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1 systems for electrochemical energy conversion and storage. Although recent studies  
2 only provided the initial ideas, flexible fiber/wire-shaped solar cells facilitated the  
3 breakthrough for novel solar architecture and portable/wearable electronics or  
4 e-textiles of the future.

5 **Keywords:** fiber/wire shaped solar cells; integrated power systems; energy  
6 conversion and storage; portable/wearable electronics

7

## 8 **Introduction**

9 Energy and environmental issues are two of the most prominent problems in  
10 modern society. For sustainable development, scalable and clean energy conversion  
11 via solar cells has attracted both academic and industrial attention since the  
12 foundation of photoelectric effect. The past years have witnessed the expanding  
13 market of photovoltaic cells based on Si, CdTe and CIGS ( $\text{Cu}(\text{In,Ga})(\text{S,Se})_2$ ), as well  
14 as the rapid development of new photovoltaic generations, such as CZTS  
15 ( $\text{Cu}_2\text{ZnSn}(\text{SSe})_4$ ), organic solar cells, dye-sensitized solar cells (DSSCs), and  
16 perovskite solar cells<sup>1, 2</sup>. In particular, the report by National Renewable Energy  
17 Laboratory indicated that the solar cell types and optimal research-cell efficiencies  
18 have been continuously refreshed in the past 40 years<sup>3</sup>. The most significant feature of  
19 typical solar cells is the planar shape with dimensional extension. Thus, the  
20 development of one- or three-dimensional (1D or 3D) photovoltaic cells will break  
21 through the limitations of conventional thinking and generate a new concept for solar  
22 energy conversion. This vision could also create opportunities for material

1 science/structure design that would be interesting as technical innovations and  
2 beneficial for special energy applications.

3 With the rapid development of traditional industry and increasing demand of  
4 modern electronics, the integrated optoelectronic products come to the age of small,  
5 portable, lightweight and smart merchandise. In recent years, wearable/portable  
6 electronics, such as Google glasses, iWatch, and smart phone, have gradually become  
7 part of human lives. To deepen the wearable concept and meet future individual needs,  
8 incorporating flexible optoelectronic devices (e.g., sensors, logic circuits, antenna,  
9 lighting elements, and power systems) in clothing, backpacks, and other belongings  
10 would become an important trend for the next generation of smart products, namely  
11 “electronic textiles” or “e-textiles”<sup>4,5</sup>. The fundamental functional units for e-textiles  
12 remain to be developed. Especially, flexible power sources with efficient energy  
13 conversion and storage can automatically generate electricity and contribute to  
14 self-driven energy systems<sup>6-8</sup>, which are very promising candidates for the  
15 portable/wearable electronics that can further enrich e-textiles<sup>9-11</sup>. Among the energy  
16 technology innovations, flexible fiber/wire-shaped energy devices, including solar  
17 cells, batteries, supercapacitors, light-emitting diodes, and field-effect transistors, are  
18 booming in recent years and attracting increasing attention<sup>12-18</sup>. Flexible  
19 fiber/wire-shaped energy devices fabricated on fiber- or wire-type conductive  
20 substrates are the suitable units for contexture because of their special linear shape  
21 and may play a role in practical e-textiles, such as wearable solar power sources and  
22 self-powered smart systems. In particular, fiber/wire-shaped solar cells demonstrate

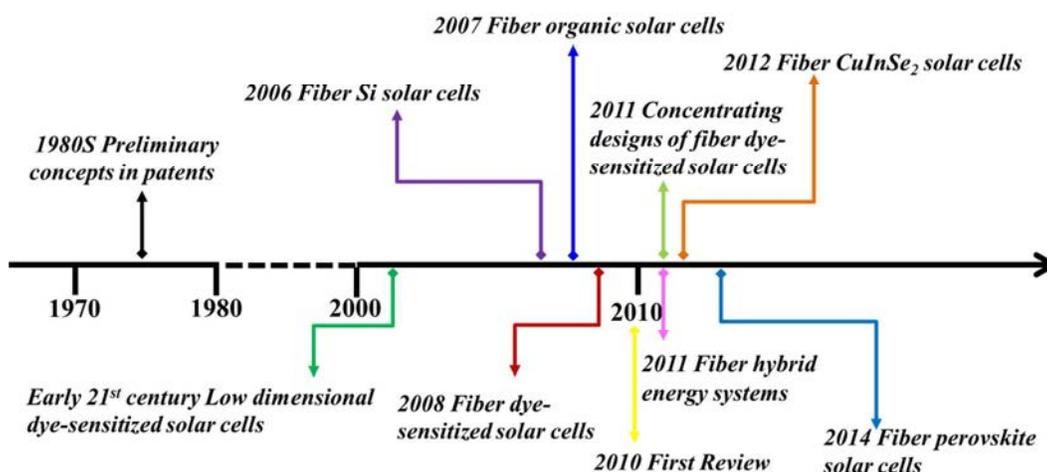
1 solar-to-electric conversion but comprise special photovoltaic architecture fabricated  
2 on fiber substrates, which endow the devices special properties and allow innovations.

3 Since 2010, several reviews have scanned the basic principle and conventional  
4 device structures of fiber-like solar cells<sup>12-14</sup>. However, the coverage and concept have  
5 been spreading because of the rapid expansion of this emerging research field in the  
6 last three years. For clarify the development tendency, the development of flexible  
7 fiber/wire-shaped solar cells, including inorganic, organic, dye-sensitized, and  
8 perovskite photovoltaic fibers, are presented. Focus was given on the properties,  
9 materials, and designs of fiber-shaped dye-sensitized solar cells (FDSCs) because  
10 they are the most developed type and could provide certain guidance to the other  
11 types. The shortcomings of recent research and future development were also  
12 summarized on the basis of status quo. In the present review, a very detailed insight  
13 into the development of fiber solar cells was provided in Part 1, covering research  
14 motivation, conventional materials, structure designs, and future perspectives.  
15 Properties of fiber solar cells associated with structure design were discussed in Part 2.  
16 Attention was paid to the materials explorations of FDSCs in Part 3; and solar devices  
17 with special energy harvesting design were provided in Part 4 to broaden the fiber  
18 based innovations. In Part 5, the cases that fiber solar cells integrated into hybrid  
19 systems were highlighted for energy management. Given the rapid development,  
20 works until March 2015 were summarized, and a fast development momentum for  
21 this highly growing field may exist in the future.

22

## 1 Development of Fiber-Shaped Solar Cells

2 The preliminary concepts of fiber/wire shaped solar cells appeared in some patents  
 3 since the 1980s, in which silicon layers formed p-n or p-i-n junctions on conducting  
 4 fiber substrates to obtain a low-cost and flexible devices<sup>19</sup>. The real progress of this  
 5 field started at the beginning of 21<sup>st</sup> century, where new types of fiber solar cells and  
 6 structure designs are booming up with rapid development of photovoltaic field<sup>13, 19-28</sup>.  
 7 The past records of fiber solar cells is presented in **Figure 1**; and continuing  
 8 development context of fiber inorganic, organic, dye-sensitized, perovskite solar cells  
 9 is summarized from Part 1.1 to 1.4, respectively.



10

11 **Figure 1** Past records of fiber solar cells<sup>13, 19-28</sup>.

12

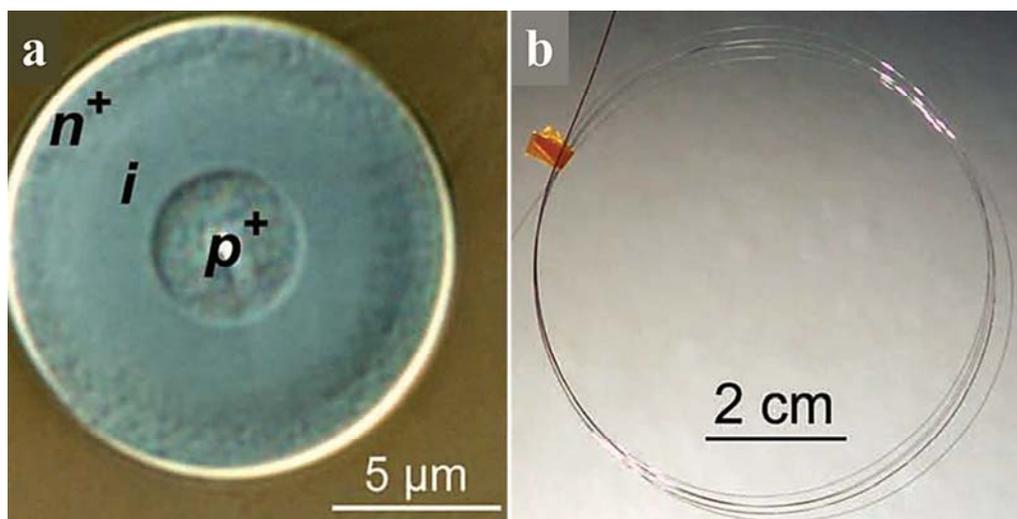
### 13 1.1 Fiber Inorganic Solar Cells

14 Silicon solar cells, based on the high natural abundance element, have experienced  
 15 a long research history. The excellent reliability and acceptable cost make them  
 16 mainstream products in the photovoltaic market. Silicon technology based on vapor  
 17 deposition could be adapted to different substrates, and it is a good choice to construct

