oxides layer (TPCO), such as ITO/PEN, FTO/PEN, ITO/PET and FTO/PET, are often chosen as the substrate in FDSSCs because of their good conductivity, outstanding transmittance and excellent flexibility. IPCE of 3.27% has been achieved by the FDSSCs (its appearance and schematic shown in Figure 4(a-b)) with the photoanode comprised TiO_2 film of 100 mm x 100 mm size prepared on ITO/PEN by doctor blade method [62]. 4%, 4.2% IPCE have been obtained for the FDSSCs fabricated via ultrasonic spray-coating technology, atmospheric pressure plasmas process respectively [49, 50]. The TPCO-based FDSSCs not only provides convenience to user, but also is manufactured by roll-to-roll mass production. So TPCO is expected to replace ITO/FTO glass for decreasing product cost [63].

However, current commercial TPCO all suffer from low heat-treatment temperature (PET: 120 °C, PEN: 160 °C). This shortage makes the photoanode with TPCO substrate unable to be annealed at high temperature, when the semiconductor film must endure calcination at high temperature of up to 450°C to remove the organic additives and improve charge transport [64]. Consequently, a low IPCE was exhibited because low charge transport enhances recombination reactions and weakens charge collection efficiency. Therefore, it is a promising direction for enhancing property of plastic FDSSCs to prepare semiconductor film on plastic substrate at low temperature [65, 66].

5.1.2. Metal substrates

Metallic foils, meshes and wires, such as Ti wire, Ti mesh, stainless steel mesh, Ti foil and the like, were deemed to be a good choice as substrate of FDSSCs [36], due to their reliable high-temperature resistance, flexibility and lower resistance than that of plastic conductive film. So the photoanode with metallic substrate is able to be sintered at high temperature for improving interconnection and charge transport [67, 68].

Among metallic substrates available, Ti exhibits excellent potential. Liang *et al.* designed a flexible fiber-type DSSC with Ti-wire photoanode and then obtained IPCE of 6.8% [69], Liu *et al.*

prepared the FDSSC with Ti mesh-supported photoanode which is co-doped by Er^{3+} and Yb^{3+} and attained 8.1% IPCE [70]. Figure 4(c) shows the FDSSCs (its schematic exhibits in Figure 4(d))

with six working electrodes and one CE. The FDSSCs achieved an IPCE of 9.1% and exhibited excellent flexibility (bending angle of near 180°) [33].

However, high price of titanium limits its wide application. Therefore, stainless steel was introduced in FDSSCs to replace Ti, due to its remarkable resistance to corrosion and low price. Cho *et al*. designed the FDSSCs used stainless steel mesh as substrate and obtained IPCE of 3.11% [71]. Unfortunately, the performance of stainless steel-based FDSSCs fails to reach to the same level with Ti-based FDSSCs. However, the conversion efficiency can be improved by modifying its structural design.

Comparing the metallic substrate FDSSCs with plastic FDSSCs, it can be found that metallic substrate possess enormous potential as flexible substrates [72]. However, metallic substrates all suffered from certain shortages, for example, metal foil has no transmittance, metal mesh owns low effective area for accepting sunlight, metal wire is hard to support the manufacture of large scale FDSSCs. In contrast, metal foil and mesh are more promising to be applied in commercial FDSSCs in large scale.

An excellent transparent CE is required to ensure adequate sunlight to reach metallic foil photoanode (via back illumination). Duan *et al*. prepared transparent Co-Se alloy CE for Ti foil FDSSCs and generated IPCE of 7.52% [73]. In addition, for mesh-substrate FDSSCs, introducing a reflecting film under transparent CE maybe is a valid way to boost efficiency. The principle how the reflecting film enhances the efficiency of mesh-substrate FDSSCs is illustrated in Figure 5. The mesh-substrate FDSSCs with a reflecting layer can utilize reflected light (② formed by reflecting back the sunlight (① lost from mesh pore.

5.2. Semiconductor films

Semiconductor film is an inevitable pathway to transport electrons in DSSCs, so its physical property and structure determine the performance of the device [56]. Among plentiful metal oxides, TiO_2 and ZnO have been employed widely in FDSSCs.

TiO_2 with a wide band gap (~3.2eV) is the most effective material for photoanode, resulting from its high cost efficiency, good stability, excellent dye-adsorbing capability, high availability and compatibly optical and electronic properties [74-76]. Though ZnO was regarded as a remarkable alternative for TiO_2 because of similar band gap energy (~3.3eV), higher electron

mobility, large excitation binding energy (60eV) against photocorrosion, availability at low cost and more morphologies than that of TiO_2 [55], the actual performance shown by ZnO-based FDSSCs failed to meet researchers' initial expectation.

According to published reports, the highest IPCE shown by ZnO-based FDSSCs is 5.24% [77], which is only 58% of the highest IPCE from TiO_2-based FDSSCs. This difference is likely

resulted from zinc oxide's poor resistance to corrosion from acidic electrolyte and acidic dye molecules (such as N3, N719, Black dye etc.), and insulating layer formed by aggregation of Zn^{2+} -contained dye molecules on the ZnO surface [78-80]. The details about the effects of semiconductor film on performance will be discussed in section 4.3.

5.2.1. Morphologies and synthesis methods

Mesoporous TiO_2 is used in FDSSCs due to their easy fabrication, good light-scattering capability and high surface area [81]. High surface area is favorable to load more dye molecules

for enhancing light absorption. In order to get higher surface area, TiO_2 particles were prepared with small diameter, but Grätzel found that the light-scattering capability was lost when the

particles' diameter is less than the wavelength of incident sunlight [82]. So double-layer mesoporous TiO_2 was designed to optimize scattering light capacity and surface area [83].