1 solar cells and detectors on flexible fibers. Early explorations since 2006 have  
2 claimed flexible fiber-type poly-Si solar cells on glass fiber<sup>22</sup> and carbon fiber<sup>29</sup>, but  
3 the device performance were very low. In 2013, Badding et al<sup>30</sup> fabricated flexible  
4 coaxial silicon fiber pores with p-i-n photodiode junction (**Figure 2**) through  
5 high-pressure chemical vapor deposition. Flexible silicon fibers with length of up to  
6 10 m were deposited, which demonstrated a potential for scalable preparations. The  
7 low light absorption of the active layers resulted in an overall conversion efficiency of  
8 0.5%. Later on, Gibson et al.<sup>31</sup> reported silica sheath fibers by using bulk glass draw  
9 techniques with low purity p-type silicon polycrystalline as starting material. The  
10 silica that formed into silicon core was etched from one side and deposited with a-i-Si  
11 and a-n-Si layers via PECVD to form a p-i-n junction. The silicon-core glass fiber  
12 achieved efficiencies of up to 3.6%, leaving room for improvements over the  
13 presented prototype. Except Si fibers, Chen et al.<sup>27</sup> prepared flexible fiber-shaped  
14 CuInSe<sub>2</sub> (CIS) solar cells by ordinal electro-deposition of CuInSe<sub>2</sub> layer, chemical  
15 bath deposition (CBD) of CdS, and then RF magnetron sputtering of ZnO and ITO  
16 layers on the Mo wire (**Figure 3a**). The single-wire solar cell achieved energy  
17 conversion efficiency of up to 2.3% and exhibited excellent long-time stability (stored  
18 at 60 °C for 0-600 h) (**Figure 3b**).

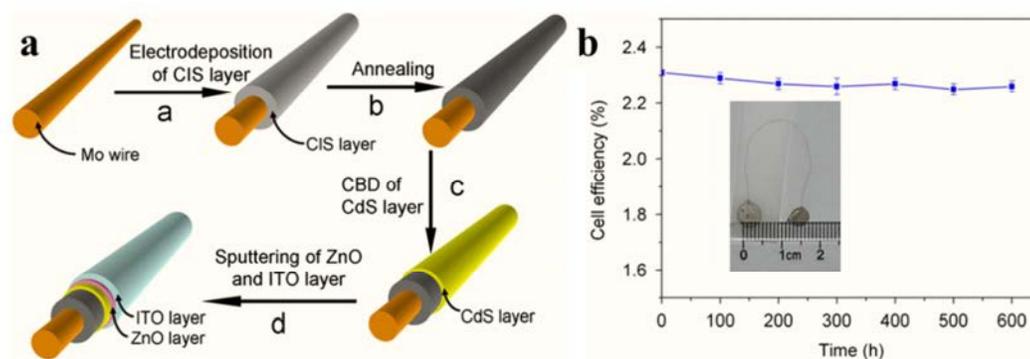
19 Mature preparation based on silicon industry, scalable technology via vapor  
20 deposition, and reliable stability of all-solid devices are the advantages of inorganic  
21 fiber solar cells, which are the key points for wearable electronics. However, a long  
22 period of efficiency improvement occurred, which remains an important issue in

1 future development. Significant improvement and great chances for applications are  
 2 available considering the performance of traditional inorganic photovoltaic cells and  
 3 the solid foundations of silicon industry.



4  
 5 **Figure 2** (a) Differential interference contrast optical micrograph of a representative  
 6 15  $\mu\text{m}$  diameter Si p-i-n junction; (b) Flexible silicon fiber solar cell of  $\sim 1$  m long  
 7 based on p-i-n junction<sup>30</sup>. Reproduced from Ref. 30 with permission, Copyright ©  
 8 2013 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

9



10

11 **Figure 3** (a) Fabrication of Mo/CIS/CdS/ZnO/ITO single-wire flexible solar cells; (b)

12 The time-dependent change in the conversion efficiency of the single-wire solar cell



1 stored at 60 °C for 0-600 h. Inert is the photo of single-wire device<sup>27</sup>. Reproduced  
2 from Ref. 27 with permission, Copyright © 2012 Elsevier Ltd.

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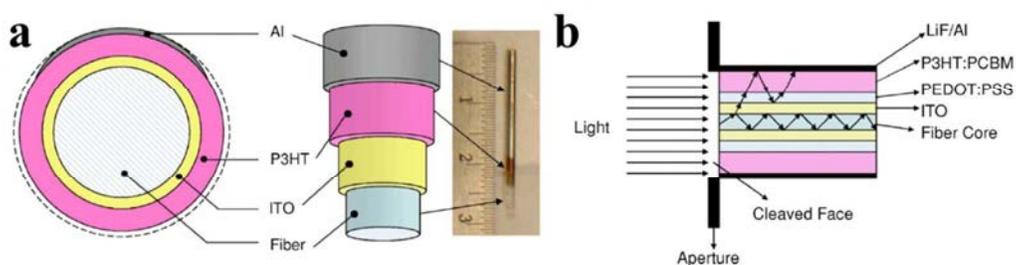
## 4 **1.2 Fiber Organic Solar Cells**

5 In traditional planar organic photovoltaics, efficient charge transport and optimal  
6 optical absorption was usually not capable. To break through this contradiction,  
7 Carroll and co-workers<sup>23, 32</sup> demonstrated fiber organic solar cells based on waveguide  
8 optical geometries in 2007 (**Figure 4**). The device with a structure of optical  
9 fiber/ITO/PEDOT:PSS/P3HT:PCBM/LiF/Al achieved efficiency of 1.1%<sup>32, 33</sup>. But the  
10 optical fibers are relatively expensive and less flexible for weaving cases. Flexible  
11 polyimide-coated silica fibers and polypropylene fibers were demonstrated as possible  
12 alternative substrates for organic photovoltaic fibers via evaporation or solution  
13 process<sup>34, 35</sup>. Driven by the goal of efficient light absorption in wide incident angles  
14 and more freedom of physical appearance, non-planar cylindrical devices with tube  
15 and glass rod substrate were also created, with efficiencies from 0.6% to 1.4%<sup>36, 37</sup>.  
16 Equivalent circuit model analysis found that the aspect ratios to obtain the optimum  
17 overall optical confinement and optimal power conversion performance were between  
18 1 and 5<sup>38</sup>. To eliminate usage of transparent oxide semiconductors, Gaudiana et al.<sup>39</sup>  
19 reported efficient flexible organic photovoltaic fibers with efficiency of up to 3.9% in  
20 2009. In their design, two twisted functional fibers, one comprising a conducting  
21 polymer and fullerene derivative and the other coated with a silver film, were  
22 wrapped together and encased. In further attempts to replacing the metal counter

1 electrode, a carbon nanotube (CNT) fiber was used to twist the fiber anode with  
2  $\text{TiO}_2/\text{P3HT}:\text{PCBM}/\text{PEDOT}:\text{PSS}$  layers, and the photovoltaic wires achieved  
3 efficiency of up to 1.8%, which maintained by ~85% after bending (bending radii of  
4 ~5 mm) for 1000 cycles<sup>40, 41</sup>. Cao and co-workers<sup>42, 43</sup> demonstrated that CNT films,  
5 CNT yarns (**Figure 5a** and **5b**), and single-layer graphene as counter electrodes can  
6 contribute to 2.3% to 2.5% efficiency. The devices show stable behavior during  
7 bending and moderate stability under inertia gas for 20 days (**Figure 5c** and **5d**).  
8 Especially, involving carbon based film as the second electrode could avoid thin  
9 layers penetrating during twisting, maintain good interface contact to the inner layer  
10 and provide satisfying light transparency. In further attempts, flexible, lightweight,  
11 easy-processable organic photovoltaic textiles (**Figure 6**) with reasonable  
12 photovoltaic performance were developed, which provide a promising avenue toward  
13 wearable electronics<sup>44, 45</sup>. Light harvesting, oscillator strength, and low charge  
14 mobility limitations are the hindrances toward efficient organic solar fibers, but  
15 cost-competitive and large-scale fabrication of traditional organic solar platforms are  
16 claimed to be possible<sup>46</sup>. Moreover, 17% efficiency can be achieved using theoretical  
17 calculations in a reflective tandem architecture<sup>47</sup>.

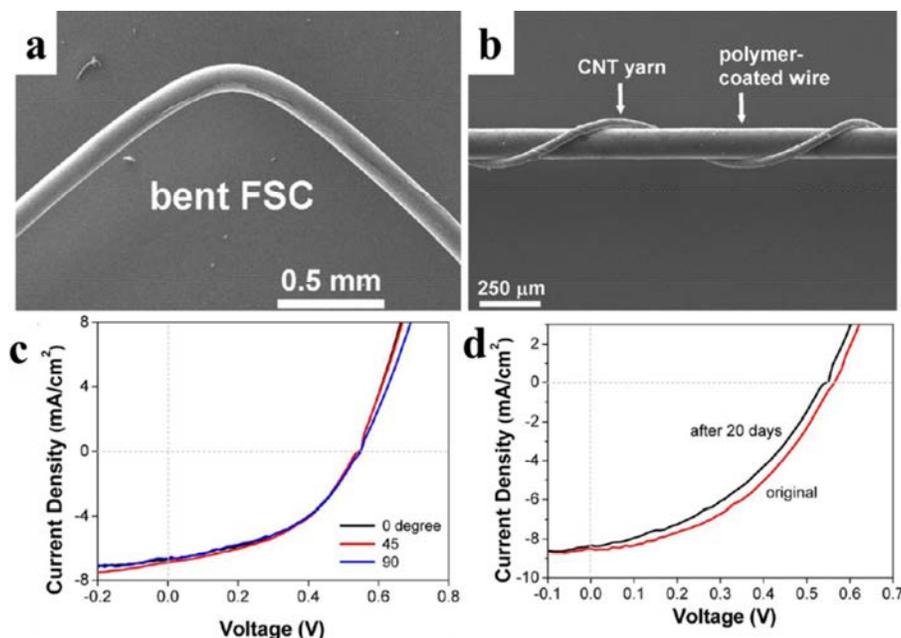
18 Organic photovoltaics provide material basis for flexible and stable fiber solar  
19 cells, which stimulates wide research motivation for wearable solar power sources.  
20 Though solution process would bring much convenient in film preparation,  
21 dip-coating rather than conventional spin-coating is used to deposit the active layers  
22 for fiber/wire type organic solar cells. The difficulties in preparing long-range,

1 uniform, and controllable thin films and forming efficient interface junctions on  
 2 curved surface is a real technical challenge for highly efficient organic fiber devices.  
 3 Further research would see the corporation of novel organic/polymer photovoltaic  
 4 materials, improved functional interfaces and advanced optical structures.



5  
 6 **Figure 4** Organic fiber solar cell based on optical fiber (a) and its light harvesting  
 7 path (b)<sup>32</sup>. Reproduced from Ref. 32 with permission, rights managed by AIP  
 8 Publishing LLC (2007).

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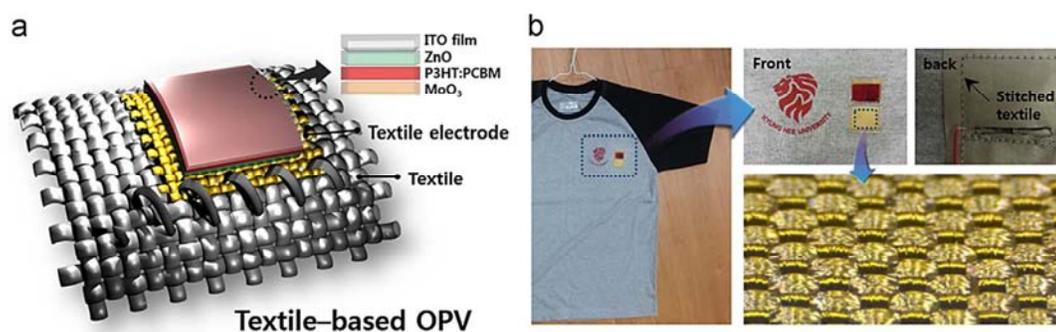


10

11 **Figure 5** (a) Organic fiber solar cell with CNT films as counter electrode (bent to  
 12 nearly 90 degrees). (b) Organic fiber solar cell based on CNT yarns. (c)  $J$ - $V$  curves of

1 a CNT films based organic fiber solar cell in straight form (0 degree) and bent to 45  
 2 and 90 degrees, respectively, showing stable behavior during bending. (d)  $J-V$  curves  
 3 of the CNT yarns based organic fiber solar cell in original state and after storage in  
 4 inertia gas for 20 days<sup>42</sup>. Reproduced from Ref. 42 with permission, Copyright ©  
 5 2012, American Chemical Society.

6



7

8 **Figure 6** (a) Concept of a stitchable organic photovoltaic textile. (b) Photographs of  
 9 the textile-based OPV integrated with clothing<sup>45</sup>. Reproduced from Ref. 45 with  
 10 permission, Copyright © 2014 Elsevier Ltd.

11

### 12 1.3 Fiber Dye-Sensitized Solar Cells (FDSCs)

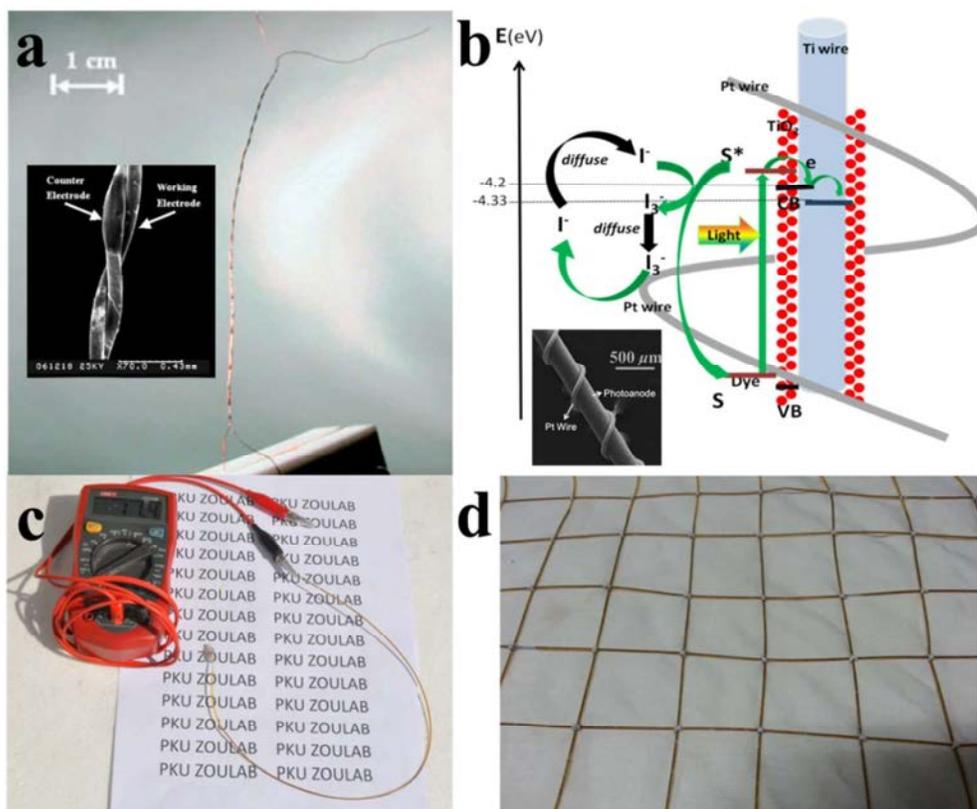
13 The tentative idea of FDSCs took shape at the beginning of the 21st century, which  
 14 made advantages of the simple, air-tolerant, and robust DSSC preparation<sup>48</sup> and  
 15 extended the typical sandwich device to lower dimensional structure<sup>20, 21</sup>. In 2008,  
 16 Zou et al. reported flexible wire-like DSSCs based on double twisted metal fibers  
 17 (**Figure 7a**)<sup>24</sup>. This design featured as linear shape, transparent conductive oxide  
 18 (TCO)-free and easy fabrication attracted wide attention<sup>49</sup>. Later on, singly twisted  
 19 FDSCs where the counter electrode (Pt wire) winding the fiber photoanode (**Figure**

1 **7b**) were prepared and the device with capillary package achieved efficiency up to  
2 7%<sup>25, 50</sup>. The above success led to follow-up FDSC explorations toward higher  
3 efficiency, cheaper materials, larger device size, better stability, concentrated designs  
4 and energy management, which included: 1) selection of commercial, inexpensive,  
5 lightweight fiber substrates for improved surface area and light harvesting; 2)  
6 synthesis of nano materials and interface engineering for efficient fiber photoanodes;  
7 3) development of cost-effective and highly active catalyst for counter electrodes; 4)  
8 low volatile electrolyte for improved device stability; 5) novel designs of fiber  
9 electrodes for 3D solar cells and concentrated devices; 6) integrated fiber energy  
10 system for multi-step energy conversion and storage.