The mesoporous film can be formed via hydrothermal process, sol-gel method, doctor blade process, electrospray, hot compression, electrodeposition (ED) and electrophoretic deposition (EPD) [54]. Synthesis methods play an important role for FDSSCs performance, the FDSSCs with the mesoporous TiO_2 film fabricated through electrospray on ITO/PEN achieves an IPCE of 6.31% [84], the mesoporous TiO_2 synthesized by EPD on ITO/PET just attained 4.93% IPCE [85].

In the fabrication process, organic binder was often used, which brought organic residuals in mesoporous TiO_2 and led to poor interconnection among nanoparticles and high recombination ratio. In particular, sintering at high temperature cannot be used to remove organic residuals from mesoporous TiO_2 on plastic substrate. Therefore, the attempts were made to enhance interconnection of mesoporous TiO_2 without damaging plastic substrate. Laser sintering [86] and

combination of pre-sintering and isostatic pressing has been applied in FDSSCs, particularly plastic FDSSCs with the mesoporous TiO_2 (Figure 6(a)) fabricated by isostatic pressing demonstrate conversion efficiency of 7.43% after pre-sintering [87]. The preparing process

without organic binder is applied in plastic FDSSCs, such as EPD and hot compression [88-90]. Unfortunately, improving performance from hot compression wasn't kept in long term [51]. However, there is no similar problem in the FDSSCs with a metal-substrate photoanode because metal-based photoanode can be sintered at high temperature.

Comparing with mesoporous TiO_2, TiO_2 nanowires (NWs, Figure 6(b)) and nanorods (NRs, Figure 6(c)) possess high surface area and electric conductivity, which make them become popular choice for excellent FDSSCs. The TiO_2 NRs or NWs was synthesized on Ti substrate through hydrothermal method since Ti substrate can endure extreme growth conditions and act as titanium source. For examples, the TiO_2 NWAs/Yb^{3+} and Er^{3+} co-doped NPs, Nb_2O_5 coated TiO_2 NWs and Nb-doped TiO_2 NPs/TiO_2 nanowire arrays photoanode all were fabricated on Ti mesh and shown remarkable conversion efficiency [70, 91, 92]. And TiO_2 nanorod arrays (NRAs) growing on carbon fibers (CFs) were employed in tube-shaped FDSSCs, generated an IPCE of 1.28% [93]. The FDSSCs with PET supported TiO_2 NRs photoanode prepared by magnetron sputtering achieved IPCE of 5.3% and fill factor of 0.8 [94]. Obviously, nanostructures have to combine with suitable substrate and preparation method for exhibiting remarkable performance.

TiO_2 NWs and NRs arrays both possess low light scattering capability compared with mesoporous TiO_2. The modified treatments were utilized to improve light scattering capability and reduce recombination reactions, increase dye loading capacity. TiO_2 nanotube arrays (NTAs) with high surface area have been investigated in DSSCs, but it possess limited active surface area and fragile nature, which limits its application in large-scale devices [95]. On the contrary, TiO_2 nanotubes array grown on Ti foil or wire by anodization can be peeled off and transferred to other flexible substrates to form photoanode. Fu *et al*. transferred TiO_2 nanotube arrays from anodized Ti foil to on ITO/PEN as photoanode and obtained 5.41% IPCE [96]. Compared with plastic substrate-supported TiO_2 nanotube arrays, metal substrate-supported TiO_2 nanotube arrays (Figure 6(d)) shows better performance and stability. For example, Liang *et al*. prepared TiO_2 NTAs on fine Ti wire as flexible photoanode and gained an IPCE of 6.8% [69]. The surface area can be enhanced through adjusting pore size of nanotube and increasing length of nanotube, but long nanotube will lead to high risk of fragmentation.

Comparing with TiO_2, ZnO possesses richer morphologies than that of the titanium dioxides known today due to higher anisotropy [97]. Meanwhile, considering its excellent photoelectric

property, it was deemed as prior choice in photoanode materials. However, its highest conversion efficiency is 7.5% in rigid DSSCs comparing with that of TiO_2 (14.3%). [98]. Therefore, the research of nanostructured ZnO semiconductor film in FDSSCs is rather limited, comparing with their plentiful ZnO nanostructures[55].

The ZnO powders (Figure 6(e)) composed of ZnO NPs (lump-like) was chemically assembled on ITO/PET as a photoanode for FDSSCs. It shows enhanced light scattering ability, superior electron transport property and IPCE of 5.16% [99]. ZnO NPs/NRs hierarchical architecture (Figure 6(f)) was synthesized on zinc foil substrate via a two-step hydrothermal growth process and exhibited a IPCE of 2.17% [100]. Ohashi *et al*. fabricating porous ZnO films included two kinds of macroscopic morphologies (flower-like particles and densely packed nanoparticles, shown in Figure 6(g)) on ITO/PET via modification of microscopic morphologies to enhance interconnection, and achieved 4.1% IPCE [101]. Chang *et al*. prepared ZnO nanoarchitectured film (Figure 6(h)) consisting of ZnO particle and ZnO nanocactus structure on ITO/PET by facile RT-chemical bath deposition method and achieved highest IPCE of 5.24% in ZnO-based FDSSCs [77].

Deposition and mechanical compression were applied to produce porous ZnO NPs photoanode and enhance interconnection among NPs. For instance, a photoanode with porous ZnO film was prepared through EPD, followed by compression, and then it was assembled for the FDSSCs demonstrated an IPCE of 3.36% [102]. Choudhury *et al*. optimized the pressure and temperature to investigate the performance of FDSSCs based on ZnO NPs film on the ITO/PET, and find optimized compression parameter (130 MPa, 70 °C) to achieve relative 45% improvement in IPCE [103]. It seems that compression is an effective way to improve performance of ZnO film, but it was found that the compression process just elevate the short-term performance of the FDSSCs because the devices undergone compression were gradually degraded with declining connection among NPs and between ZnO film and conductive layer. Long-

term stability of FDSSCs may rely on new materials or novel fabrication and post-treatment technology.