11 All the trials and errors contributed to the continuous FDSC performance  
12 improvement, from < 0.5% to > 9% throughout the years<sup>51, 52</sup>. For example,  
13 well-designed device conformation, film preparation and interface engineering on  
14 fiber photoanodes improved dye-adsorption amount, enhanced light collection, better  
15 electron injection, and inhibited charge recombination, thereby achieving 7.2% to 8.6%  
16 efficiency<sup>53-55</sup>. Ultra-long FDSCs of more than 30 cm were fabricated with efficiency  
17 up to 4% (aspect ratio of 310), which are the longest single FDSCs with the highest  
18 power output and could be applied to solar fabrics (**Figure 7c** and **7d**)<sup>56, 57</sup>. For price  
19 considerations, replacing Pt-based counter electrode with cost-effective materials with  
20 high specific area is a sustainable way, achieving efficiencies of 7.2% to 8.5%<sup>58, 59</sup>. To  
21 take a further step toward lightweight and inexpensive power sources, non-metal wire  
22 substrates have been applied to either or both electrodes<sup>60-63</sup>. Especially, all-carbon

1 based FDSCs<sup>60</sup> and quantum dot (QDs) sensitized fiber solar cells<sup>64</sup> with moderate  
2 device performance were developed, and more breakthroughs are on the way. Typical  
3 volatile liquid electrolyte may prevent device stability in the long run, whereas gel  
4 and solid electrolytes can reasonably supply the solutions for stable FDSCs<sup>65-67</sup>.  
5 FDSC exploration was also expanded to innovations concerning light collection for  
6 high power output. Given the symmetric columnar structure, fiber photoanodes  
7 endowed FDSCs with capacities such as 3D light harvesting and less-independent  
8 incident light angles, which contributed to flexible multi-fiber solar device of 9.1%  
9 efficiency<sup>52</sup> and concentrating modules with fold-improved power output<sup>25, 68</sup>. Except  
10 for solar-to-electrical generation, highly performing FDSCs can be integrated with  
11 other fiber shaped energy conversion devices, such as supercapacitor, battery and  
12 nanogenerator, for hybrid energy systems<sup>26, 69, 70</sup>, thereby providing more flexible  
13 energy management.

14 From the view of energy conversion efficiency, the performance of FDSCs is very  
15 close to traditional planar DSSCs. The development of FDSCs could represent the  
16 directions of all fiber solar cells, namely, 1) new materials and interface engineering  
17 for continuously improved performance; 2) efficient fiber electrodes for structure  
18 designs; 3) advanced solar energy system based on fiber photovoltaics. All the  
19 attempts and progress will push the fiber photovoltaics to higher level, and make  
20 chance for practical solar power for portable/wearable electronics.



1  
 2 **Figure 7** (a) Flexible wire-like dye-sensitized solar cell based on twisted metal  
 3 electrodes<sup>24</sup>. Reproduced with permission from ref. 24, Copyright © 2008  
 4 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (b) Schematic of singly twisted  
 5 FDSCs where the counter electrode (Pt wire) winding the fiber photoanode with  
 6 working mechanism<sup>25</sup>. Reproduced from Ref. 25 with permission from the Royal  
 7 Society of Chemistry. (c) Ultra-long FDSC of more than 30 cm (with aspect ratio of  
 8 310) displays  $I_{sc}$  of 11.4mA under natural light conditions (13:50 on October 16,  
 9 2013); (d) Flexible fabric of flexible FDSCs on the cloth substrate<sup>56</sup>. Reproduced  
 10 from Ref. 56 with permission from the Royal Society of Chemistry.

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#### 12 1.4 Fiber Perovskite Solar Cells

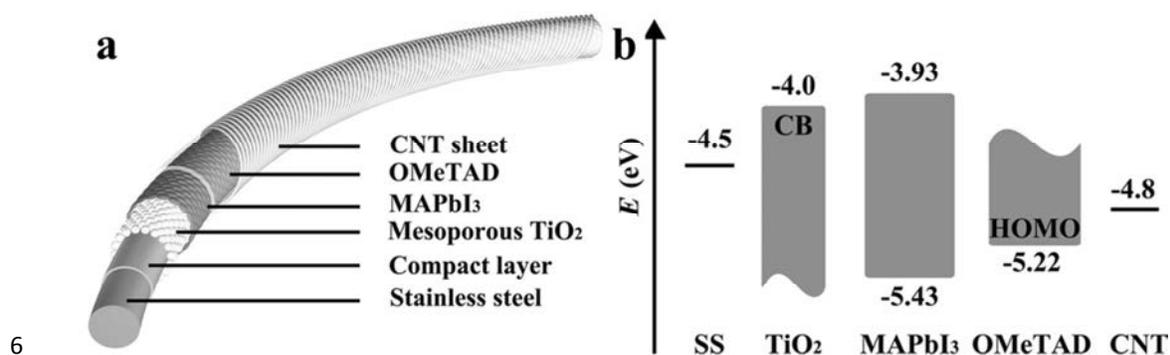
13 The new photovoltaic generation, perovskite solar cells<sup>71</sup> have attracted worldwide

1 attention, whose efficiency has been improved to  $> 20\%$  in five years<sup>3</sup>. Solid state,  
2 high performance and easy solution processing are the notable features of perovskite  
3 solar cells, which make them suitable candidates for efficient, flexible, lightweight,  
4 and liquid-free photovoltaic fibers. Peng's group<sup>28</sup> demonstrated perovskite fiber solar  
5 cells (**Figure 8**) by winding an aligned multi-walled carbon nanotube sheet onto a  
6 TiO<sub>2</sub>-modified steel wire (SS) and incorporating perovskite layer between them via a  
7 one-step solution process. The coaxial fiber devices achieved efficiency of 3.3% and  
8 the photovoltaic parameters ( $V_{oc}$ ,  $J_{sc}$ , and  $FF$ ) remained almost constant under  
9 different incident light angles. No damage was observed on the sheet electrode and  
10 the energy efficiencies remained at 95% after bending for 50 cycles. With two-step  
11 solution process, the corresponding perovskite fiber solar cells based on ZnO array  
12 achieved 3.8% efficiency<sup>72</sup>.

13 Although the efficiency is only comparable with fiber inorganic and organic solar  
14 cells at present, the device performance are rather promising as initial trials. The  
15 above trials further confirm the openness of the fiber system to new photovoltaic  
16 types. Due to the excellent performance of perovskite solar cells, fiber perovskite  
17 solar cells have a great chance of catching up via further modification, and they might  
18 achieve equal or even higher efficiency than FDSCs. For practical applications, more  
19 research is needed to explore repeatable and scalable technology. But the perovskite  
20 layer is sensitive to moisture and hole transport material Spiro-OMeTAD  
21 [2,2',7,7'-tetraakis(N,N-di-p-methoxy phenylamine)-9,9'-spirobifluorene], suffer  
22 from de-doping process in the air, the stability and lifetime of fiber device need



1 further improvement. Another drawback is the toxic elements, environmentally  
2 friendly alternatives are welcome. From this point of view, opportunities are there for  
3 novel photovoltaic types as well as fiber photovoltaics. In the ideal situation, reliable  
4 packages could help with the stability and pollution leakage problem at the same time.  
5 But some systemic work remains to be open before the scheme is acceptable.



6  
7 **Figure 8** (a) Structure and (b) energy level diagram of flexible perovskite fiber solar  
8 cells based on carbon nanotube sheet<sup>28</sup>. Reproduced from Ref. 28 with permission,  
9 Copyright © 2014 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

10  
11 As summarized in **Table 1**, fiber/wire-shaped solar cells, including inorganic,  
12 organic, dye sensitized, and perovskite, have made significant advancements. Fiber  
13 devices with length  $\leq 5$  cm were reported in most present literature. For high power  
14 output and weaving process, ultra-long, efficient and flexible fiber solar cells remain  
15 one of the most tempting challenges in this field. Compared with other fiber solar  
16 cells, FDSCs have achieved superior efficiency and larger device size. Moreover,  
17 FDSCs are easy and scalable to fabricate, as well as less sensitive to environmental  
18 factors. These advantages make them an attracting topic in material studies and

1 achieve rapid progress.

2

**Table 1 Photovoltaic parameters of typical fiber inorganic, organic, dye-sensitized and perovskite solar cells**

Structure	Feature	<i>L</i> (cm)	<i>J</i> <sub>sc</sub> (mA/cm <sup>2</sup> )	<i>V</i> <sub>oc</sub> (V)	<i>FF</i>	<i>η</i> (%)	Ref.
CF/p-poly Si/n-poly Si/Ag	Hairlike single fiber	4	1.7	0.14	0.246	0.04	29
Capillary fiber/p-Si/i-Si/n-Si/Ti-Au	coaxial silicon fiber with p-i-n junction	1	2	0.22	0.55	0.5	30
Al-Pt/p-Si/i-Si/n-Si/ITO	silicon-core single fiber	<1	16.6	0.3	0.52	3.6	31
Mo/CuInSe <sub>2</sub> /CdS/ZnO/ITO	Non-Si flexible fiber	4	13.0	0.34	0.522	2.31	27
Optical fiber/ITO/PEDOT:PSS/P3HT:PCBM/LiF/Al	Waveguide light-collection	~1.4	~7.0	/	/	1.1	32
SS/TiO <sub>x</sub> /P3HT:PCBM/PEDOT:PSS/LiF/Ag/SS	ITO-free; twisted fibers; 5-30 cm	/	11.9	0.60 7	0.538	3.87	39
Ti/TiO <sub>2</sub> /P3HT:PCBM/PEDOT:PSS/CNT fiber	flexible and stable	/	9.06	0.52	0.38	1.78	40
SS/ZnO/P3HT:PCBM/PEDOT:PSS/CNT yarn	CNT yarn; stable	/	8.49	>0.5 6	/	2.3	42
SS/ZnO/P3HT:PCBM/PEDOT:PSS/graphene	Single-layer graphene sheets	/	8.14	0.57 0	0.545	2.53	43
Ti/TiO <sub>2</sub> /N719/I <sup>-</sup> <sub>3</sub> /Pt	TiO <sub>2</sub> colloids; 3D light-harvesting	4.8	12.28	0.72 8	0.786	7.02	25
Ti/TMCA-TiO <sub>2</sub> /N719/I <sup>-</sup> <sub>3</sub> /Pt	Micron-core array; multilayer structure	5-7	16.04	0.70 2	0.717	8.07	54
Ti/TiO <sub>2</sub> /N719/I <sup>-</sup> <sub>3</sub> /graphene-Pt	Graphene/Pt composite fiber	/	/	0.74 4	0.63	8.45	58
Ti/TiO <sub>2</sub> /N719/I <sup>-</sup> <sub>3</sub> /TiN/carbon fiber	Helical photoanode; Non-Pt	/	19.35	0.64	0.58	7.20	59
Ti/TiO <sub>2</sub> nanotube/N719/I <sup>-</sup> <sub>3</sub> /Pt	Flexible; ordered TiO <sub>2</sub> nanotube arrays	/	15.9	0.67 3	0.80	8.6	55
(Ti/TiO <sub>2</sub> /N719) <sub>6</sub> /I <sup>-</sup> <sub>3</sub> /Pt	Multi-fiber anodes	1.7	18.6	0.66	0.74	9.1	52
	Facile synthesis	5	/	/	/	Avg. 7.2	53
Ti/bilayer-TiO <sub>2</sub> /N719/I <sup>-</sup> <sub>3</sub> /Pt	Longest single device; highest power output	9.0	12.58	0.68 3	0.712	6.12	56
		31.5	8.09	0.71 6	0.692	4.01	56
CNT yarn /TiO <sub>2</sub> /N719/I <sup>-</sup> <sub>3</sub> /CNT yarn-Pt	Flexible; all carbon device	/	19.8	0.54	0.319	3.4	73

Optical fiber/ITO/TiO <sub>2</sub> NW/N719/I <sup>-</sup> I <sub>3</sub> <sup>-</sup> /Pt	Waveguide; cylindrical electrode	2-3	18.50	~0.6	/	6	74
Ti/TiO <sub>2</sub> /N719/T <sup>-</sup> -T <sub>2</sub> /CNT fiber	Flexible; non-I <sub>2</sub> electrolyte	/	15.49	0.68 3	0.69	7.33	75
SS/TiO <sub>2</sub> /CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> / Spiro-OMeTAD/CNT	Flexible coaxial perovskite solar fiber	<4	10.2	0.66 4	0.487	3.3	28
SS/ZnO/CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> / Spiro-OMeTAD/CNT	ZnO nano-obelisk array; mild condition	<5	~12	~0.6 5	/	3.8	72

1

## 2 2 Properties of Fiber Solar Cells

3 Evolved from sandwich-type structure, the three dimensional structure endows  
4 fiber solar devices with special feature in light harvesting and charge transport, which  
5 further affects characterizations and applications. In principle, fiber solar cells with  
6 column symmetry should share similar properties. Since extensive studies of FDSCs  
7 have been well developed, we present the properties of fiber solar cells with FDSCs  
8 as a typical representative. In addition, compared with solid fiber solar cells, the  
9 liquid-based interfaces could relate to the high performance of FDSCs. Thus, we  
10 discuss the properties of fiber solar cells with focus on FDSCs in this part.

11

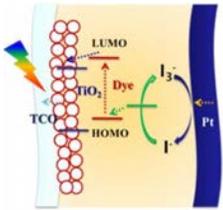
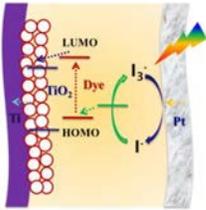
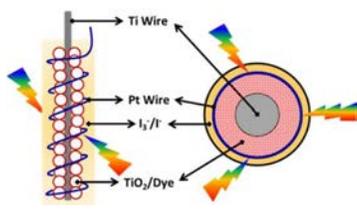
### 12 2.1 From Planar Structure to Fiber Style

13 FDSCs differ with typical and inverted DSSCs in composition, shape, flexibility,  
14 light collection, electron injection, and electrodes (**Table 2**). With  
15 Ti/TiO<sub>2</sub>/Dye/electrolyte/Pt composition, FDSCs do not need TCO-based substrates,  
16 which are important in the total traditional DSSC cost. Different from traditional  
17 planar flat appearance, FDSC devices are in the form of wires, fibers, rods, or tubes;  
18 and flexible devices can be achieved by increasing the length/diameter ratio together

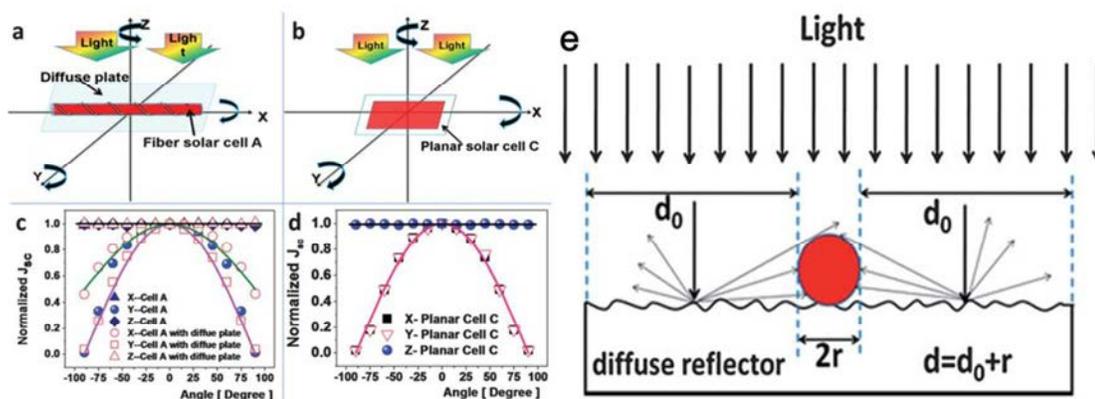
1 with soft packages. These FDSC properties indicate a promising role in  
2 portable/wearable applications, which can be woven into flexible textiles. Since the  
3 utilization of high-melting-point fibers rather than polymer conductive substrates (e.g.,  
4 ITO/PET, ITO/PEN)<sup>76, 77</sup>, no temperature limitation exists during functional fiber  
5 preparation. Furthermore, given the symmetric cylindrical structure, FDSCs can  
6 harvest solar radiation in 3D space, more freedom than the front or back illumination  
7 (**Figure 9a to 9d**)<sup>25</sup>. Experiments of various fiber solar cells have shown constant  
8 performance with different incident light angles<sup>24, 28, 42</sup>. Therefore, a simple diffusing  
9 plate under FDSC can improve light harvesting for higher power output via diffuse  
10 reflection illumination (**Figure 9e**). For example, power output could be greatly  
11 enhanced for fiber solar cells in conjunction with conventional rough substrates (e.g.,  
12 foam board and white paper) and mirrors via light scattering and reflecting<sup>50, 56, 78</sup>.  
13 Nonetheless, reduced incident light in inverted DSSCs and FDSCs by electrolyte  
14 absorbance was observed compared with the light path of the front illuminated style.  
15 Reducing electrolyte thickness can lead to better solar energy harvesting. Under  
16 illumination, the photoelectrons from the excited dyes were injected into the  
17 conductive band of semiconductor layer and further collected through the current  
18 collector. The direction of electron injection in FDSCs and inverted DSSCs is along  
19 the incident direction, which is opposite to that of typical DSSCs. Furthermore, unlike  
20 DSSC and inverted DSSC cases, no explicit area/size ratio for the two electrodes is  
21 required, and they can be twisted together, packaged in parallel or film covered single  
22 fibers, which provide much freedom and convenience for structural design and

1 material innovations. For fiber devices, the contact between electrolyte and sealing  
 2 material is at two ends; smaller than that of the planar devices, which could reduce  
 3 leakage rate. All these differences can essentially reflect the structure evolution from  
 4 DSSCs to inverted DSSCs and further to FDSCs. And similar phenomena could exist  
 5 in other types of fiber solar cells.  
 6

**Table 2 Comparison of typical DSSCs, inverted DSSCs, and FDSCs**

Type	DSSCs	Inverted DSSCs	FDSCs
<b>Diagram</b>			
<b>Composition</b>	TCO/TiO <sub>2</sub> /Dye/electrolyte/Pt/TCO	Ti/TiO <sub>2</sub> /Dye/electrolyte/Pt/TCO	Ti/TiO <sub>2</sub> /Dye/electrolyte/Pt
<b>Shape</b>	Flat	Flat	Wire/fiber/rod/tube
<b>Flexibility</b>	TCO substrate; area/thickness ratio	flexible substrate; area/thickness ratio	Soft package; length/diameter ratio
<b>Light</b>	front illuminated;	back illuminated; TCO/Pt	3D light harvesting;
<b>Collection</b>	TCO → TiO <sub>2</sub> /Dye	→ electrolyte → TiO <sub>2</sub> /Dye	Tube → electrolyte → TiO <sub>2</sub> /Dye
<b>Electron Injection</b>	Dye → TiO <sub>2</sub> → TCO	Dye → TiO <sub>2</sub> → Ti	Dye → TiO <sub>2</sub> → Ti

Anode/Cathode=1/1;      Anode/Cathode=1/1;      No explicit ratio; twisted, in  
**Electrodes**                      face-to-face                      face-to-face                      apparel or single fiber



1  
 2 **Figure 9** (a) 3D light-collecting schematic of the fiber-shaped solar cell (Cell A); (b)  
 3 3D light-collecting schematic of the traditional plate solar cell (Cell C); (c) the angle  
 4 dependences of the normalized short-circuit currents of the Cell A and those of the  
 5 composite Cell A; (d) the angle dependences of the normalized short-circuit currents  
 6 of the plate Cell C. (e) Schematic of the diffuse reflection illumination mode<sup>25</sup>.  
 7 Reproduced from Ref. 25 with permission from the Royal Society of Chemistry.