By contrast with plastic-substrate photoanode, metal-substrate photoanode can be sintered at high temperature (450-500°C) to reinforce interconnection and electron transport. For instance, the ZnO nanosheets (Figure 6(i)) prepared on Ti foil by simple hydrothermal can be annealed in

air at 450°C for 3h and combined with PEDOT/ITO/PET CE to yield IPCE of 2.97% [104]. The FDSSCs used ZnO nanorod arrays coated stainless steel wire (Figure 6(j)) and Pt wire as photoanode, CE and showed IPCE of 0.96% [105]. Moreover, ZnO NR, NWs and NSs all were grown on stainless steel mesh to be applied in FDSSCs, but their conversion efficiencies are less than 3% (1.11~2.71%) [106-108].

Therefore, several conclusions can be reached: (1) TiO_2 FDSSCs achieved higher conversion efficiency than ZnO FDSSCs commonly; (2) except on Ti substrates, TiO_2 is hard to grow on flexible substrates (TPCO, stainless steel) directly, but ZnO can grow on plentiful substrates with various morphologies directly; (3) TiO_2 prepared on substrate via spin-coated, sol-gel, mechanical compression and deposition shown relative low performance (in mechanical stability, electron transport and collection). (4) In addition, the combination of two nanostructures (with hierarchical or mixed structure) can improve conversion efficiency by increasing surface area and enhancing light-scattering capacity.

Therefore, combining titanium oxide's advantages with zinc oxide's merits, namely preparing TiO_2 or TiO_2/ZnO mixed photoanode on ZnO template, is likely to create more morphological possibilities to develop FDSSCs. On a rigid ITO glass, Xin *et al*. prepared TiO_2 flower-like cluster by converting ZnO nanoflowers in $(NH_4)_2TiF_6$ and boric acid (H_3BO_3) solution, which make photoelectric conversion efficiency increases to 2.73% from 1.16%. This report shows that flower-like structure owns a fast electron transport time and charge collection efficiency was nearly 100% [109]. Therefore, it is feasible that fabricating TiO_2 with various morphologies on flexible substrates via using ZnO as template, then employing it as flexible photoanode to improve conversion efficiency.

5.2.2. Post treatments

In general, FDSSCs with pure and unmodified semiconductor film are likely to achieve a high performance by post-treatments. Several optimized treatments were frequently used in FDSSCs to enhance their performance, such as doping, passivation, plasma and coating treatment.

The doping treatments as an effective way to improve IPCE in DSSCs were also introduced in FDSSCs to enhance their performance. Those materials that are able to be doped in metal oxides include metallic ions, precious metals and nonmetallic elements. Among those materials,

magnesium (Mg), niobium (Nb), ytterbium (Yb) and erbium (Er) have been investigated for increasing IPCE of FDSSCs. Ti meshes supported TiO_2 NRs/Er^{3+}-Yb^{3+} co-doped TiO_2 NPs (Figure 6(k)) have been reported with a relative efficiency improvement of 86% comparing with TiO_2 NRs supported pure TiO_2 NPs [70]. Similarly, Ti mesh supported TiO_2 NWs/ Nb-doped TiO_2 NPs is also reported with a relative efficiency promotion of 45% compared with non-doped device [92]. Mg^{2+} doped TiO_2 NRs/CNT incorporated TiO_2 NRs synthesized on ITO/PET by electrospun and ultrasonic treatment obtains 29.4% improvement in IPCE by contrast with the non-doped similar device [112]. In addition, there are plentiful elements employed for doping treatment in rigid DSSCs to improve conversion efficiency, which can also be introduced in FDCCSs, such as lanthanum (La), nitrogen (N), carbon (C), sulfur (S), chromium (Cr) and neodymium (Nd) [54].

Being different from doping process, coating treatment introduce thin layer on top or surface of semiconductor layer. In general, coating layer can increase electron collection efficiency or connection between electrolyte and semiconductor layer to reduce recombination, which Nb_2O_5-coated hierarchical TiO_2 NW-NS arrays (Figure 6(l)) achieved obvious improvement in the current of short circuit (J_{sc}=10.5mA/cm^2) and voltage of open circuit (V_{oc}=0.75V) compared with uncoated FDSSCs [91]. Furthermore, Peiris *et al*. reported that $Mg(OH)_2$ coated TiO_2 achieved a highest voltage of open circuit (V_{oc}=0.847V) in FDSSCs, due to the blocking effect [113]. In addition, $TiCl_4$ treatment was also employed to coat TiO_2 on the surface of semiconductors for reducing recombination and increasing dye-absorbing capacity [110].

Plasma treatment is a valid pathway to improve IPCE via passivating the surface of semiconductor or increasing dye absorption capacity. Specifically, the UV-O_3 plasma treatment was used to add functional group on surface of semiconductor and decrease recombination at the ZnO photoanode/electrolyte interface. Early in 2010, UV-O_3 treated ZnO film showed a relative improvement of 20% in IPCE [102]. While fluorine plasma treatment can etch surface of semiconductor to increase the nanopore for enhancing dye pick up. TiO_2 NPs photoanode treated by fluorine plasma also revealed the increase of 25% in IPCE [114].

The passivation is less reported in FDSSCs. It can be carried out through acid treatment,

therefore the semiconductor needs strong resistance to corrosion [115]. Due to low acid resistance, ZnO is not suitable to go through passivation in intense acid for enhancing property.