8

9 For flexible and wearable energy devices, the cohesion and adhesion of active  
 10 layers would be crucial factors during textile manufacturing and weaving operations,  
 11 as well as in maintaining device performance. Therefore, bulk fiber electrode with  
 12 good conductivity and catalytic performance, like Pt wire, would be a good choice for  
 13 the counter electrode. The dye-sensitized semiconductor oxides of the photoanode can  
 14 form cracks upon deformation. Experiments have shown that damage induced by  
 15 tensile deformation influence the device efficiency more than local damage, which  
 16 indicates the practicability of preparing solar textiles from fiber devices, if the tension

1 is well controlled<sup>79</sup>. Typically, thinner layer with good adhesion is preferable to  
2 prevent deep cracks, whereas thicker functional layer is preferable for higher  
3 efficiency<sup>50, 53, 80</sup>. A balance between flexibility and efficiency need further  
4 consideration of practical factors.

5

## 6 **2.2 Characterization of Fiber Solar Cells**

7 For typical sandwich-type solar cells, the illumination area is the apparent active  
8 area or masked geometrical area (**Figure 10a**). However, for fiber solar cells, only  
9 part of the photoanode is illuminated and the surface area of curved fiber is not the  
10 reasonable illumination area. According to the projection relation, the cross sectional  
11 area of the fiber photoanode is the position for direct solar energy harvesting (**Figure**  
12 **10b**). Thus, in most recent works<sup>50, 73, 81</sup>, the project area was used for efficiency  
13 calculation. The illumination area ( $A$ ) is calculated as:

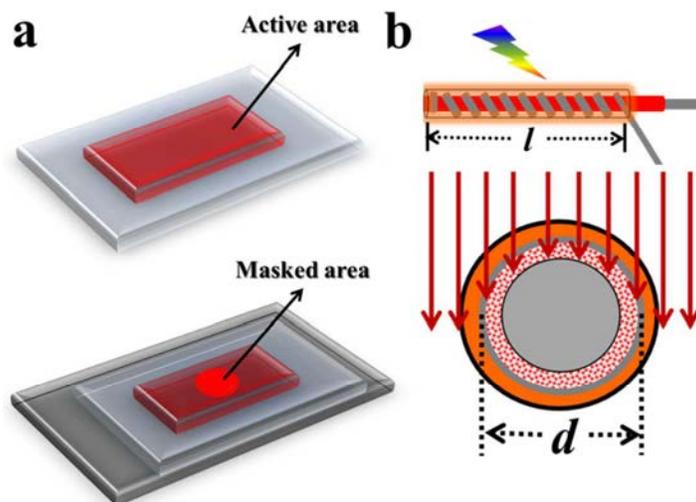
$$14 \quad A = d * l$$

15 where  $d$  and  $l$  is the average diameter and effective length of fiber photoanode,  
16 respectively.

17 The short-circuit current density ( $J_{sc}$ ) and the solar energy conversion efficiency ( $\eta$ )  
18 is calculated by:

$$19 \quad J_{sc} = I_{sc}/A \text{ and } \eta = J_{sc} \times V_{oc} \times FF \times 100\%$$

20 Where  $I_{sc}$ ,  $V_{oc}$ , and  $FF$  is the short-circuit current, open-circuit voltage, and fill  
21 factor, respectively.



1

2 **Figure 10** (a) The illumination area of typical sandwich-type solar cells is the  
 3 apparent active area or masked geometrical area; (b) According to the projection  
 4 relation, the cross sectional area of the fiber photoanode is the position for direct solar  
 5 energy harvesting.

6

7 Photoanode with a smaller fiber diameter would lead to higher efficiency due to  
 8 smaller active area under the same length<sup>82</sup>, which indicates fiber substrates of  
 9 micro/nanoscale might contribute to efficient devices. Notably, the size measurement  
 10 (length and average diameter) error in short devices is larger than that in larger  
 11 devices. Thus, device size should be considered in terms of measurement accuracy, as  
 12 well as preparation difficulty, during efficiency comparison. For practical application  
 13 and a more reasonable comparison with conventional DSSCs, the cross sectional area  
 14 of the capillary tube is recommended to represent the cross section of the incident  
 15 light<sup>52, 82</sup>. Reducing tube thickness and increasing its effective partial filling would  
 16 benefit high-performance devices. Additionally, testing environment should be



1 emphasized because of the 3D light harvesting property. Given the discrepancy of  
2 different voltage scanning directions, the average of the forward and backward tests is  
3 recommended as the final result for FDSCs<sup>56</sup>.

4 Current literatures have dealt with the flexibility of fiber solar cells, but there is no  
5 uniform parameter as standard. The aspect ratio (the ratio of length and diameter),  
6 photovoltaic performance under various bending conditions (e.g. bending angles,  
7 bending radius, bending cycles), morphology after bending cycle(s), were typically  
8 experimental evidence for discussions.

9

### 10 **2.3 Reasons for excellence of liquid-based FDSCs**

11 To achieve efficient fiber solar devices, the key points are uniform functional layers  
12 and effective interface contact for efficient light collection, photoelectron generation,  
13 and charge transport. These points might be easier for traditional flat substrates, in  
14 which efficient solar cells have been fabricated. However, fiber substrates, which  
15 exhibit cylindrical surface and some uneven positions formed in the drawing process,  
16 may cause coating defects, layer fractures, and film perforation. These factors may  
17 exert significant adverse effects on thin film devices, such as invalid p-n junction,  
18 series charge recombination, or even short circuit. These problems become more  
19 pronounced in devices of large length and that is the reason why we emphasize device  
20 size in energy conversion efficiency comparison. Fortunately, a semiconductor oxide  
21 layer of more than 10 microns was enough to fill these grievances and cover the  
22 whole substrate in FDSC. Additionally, light collection can be weakened when the

1 active layer is covered by a hole transport layer and/or outer metal current collector,  
2 which may limit photoelectron generation. Liquid electrolyte and twisted electrodes  
3 can mitigate this effect to a certain degree. Moreover, considering the interface  
4 contact, charge transfer at the solid-liquid heterojunction (photoanode/electrolyte  
5 interface) might be more efficient than that at solid-solid heterojunction. These are the  
6 possible reasons why FDSCs are currently more efficient than most other fiber solar  
7 cells. However, the possibility that real efficient solid-solid heterojunction would be  
8 discovered as evidence of technology development cannot be ruled out. In fact, higher  
9 current density short-circuit current ( $J_{sc}$ ) or/and open-circuit voltage ( $V_{oc}$ ) can be  
10 expected in other solar cell types, even with lower fill factor ( $FF$ ) than liquid-based  
11 FDSCs. We are quite optimistic that highly efficient solid fiber solar cells will be  
12 introduced in the near future.

13

### 14 **3 Materials for FDSCs**

#### 15 **3.1 Substrates**

16 The substrate, which provides support to the whole device, is an important part of  
17 FDSCs. Mechanical strength, conductivity and specific surface area comprise the  
18 basic factors for fiber substrates. For flexible fiber devices, the aspect ratio of fiber  
19 electrodes determines FDSC flexibility. Therefore, flexible fiber substrates with  
20 smaller diameter and larger length would be ideal candidates for practical applications.  
21 Another challenge is that the weight and cost of fiber substrate should be as low as  
22 possible, and this is indeed a consideration for wearable applications. Conductive

1 metal fibers, carbon-based fibers, polymer fibers, and optical fibers have been applied  
2 to FDSCs.

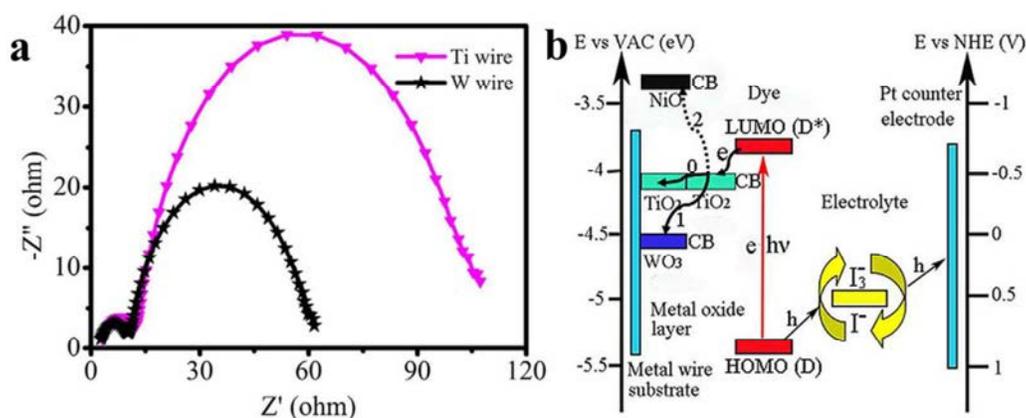
3

### 4 **3.1.1 Metal Wires**

5 Metal fibers are the most commonly used substrate for FDSCs owing to their good  
6 conductivity, mechanical strength, long-range flexibility, and high-temperature  
7 tolerance. Various metal wires<sup>57, 82-84</sup> including titanium, stainless steel, wolfram,  
8 aluminum, nickel, and zinc, have been utilized to collect photocurrent in the  
9 photoanode, as well as to support the whole device. Though started with stainless  
10 steel wire<sup>24</sup>, most of the reported FDSCs and the most efficient FDSCs (7% to 9%  
11 efficiency) are fabricated with Ti-based photoanodes<sup>52-55</sup>. Annealing process of  
12 titanium wires can decrease the work function and increase Fermi level, which  
13 contributes to lower Schottky barrier<sup>57</sup>. However, W oxides formed during annealing  
14 leads to lower recombination resistance at the photoanode/electrolyte interface and  
15 results in lower open circuit voltage; the insulating Al<sub>2</sub>O<sub>3</sub> and NiO layers, which could  
16 prevent the photoelectrons from being injected into the substrate (**Figure 11**), lead to  
17 poor efficiency<sup>82, 83</sup>. However, the above observations cannot exclude cheap metal  
18 substrate application for DSSCs because low-temperature-based process can avoid  
19 metal oxidation. However, active metals exposed to electrolyte may cause  
20 galvanic-battery reactions along with metal corrosion and oxidized species  
21 consumption, which eventually result in a non-functional device<sup>85</sup>. Therefore,  
22 covering the metal substrates via dense titanium oxide layer is a possible solution<sup>57</sup>.

1 Moreover, cheap and light fiber substrates with titanium-coated layer may also play a  
2 role in high-purity titanium wire placement.

3 Furthermore, metal wires support the active catalytic components as counter  
4 electrodes in parallel with the photoanode<sup>69, 86, 87</sup>. Given the good conductivity,  
5 studying the electrochemical catalytic performance of new materials may be  
6 beneficial regardless of the impact of resistance.



7  
8 **Figure 11 (a)** EIS measurements for the FDSSCs based on W and Ti wires with TiO<sub>2</sub>  
9 photoanodes. The W based FDSCs showed a drastic decrease in the internal resistance,  
10 which means a larger recombination rate in photoelectrode, and which might be due  
11 to the high-level conduction band mismatch between the TiO<sub>2</sub> working electrode and  
12 the WO<sub>3</sub> layer. (b) Energy states diagram for different metal wire-based FDSC  
13 featuring the operation principle, and the dashed line shows the photoelectrons cannot  
14 transfer from TiO<sub>2</sub> nanoarray to NiO film<sup>83</sup>. Reproduced from Ref. 83 with  
15 permission, Copyright © 2014, Rights Managed by Nature Publishing Group.

16

### 17 3.1.2 Carbon-based Fibers

18 Carbon-based fibers are attractive candidates as electrodes for energy devices

1 because of their salient flexibility, low density, high specific surface area, moderate  
2 conductivity, anti-corrosion property, and high strength<sup>49, 88, 89</sup>. Carbon fibers  
3 composed of micron-sized single fibers are commercial products and are utilized in  
4 FDSCs since 2011<sup>90</sup>. With even higher specific surface area, carbon nanotube (CNT)  
5 fibers based on nanosized monofilament were also applied in fiber solar cells by  
6 Peng's<sup>91</sup> and Cao's<sup>92</sup> groups. Core-sheath CNT/reduced graphene oxide (RGO)  
7 nanoribbon, CNT/RGO composite, and RGO fibers<sup>93</sup> (**Figure 12a**) benefit from the  
8 convenience of physical and/or chemical modifications, which bring more concept for  
9 materials science and device designs.

10 Carbon-based fibers can be used as supporters for dye-sensitized TiO<sub>2</sub> layer, such  
11 as nanoparticles, nanorods (NRs), nanotube, and nanowires (NW)<sup>60, 94, 95</sup>. Wang et al.  
12 first demonstrated CF/TiO<sub>2</sub> NR array-based FDSC (**Figure 12b**) with efficiency of  
13 0.76%; bunched-NR based device achieved higher efficiency of 1.28% because of  
14 higher *J*<sub>sc</sub> attributed to the larger surface area<sup>94</sup>. FDSC with CF/TiO<sub>2</sub> NW photoanode  
15 obtained an impressive conversion efficiency of 2.48%<sup>95</sup>. The carbon fiber bundle  
16 diameter directly affects the device performance originating from the electrode  
17 conductivity, loading amount of dye-sensitized nano-TiO<sub>2</sub>, and apparent device area<sup>60</sup>.  
18 Semiconductive CNT fibers were directly sensitized as photoanode, and the device  
19 packaged in the typical sandwich format achieved power conversion efficiency of  
20 2.2%<sup>91</sup>. Nonetheless, these FDSCs are usually not self-standing, and additional  
21 substrate is required to support the device, which can be a problem for carbon-based  
22 fiber photoanode. Moreover, obtaining the accurate active area is not easy due to the

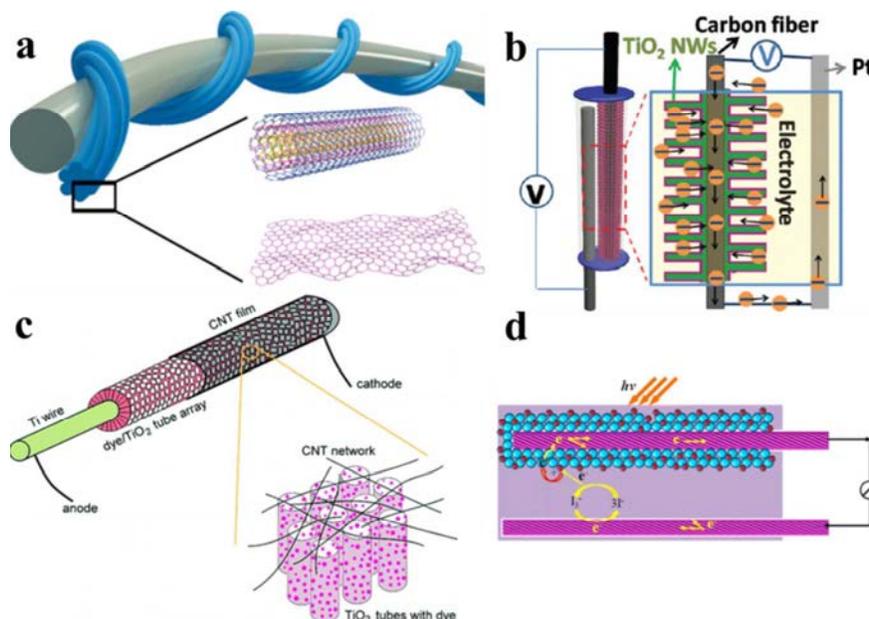
1 disperse state in the electrolyte, and the apparent area in the dry state is used for  
2 efficiency calculation. However, these problems can be avoided when these fibers  
3 were used in the counter electrode.

4 Commercial carbon fibers consisting of single fibers were used as counter electrode  
5 or current collector in some innovative works, with efficiencies of 3.0% to 7.2%<sup>59, 90,</sup>  
6<sup>96, 97</sup>. A bunch of carbon fibers showed limited catalytic activity toward  $I_3^-/I^-$  redox  
7 couple, with 2.7% efficiency<sup>25</sup>. The structural defects on the nanotubes formed main  
8 catalytic sites for redox reactions by directly utilizing CNT fiber as the counter  
9 electrode, and 3.4% efficiency was reported<sup>98</sup>. The device performance showed some  
10 yarn diameter dependence because of the combination of active sites, conductivity,  
11 and electrode contact. With similar device structure, Peng et al. reported device  
12 efficiency up to 4.6% with CNT fiber<sup>99</sup> and 3.8% with graphene fibers<sup>58</sup>. CNTs can be  
13 rolled or twisted to fibers since they are usually grown as a thin film on planar  
14 substrate. To make advantage of the CNT thin films for device design, Cao et al.<sup>92</sup>  
15 reported single-wire DSSC (1.6%) via wrapping carbon nanotube film around the  
16 fiber photoanode (**Figure 12c**). Efficiency of 2.6% was achieved with the assistance  
17 of an Ag wire to overcome the CNT sheet resistance. Through film thickness  
18 optimization, similar coaxial structure offers a high energy conversion efficiency of  
19 4.1% and stability during deformation<sup>100</sup>. These “CNT-enriched fiber solar cells” are  
20 attracting increasing attention given the attractive structures, moderate performance,  
21 and light weight<sup>101-104</sup>.