In addition, FDSSCs also employed the materials with good optical and electronic property to

modify oxide semiconductors for enhancing electron diffusion capability or light absorbing capacity of photoanode. For instance, the photoanode with 3D graphene decorated nanocrystalline TiO_2 films (3DGT) shows a relative augment of 56% in IPCE compared with pristine TiO_2 based photoanode [116], Au nanoparticles modified TiO_2 film achieves a relative improvement of 14% in IPCE [85].

Among the treatments mentioned above, the doping treatments demonstrated incredible capability for increasing conversion efficiency, in particular the doping treatment with lanthanide-series ions. Lanthanide-series ions possess plentiful types and excellent upconversion ability. So far, there are just several kinds of lanthanide-series ions applied in FDSSCs, photoanode doped with other kinds of ions is also worthy investigating.

5.3. Sensitizers

Sensitizer (namely dye) acting as the role to transfer sunlight to current, its property increases the amount of visible light absorption of FDSSCs. The high-efficiency dye possesses four key parameters at least:

- Higher energy of Lowest Unoccupied Molecular Orbital (LUMO) than the energy of semiconductor edge for facilitating injection of excited electrons.
- More functional groups in the dye molecule for enhancing adsorption of dye molecule on the semiconductor surface.
- Wide absorption spectrum to increase incident light harvest and conversion efficiency.
- High stability under intense sunshine and strong redox couple and long lifetime.

With similar operational principle, all sensitizers applied in rigid DSSCs are able to be used in FDSSCs. Sensitizer can be divided into two types—metal organic sensitizer and metal-free organic sensitizer. Their advantages and shortages will be discussed in next section briefly.

5.3.1. Metal organic sensitizers

Metal organic sensitizer is a metal complex, such as zinc-based porphyrin [117], ruthenium-based porphyrin [118], ruthenium-based pyridine [119] and the like. Among metal organic dyes, ruthenium-based-dye family is a standard sensitizer and boosting performance of DSSCs due to their favorable electrochemical properties, highly stable oxidized states, wide absorption spectrum [120, 121].

In the Ru-based-dye family, N3 and N719 are common commercial sensitizers, combined with TiO_2-nanostructured semiconductor to achieve 11.18% IPCE [122], 11.04% IPCE [123] for rigid DSSCs. In FDSSCs, N719 generated highest IPCE of 9.1%, which is also higher than that of metal-free organic dye. Therefore, Ru-based dyes are prime choice for sensitizer in FDSSCs. However, their merits fail to cover up their high cost and hazardous nature.

In addition, Ru-based dye is not suitable to be applied in ZnO-based FDSSCs since Ru dye can remove Zn^{2+} from ZnO lattice resulting in an insulating layer (agglomerate of ZnO^{2+} and dye molecules on ZnO surface) which blocks injection of photo-excited electron to semiconductor [55]. Figure 7 presents that the conversion efficiency deriving from metal organic DSSCs is higher than that of metal-free organic DSSCs. This plot proves powerfully that metal organic dye is the key to improve FDSSCs' performance. Therefore, synthesizing novel nontoxic and pollution-free metal organic dyes will bring breakthrough for creating higher conversion efficiency, for example, the rigid solar cells sensitized by zinc-based porphyrin dye achieved highest IPCE of 14.3%.

5.3.2. Metal-free organic sensitizers

With increasing demand for low production cost, high procurability, eco-friendly devices, increasing effort has been made to exploit alternatives for metal organic dyes. Metal-free organic sensitizers are considered as promising replacement and classified into synthetic metal-free organic sensitizers and natural pigments.

Natural pigments are derived from natural plants by grind, squeeze, filtration and purification [124], which result in its low cost and abundance. However, there is no relevant report about using natural pigment in FDSSCs, due to low performance in rigid DSSCs. Figure 8(a-e) present the absorption spectra of natural pigments deriving from purple cabbage, carrot, purple grape skin, mulberry, potato respectively. It can be observed from these pictures that natural pigments possess narrow light-absorbing band (weak light-absorbing capacity). Among the curves shown in Figure 8(f), pink curve exhibits the highest conversion efficiency of 0.162%, which might be due to large amount existence of non-activated components [125].

In addition, several attempts were made to modify natural pigment and synthesize metal-free organic sensitizers. Among synthetic metal-free organic sensitizers, D149 was applied in FDSSCs and shown an IPCE of 5.24% [77]. Therefore, synthetic metal-free organic sensitizers are

expected to have specific absorption spectrum and high purity of the dyes in FDSSCs.

5.3.3. Improvement from self-assembly and co-sensitization.

In process of the transferring light to current, excited electrons which inject into the semiconductor are from first layer of dye most closely attached on the semiconductor. The dye molecules under first-layer dyes or cluster on surface of semiconductor, which excited electrons fail to inject into semiconductor, and join in recombination reactions.

Dyes are good at taking in the light in certain wavelength range, out of which it shows low light-absorbing capability [126, 127]. Limited light-absorbing capability hindered development of FDSSCs. Moreover, aggregation and stability of sensitizer also are negative factor in FDSSCs' development. Thus, various/a series of attempts have been made to change structure or organic groups of sensitizer molecules for boosting their light-absorbing capability, aggregation-inhibiting ability and stability. The dye with donor/acceptor linked by conjugated π bridge (D-π-A) exhibits strong absorption and high conversion efficiency. Therefore, various attempts have been made to develop the D-π-A dyes for improving performance. Chiu et al. prepared diketopyrrolopyrrole dye containing D-π-A (TEDBn), achieved conversion efficiency of 7.2% [128]. Kang et al. designed and synthesized four D–π–A structured Zn(II)–porphyrin sensitizers with extended π-conjugation, used them by co-sensitization, and then harvested 10.2% conversion efficiency(SGT-012 and SGT-052) [129].

Compared with D–π–A molecules with one donor group, dyes with two donor groups (D–D–π–A)exhibit extended absorption region, enhanced molar extinction coefficient, avoided the π–π aggregation and increased thermostability. Zhang et al. prepared a new D–D–π–A dye (CPAR-1), and achieved 5.9% conversion efficiency [130]. Han et al. synthesized WY6 dye with D–D–π–A structure on T0 dye, promoted conversion efficiency from 4.24% to 5.24% [131]. Qian et al. synthesized D–D–π–A type dyes (QX07) and harvested conversion efficiency of 8.28% [132].

Moreover, some sensitizers with modified D–π–A structures also are designed and synthesized widely [133-136], such as D–A–π–A, D–π–A–π–A, A–π–D–π–A, A–D–π–D–A etc. Eiamprasert et al. found that the asymmetric double D–π–A di-anchoring architecture consisting of two different D–π–A units in a single molecule not only provides an alternative route to harvest the panchromatic solar spectrum, but can also be used to regulate the charge recombination [137].

In the dye-absorbing process, dye molecules are randomly adhered on the surface of semiconductor layer, thus there are a lot of clusters of dye molecule on the surface, which weaken the efficiency of dye and the light-absorbing capability of photoanode [138]. Therefore, it is reliable for improving light-absorbing capacity to combine with a kind of co-adsorbent to facilitate self-assembly of dye molecules on the semiconductor surface, or to employ two kinds of dyes with different size or inverse light-absorbing band to sensitize photoanode [139-143].

In a rigid DSSCs, Yu *et al*. found that chenodeoxycholic acid (CDCA) as co-adsorbent hinder the phthalocyanine aggregation [138]. Li *et al*. used three di-n-alkylphosphinic acids (DPAs) as co-adsorbent, and then accelerated conversion efficiency increase to 6.09% from 5.53% [144]. In terms of co-sensitized method, Lee *et al*. designed the FDSSCs co-sensitized by an organic dye (JH-1) and a squaraine dye (SQ2), achieved IPCE of 6.31% [84]. In addition, Xue *et al*. prepared the rigid solar cell sensitized by D131and SQ2 dye with cocktail or layered method (its schematic shown in Figure 9(a)). D131 dye and SQ2 dye possess complementary light-absorbing spectrum (shown in Figure 9(b)). According to the Figure 9(c) and 9(d), the solar device sensitized with layered method exhibited higher conversion (4.1%) efficiency than that with cocktail co-sensitized method (3.3%) [145].

These reports above offer some novel approaches to enhance FDSSCs' performance. Namely, synthesis of novel D–π–A sensitizers can enhance light-absorbing capability. For the further promotion in light absorption, the co-sensitization by two dyes owning complementary light-harvesting spectrum in with layered-sensitized method will be promising. In respective dye-absorbing process, using co-adsorbent are efficient ways to restrain dye aggregate and to

enhance light-harvesting capability, thus improving conversion efficiency tremendously.

5.4. Electrolytes

Electrolyte works as the aisle provides the internal electrical conductivity in light-to-current process and is divided into liquid, quasi-solid (gel) and all-solid electrolyte. Among the three electrolytes, all-solid electrolyte is hardly employed in FDSSCs since its low charge mobility, high resistance and poor interfacial connection leads low conversion efficiency. On contrary, the liquid and quasi-solid electrolytes are often used in FDSSCs because their high ion mobility and remarkable interfacial connection with dyes result in high conversion efficiency [146-148].

Liquid electrolyte is employed widely in FDSSCs [149]. Employing liquid electrolyte in FDSSCs can obtain excellent connection between dye layer and electrolyte to accelerate dyes' redox reactions, but liquid electrolyte always suffer from instability, volatility and difficult seal. Moreover, utilization of liquid electrolyte increases the risk of leakage because FDSSCs endure more bends. So the quasi-solid electrolytes are a better option for FDSSCs due to its good ion mobility, low flow, long lifetime and easy seal [150, 151].

Attempts have been made to study quasi-solid electrolyte, for instance, Chen *et al*. reported that the MSNs based gel-electrolyte exhibits high reflectance in the visible wavelength and enhanced the ionic conductivity and achieved IPCE of 5.45% [152]. Bella *et al*. claimed the paper-based quasi-solid dye-sensitized solar cells with the IPCE of 5.2% [153], even the FDSSCs with dual-function quasi-solid-state electrolyte obtained IPCE of 7.2% which is close to performance of liquid-electrode FDSSCs (7.35%) [154]. The above reports have proved excellent property and tremendous potential of quasi-solid electrolyte in FDSSCs.

In terms of redox couple, iodide-based redox couple (I^-/I_3^-) is most frequently employed as mediator in FDSSCs, but its corrosion effect, intense visible light absorption and low redox potential push researchers to look for replaceable redox [55, 80]. Given no variation between flexible and rigid DSSCs in operational principle, the redox couples employed in rigid DSSCs, such as cobalt-based redox couple (Co^{2+}/Co^{3+}), vanadium redox couple (V^{2+}/V^{3+}), organic sulfide redox couple (T_2/T^-) and copper-based redox couple (Cu^+/Cu^{2+}) all can be introduced directly into FDSSCs [155-158]. Therefore, quasi-solid electrolytes combine with high-performance redox couple will be first choice for FDSSCs and make FDSSCs more promising and competitive.

5.5. Counter electrodes

Counter electrode works as a vital role transferring charge and reducing I^-_3 to I^- in FDSSCs [159]. High-efficiency CE is an indispensable part for FDSSCs with prominent IPCE. Generally, high-performance CE should have the characteristics as follows [29] :

- Low resistance to reduce loss of voltage of open circuit (V_{oc}).
- High catalysis to facilitate reduction of tri-iodide (I^-_3)
- Robust stability to improve lifetime.

 Excellent transmittance when light needs incoming through CE.

According to the constituent elements, CE is classified as metal-based, carbon-based and composite-based electrodes. Their respective features are discussed as follows:

5.5.1. CEs based on carbon materials

Carbon materials have plentiful kinds (such as carbon nanotubes, carbon nanoparticles, graphene and so on), abundant resource and decent conductivity, which make them become good catalyst in CE.

Graphene is considered as a revolutionary material because of its optical, electrical and mechanical properties [160]. Using graphene in flexible CE is a promising way to enhance IPCE. Lukaszkowicz *et al*. prepared graphene-based CE by depositing graphene on the PET/ITO for

FDSSCs, and then attained conversion efficiency (3.95%) closed to that of Pt-based CE (4.39%) [161]. Sahito *et al*. fabricated textile fabric-supported graphene nanosheets as CE for DSSCs, the solar cells achieved excellent IPCE of 6.93% [162]. Obviously, the graphene possesses enormous potential as CE. But now, the yield and the price of graphene both are the obstacle for its application in FDSSCs.

CE based on low-cost carbon material is a current study hotspot. Tathavadekar *et al.* demonstrated that a flexible conducting carbon cloth (shown in Figure 10(b-c)) obtained by one-step pyrolysis (illustrated in Figure 10(a)) of cellulose fabric was assembled with rigid photoanode and achieved the IPCE (5.8%), which is close to conversion efficiency of same device with Pt/ITO/glass CE (7%) [163]. In rigid DSSCs, Cha *et al*. claimed that a honeycomb-like porous carbon (its raw material, macroscopic and microscopic morphology shown in Figure 10(e-f)) derived from fallen leaves was used as flexible CE and achieved 5.52% IPCE [164]. This report provided a good direction—preparing the CE based on natural plant templates. In the nature, plentiful plants own elegance microstructure, so carbon-based CE deriving from natural plant will result in a reduction in cost.

Besides, combining carbon materials with metal or metal compounds is a usual method to boost catalytic activity [165]. Like the report, preparation of a Pt/SWCNT CE at low temperature for FDSSCs has an increasing of 25.74% in efficiency, compared to the flexible DSSC with an unmodified Pt CE [166]. However, carbon materials directly deposited on a flexible substrate shows a poor mechanical stability.

5.5.2. CEs based on metals and metal compounds

Transition metals, transition metal compounds and alloys have been researched to explore the possibility as CE in DSSCs. Among those metal materials, platinum is a remarkable catalytic material and generally used as CE by loading platinum on flexible substrate. For instance, Pt NPs/ITO/PET CE prepared by a pulsed-mode electrochemical deposition (Pulse-ECD) technique was employed in FDSSCs and generated an IPCE of 4.3% [167], other FDSSCs with Pt/Ti CE achieved an IPCE of 6.13% [168].

But its high price limits its application in commercial field, a lot of attention has been paid to find alternatives for Pt. According to reports, several transition metal compounds show high catalytic potential [29], in particular transition metal sulfides [156]. Cheng *et al.* employed the molybdenum sulfide thin film synthesized on conductive plastic substrates (MoSx/ITO/PEN) as CE and achieved a conversion efficiency of 4.39% [169]. Yue *et al*. reported the IPCE of 7.2% was obtained by employing NiS/Pt/Ti CE [170]. Zhang *et al*. fabricated a transparent ITO/CuS NN film (shown in Figure 11(b)) followed the fabrication processes shown Figure 11(a), and then employed it as CE to gain IPCE of 6% [171]. Obviously, using transition metal sulfides as CE materials accelerates the commercialization of cost-efficiency FDSSCs.

Although metal and metal compounds have excellent catalytic potential, the metal and metal-compound CEs fabricated on flexible substrate via deposition have poor mechanical stability. Therefore, fabrication of CE with high mechanic stability is desirable technique. Khan *et al*. fabricated a new type of embedded metal-mesh transparent electrode (EMTE) with in-situ electrodeposited catalytic Pt NPs as a high-performance CE (denoted as PtNP-EMTE) for lightweight flexible bifacial DSSCs. The conversion efficiency under rear illumination is 4.87% comparing to that under front illumination (5.67%) [172]. Its fabrication process and morphological characterization exhibit in Figure 12(a), 12(b), 12(c) respectively. Figure 12(d) reveals that PtNP-EMTE possesses excellent optical transmittance. The results indicate that PtNP-EMTE is likely to enhance performance of the FDSSCs with metal foil substrate-based photoanode. However, this process needs advanced equipment and complicated technology such as sputtering and evaporating [173, 174]. So it is essential to find a simple approach to fabricate

high catalytic-performance and mechanical-stability CE.

5.5.3. Composite CEs

In terms of respective advantages of metal-based and carbon-based CE, composite CE is comprised of single polymer or combination of several materials among metals, metal compounds, carbon materials and polymers. For instance, Zhu at al. claimed a composite Pt/carbon spheres (Pt/CS, shown in Figure 13(a)) CE was prepared and used in plastic FDSSCs, achieved IPCE of

3.94% [175]. Liu *et al*. prepared polypyrrole/reduced graphene oxide (PPy/RGO, shown in Figure 13(b)) CE on plastic conducting substrates for FDSSCs via facile two-step electrochemical process at low temperature and reached IPCE of 4.25% [176]. Bao *et al*. prepared the platinum NPs coated TiO_2 NTs/Ti mesh CE which showed superior electrocatalytic activity, good stability and low series resistance and achieved IPCE of 5.6% [177]. Obviously, composite CE possesses enormous potential in catalyst, but it could not meet a long-term stability. Therefore, Yin *et al*. prepared flexible Ag@ZrO_2/C nanofiber film by electrostatic spinning as CE in flexible plastic DSSCs for enhancing device's conversion efficiency and mechanic stability. As a result, this device achieved an IPCE of 4.77% [178]. However, this needs a long fabricating time and complicated processes.

Fortunately, conductive polymers, such as PEDOT and PANI, were introduced as CE to simplify fabrication procedure of CE and develop high-performance CE due to high conductivity, excellent doping-capacity and stability. Among conductive polymers, PEDOT not only catalyzes the reaction of tri-iodide/iodide redox couple solely due to the formation of charge-transfer complexes between iodine species and polymeric rings [104], but also combines with other nanomaterials to improve catalytic performance. For instance, Ke *et al*. prepared the flexible PEDOT:PSS/Triton CE by spin-coating. The FDSSC shows a higher catalytic (3.74% IPCE) than that of Pt/Triton CE (3.52% IPCE) [179]. Ma *et al*. synthesized 3, 4-ethylenedioxythiophene

(PEDOT) on ITO/PEN (shown in Figure 13(c)) by electrochemical methods as flexible CE, combined with rigid photoanode, subsequently achieved IPCE of 7.18% [180]. Besides, Peng *et al*. produced a nano-TiC/graphene/PEDOT:PSS composite material on ITO/PET by spray printing for

FDSSCs as CE, the conversion efficiency of 4.5% is higher than that of similar device with Pt CE (4.3%) [181]. The electrodes mentioned above all possess excellent flexibility and easy-implemented fabrication process.

In addition, polyaniline is also employed to prepare high-performance CE, for example,

Motlagh designed the polyaniline-coating carbon fabric (PANi/CF) as a novel textile-based CE, and attained IPCE of 3.81% [182]. Yue *et al*. prepared the nickel sulfide/polyaniline/titanium (NiS/ PANI/Ti, shown in Figure 13(d)) counter electrode through electrodeposition and assembled with Ti foil photoanode to be a FDSSCs. As a result, this device achieved better IPCE (7.35%) compared with the FDSSCs with the Pt/Ti CE (6.24%) [183]. Given what mentioned above, the composite CEs based on combinations of conductive polymers and transition metal sulfides will be a good direction for developing the high-catalytic-performance, strong-mechanical-stability, low-cost and easy-prepared CE, particularly when nanoparticles of transition metal sulfide just attach on surface of porous polymer, but not be covered by polymer.

6. Applications

With the ever-increasing deployment of micro/nano electronic systems, the need grows for self-sustaining, constant power sources. The scientific and industrial communities are actively pursuing an intensive effort towards integrating energy harvesting and storage devices able to drive low power electronics. In this framework, flexibility represents a mandatory requirement to cover non-planar or bendable surfaces, more and more common in nowadays-electronic devices. Here we present three innovative integrations of FDSSCs with multiple types of electronics into a single device.

6.1. Integrating with portable electronic device

Electronic devices have been a kind of symbol of our age, in particular portable handheld devices (namely mobile terminals). The mobile terminals bring tremendous convenience to us. However, mobile terminals still suffer from limited stored power energy and frequent charge because the most just depend on the power from Li-ion battery, which limited utilization of mobile terminals. Therefore, the portable power with lightweight, small volume, highly cost efficiency and sustainability was expected widely for supporting long service time or a long journey.

The FDSSCs are the device meeting those conditions mentioned above. Flexibility of FDSSCs makes itself to adapt non-planar or bendable surfaces, and complicate space, which also can optimize user's experience. FDSSCs can be manufactured with various sizes for different needs. Xu *et al*. fabricated the FDSSCs which consisted of a CuS/PET CE and a Ti foil-based photoanode and shown tremendous potential in wearable energy-harvesting device [184]. Its

schematic diagram and corresponding device prototype are presented in Figure 14(a), 14(b).

Figure 14(c) shows the J-V curves of FDSSCs with different CEs. Subsequently, five CuS-based FDSSCs were assembled in series, and then three LEDs were lightened successfully (shown in inset photo). In addition, this device kept 91% of the original conversion efficiency after 500 bending cycles (namely 9% decrease, shown in Figure 14(d)) with excellent stability. This report provided a powerful witness for practical application of FDSSCs. With the development of technology, the stability of flexible device will be optimized further. Given its flexibility and thinness for easy carry, it will be more popular to integrate with portable electronic device.

6.2. Integrating with another energy-harvesting device

Harvesting solar energy via solar cells is changing status of the global energy crisis and environmental pollution. However, solar energy is intermittent or even impossible to access in certain circumstances, such as a cloudy day or indoor environment. Meanwhile, the development of wearable electronic device drives the demand for flexible power sources rapidly.

Therefore, researchers proposed to combine solar cells with nanogenerator (NG) to overcome the challenge. Pu *et el.* developed a wearable power textile consisted of a grating-structured textile-based triboelectric nanogenerator (TENG) [185]. Subsequently, Yun *et al*. integrated the

TENG and a fiber-based FDSSC into a cloth as shown in Figure 15(a) [186]. The TENG, consisted of a slider fabric located on the sleeve and a stator fabric fixated underneath the arm (Figure 15(b)), was aimed to harvest the swing energy of arms during walking and running. The relative sliding of the slider and stator fabric will generate alternating current between the interdigitated electrodes in the fabric. The fiber-based FDSSC was prepared via winding Pt wire CE around TiO_2 coated Ti wires photoanode in a flexible plastic tube (Figure 15(c)). The FDSSC are connected with the TENG fabric via a bridge rectifier (Figure 15(d)). Figure 15(e) shows short circuit current of FDSSCs, TENG fabric and integrated device. Notably, the short circuit current of integrated device is larger than that of either. The integrated energy-harvesting device possesses stronger capability in harvesting energy application for portable electronics.

6.3. Integrating with energy-storage device

Solar cells only generate electric power, but fail to store electric energy. Traditional solar cells are unable to supply the power in the night, or the electric energy generated in the day is likely to be wasted without utilization. Therefore, stored-energy devices are needed to store

energy for avoiding waste or showing effect at the nighttime. Supercapacitor (SC) is deemed as the most

promising stored-energy device in storage of electric energy. Particularly, flexible SC can combine with FDSSCs, and this combination will bring a revolution in solar cells, due to its excellent flexibility, thin thickness, lightweight, long lifetime and high power density. A harvesting-storage device integrating flexible SC with FDSSCs can provide the temporal flexibility to balance local power generation and consumption, and improve the overall energy utilizations. In other words, the FDSSCs in harvesting-storage device generate power continuously, and supercapacitor in harvesting-storage device stores the excess or unused energy for future utilization.

To this purpose, Scalia *et al*. designed an innovative device consisting of a flexible architecture integrated a TiO_2 nanotube-based FDSSCs with a graphene-based electrical double layer flexible SC [187]. In this report, FDSSCs were integrated with flexible SC, shown in Figure 16(a). This device is a dual functional electronics (shown in Figure 16(b-c)). Its operational principle can be described by two steps. Namely, two electrodes of the capacitor were connected with photoanode and CE of FDSSCs respectively, and integrated device was placed under simulative sunlight to charge the capacitor (photo-charge, Figure 16(d)). The connected electrodes were switched off when the capacitor stored adequate electric energy. Thus, load can be connected with capacitor to attain power when power was needed by load (discharge, Figure 16(e)). This device can overcome the temporal and territorial limitation that the energy-harvesting device faced with, and promote good experience.

In addition, the energy harvesting-storage device integrated NG, DSSCs and SC were also reported [188]. This report provided creative direction for application of FDSSCs, namely developing flexible/portable integrated energy-harvesting device based on FDSSCs, NG and SC.

7. Developmental tendency

With increasing demand for eco-friendly life, high cost-efficiency and portable power source, FDSSCs as promising and competitive candidate have been investigated widely. The highest conversion efficiency is less than 10%. Therefore, more effort must be made to optimize FDSSCs for higher conversion efficiency and cost-efficiency.

Currently, the most of substrates present the incompatibility between high conductivity and high transmittance, which result in enormous/serious light loss or weak/insufficient charge collection. Novel film technique should be developed to break this incompatibility for higher

transmittance and conductivity.

For semiconductor thin films, surface area and light-scattering capability are two opposite parameters for increasing sunlight utilization. And hierarchical structure, core-shell structure and cocktail film of metallic oxides are potential way to improve in both high surface area and high light-scattering capability. Since crack and separation of film result in high recombination, low charge collection and unstable output, forming firm/strong and stable interconnection between semiconductor layer and substrate is worth investigating. Moreover, employing the photoanode doped with upconversion ions that can turn near infrared radiation (NIR) into visible light also is effective/desirable way to boost sunlight utilization of photoanode further.

The nontoxic-metal or metal-free organic sensitizers has been popular sensitizers under eco-friendly challenge. The sensitizers with D-π-A or modified D-π-A structure, such as double donor group (D-D-π-A) and asymmetric double D-π-A di-anchoring architecture consisting of two different D-π-A units in a single molecule, have been confirmed that they are possible/likely shown high conversion efficiency. Thus, synthesizing novel modified D-π-A sensitizers will be a possible way to increase the performance of FDSSCs. Moreover, their co-sensitization also is a studied focus/promising direction for high performance.

The quasi-solid (gel) electrolyte with higher carry mobility is a research focus due to its significance for the long-term stability and high conversion efficiency of FDSSCs. Meanwhile, close interconnection between sensitized nanostructured semiconductor layer and electrolyte also play a crucial role in high-efficient reductions of sensitizer/charge cycles. Therefore, developing the quasi-solid electrolyte with excellent flow/penetration ability under certain condition will accelerate the application of FDSSCs significantly.

In addition, the growing demand for low product cost, high-efficiency catalyst and mechanical stability accelerates researches of Pt-free CE. The combination of metal sulfides nanoparticles and conductive polymer shows enormous advantages in product cost, catalytic ability and mechanical stability.

8. Conclusion

Growing markets for portable electronics need high-performance and low-cost flexible film solar cells as power sources. In this review, the contrast between rigid and flexible DSSCs was

made for understanding the differences. Representative investigations were selected and discussed according to their functionalities, such as substrate materials, semiconductor (morphology and optimization), sensitizers, electrolytes and counter electrode. The metal-substrate FDSSCs (9.1%) possess more potential to achieve high conversion efficiency than plastic-substrate FDSSCs (7.6%), in particular Ti-substrate FDSSCs. Compared with TiO_2, ZnO with better electron mobility, resistance of photocorrosion lower their conversion efficiency. But the limited flexible substrates made TiO_2 hard to obtain high conversion efficiency except Ti. So preparing TiO_2 with various morphologies by employing ZnO template was mentioned for expanding applications of TiO_2 in FDSSCs. Sensitizers decide the light-absorbing capability of photoanode and affected the conversion efficiency directly. Ru-based sensitizers possessed remarkable light-absorbing capacity, its high prices, low yield, toxicity and low adaptability for ZnO drive researchers to look for alternates for eco-friendly devices. Co-adsorbent and co-sensitization were recommended to be used in order to improve light-absorbing capability.

Quasi-solid or all-solid electrode will be better choice for FDSSCs, and show similar efficiency as liquid electrolyte. Moreover, combining the metal sulfides nanoparticles with high catalytic capability and the transparent conductive polymer with excellent adhesion will enhance catalytic properties and mechanical stability of CE effectively. In addition, this review provides three promising applications of FDSSCs for portable electronic devices.

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