22 A more challenge to single carbon-based electrode is the all-carbon based FDSCs

1 reported from several groups, which employed carbon-based fibers as substrates of  
2 both electrodes (**Figure 12d**), thereby achieving 1.9% to 3.4% efficiency<sup>60, 62, 73, 95, 105</sup>.  
3 Even with non-liquid electrolyte, the all-carbon-based photovoltaic fibers  
4 photoconversion efficiency of 2.6% was achieved with all-carbon-based photovoltaic  
5 fibers, which is independent of incident illumination and cell shape or position<sup>61, 106</sup>.  
6 Despite the low efficiency, these works may provide a promising avenue toward  
7 lightweight and flexible solar cells.

8 The above results indicate that carbon-based fibers are promising materials for  
9 efficient FDSCs. However, the carbon-based fiber only is not enough for efficient  
10 charge exchange at the cathode/electrolyte interface, which resulted in relatively low  
11  $J_{sc}$  and  $FF$ . Thus, catalysts should be employed to avoid this restriction. In most cases,  
12 carbon-based fibers were used as supporting substrate because of their limited  
13 catalytic property but high specific surface area for additional chemical modifications.  
14 Various kinds of catalytic materials are discussed in Part 3.3.



15

1 **Figure 12** (a) FDSCs based on CNT and RGO fibers<sup>93</sup>. Reproduced from Ref. 93 with  
2 permission, Copyright © 2014, American Chemical Society. (b) Schematic of  
3 configuration of the CF/TiO<sub>2</sub> NR array-based tube-shaped DSSC<sup>94</sup>. Reproduced from  
4 Ref. 94 with permission, Copyright © 2012, American Chemical Society. (c) Flexible  
5 single-wire DSSCs by wrapping a carbon nanotube film around Ti wire-supported  
6 TiO<sub>2</sub> tube arrays as the transparent electrode<sup>92</sup>. Reproduced from Ref. 92 with  
7 permission, Copyright © 2011, American Chemical Society. (d) All-carbon-based  
8 FDSC with highly flexible aligned carbon nanotube fibers as catalytic counter  
9 electrode and current photoanode collector<sup>105</sup>. Reproduced from Ref. 105 with  
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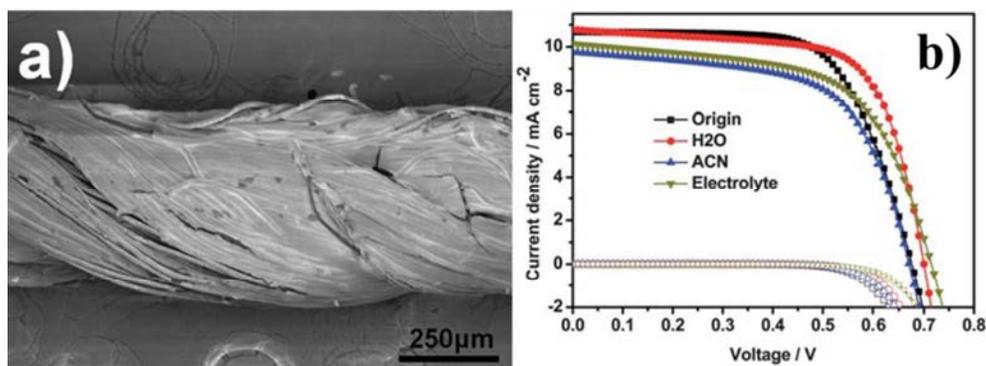
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### 12 **3.1.3 Polymer Fibers**

13 Polymer fibers are flexible, lightweight, low-cost and commercially available  
14 materials, but not naturally conductive. The first challenge in practical  
15 electrochemical cell application is preparing conductive fiber substrates. Zou et al.<sup>4</sup>  
16 prepared flexible conductive threads on insulating thread substrates via dip-coating  
17 conductive PEDOT:PSS dispersion. The conductive threads showed good resistance  
18 to the solvents and achieved 4-5% efficiency as counter electrode in FDSCs (**Figure**  
19 **13**). Fan et al.<sup>107</sup> reported Ni-deposited polymer materials as photoanode substrates  
20 for ZnO-based DSSCs, such as polybutylene terephthalate, polypropylene,  
21 polyethylene terephthalate, polyamide, and bio-mimetic veins. Lightweight DSSCs in  
22 the shape of wires, nets, and veins were fabricated via wet process.



1 Polymer fibers are raw materials for cloths, hats, and adornments. Polymer  
2 fiber-based energy devices may play a role in wearable electronics, which is one of  
3 the most important research motivations. Nevertheless, more efforts should be paid  
4 for better device performance and practical module design.



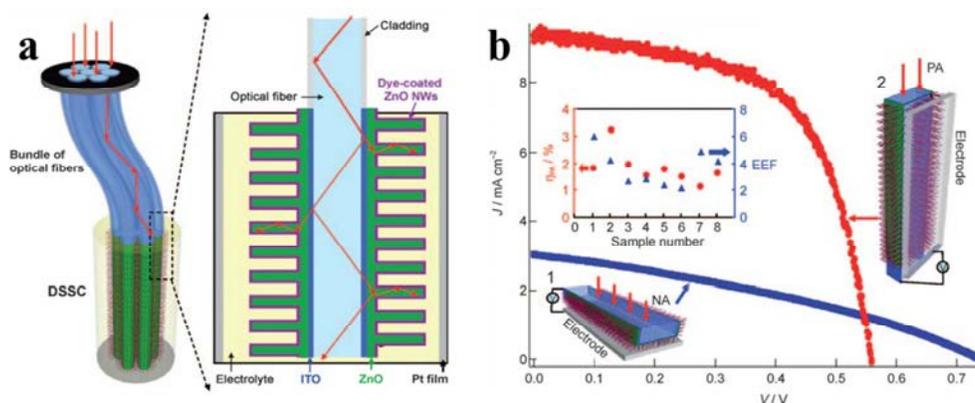
5  
6 **Figure 13** (a) Morphology of the conductive thread based on insulating thread  
7 substrates and PEDOT:PSS dispersion. (b)  $J$ - $V$  curves of the fiber-shaped solar cells  
8 using conductive thread electrodes soaked in water, acetonitrile, and electrolyte,  
9 which corresponds to efficiencies of 4-5%<sup>108</sup>. Reproduced from Ref. 107 with  
10 permission from the Royal Society of Chemistry.

11

### 12 3.1.4 Optical Fibers

13 In 2009, Ferenets et al. designed a fiber solar device with optical  
14 fiber/ZnO:Al/TiO<sub>2</sub>/N719/electrolyte/C, but its light harvesting was extremely  
15 hampered by the outmost carbon catalytic layer<sup>109</sup>. In the same year, Wang et al.<sup>63</sup>  
16 developed a hybrid photoanode with optical fiber/ITO/ZnO NW arrays for 3D DSSCs  
17 (**Figure 14**). The internal light reflection along the fiber expanded the opportunities  
18 for solar energy conversion, and the waveguide was extracted to the ITO/ZnO/Dye

1 interface because of its higher refractive index. These FDSCs achieved an efficiency  
 2 of 3.3%, and a corresponding efficiency enhancement factor (EEF) of 4.3 has been  
 3 achieved by converting the 2D DSSC to the 3D DSSC. Additionally, by integrating  
 4 optical fiber/ITO/TiO<sub>2</sub> NW hybrid fiber electrodes with the cylindrical counter  
 5 electrode, an impressive efficiency of up to 6% has been demonstrated with EEF up to  
 6 9<sup>74</sup>.



7  
 8 **Figure 14** (a) A 3D DSSC was constructed based on ZnO NWs vertically grown on  
 9 an optical fiber. The internal light reflection along the fiber was guided to the  
 10 photoanode. (b)  $J$ - $V$  curves of a DSSC under one full-sun illumination oriented 1)  
 11 normal to the fiber axis (NA, 2D) and 2) parallel to the fiber axis (PA, 3D). A  
 12 corresponding EEF of 4.3 has been achieved by converting the 2D DSSC to the 3D  
 13 DSSC. The inset shows a plot of EEF and the corresponding energy conversion  
 14 efficiencies for eight 3D DSSCs<sup>63</sup>. Reproduced from Ref. 63 with permission,  
 15 Copyright © 2009 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

16

17 Given the special light harvesting characteristic via modified optical fibers, the  
 18 direction of light reflected to the sensitized layer and photoelectron guided to the

1 current-collecting electrode is similar to the cases in traditional planar DSSCs. Thus,  
2 ITO is involved as the transparent conductive layer on the fiber substrate. Moreover,  
3 the counter electrode can contribute to additional light reflection for solar energy  
4 harvesting especially in closed structures. Theoretical investigations have shown that  
5 DSSCs wrapped around an optical fiber can achieve as promising performance as  
6 standard geometry<sup>110-112</sup>. Some flexible or concentrating devices based on 3D solar  
7 cells may be expected in the future.

8 As previously mentioned, DSSCs can be assembled on various fiber substrates  
9 with quite high performance. With flexible FDSCs, photovoltaic braid can be woven  
10 as a network of single-fiber devices<sup>56</sup>. DSSCs with TCO-free flexible mesh substrate  
11 can also be treated as an extension to fiber-based photovoltaic devices<sup>113-120</sup>, which  
12 are woven first and then applied as a device. Both approaches, which are similar in  
13 appearance, can play a role in future wearable electronics, but sealing small tubes  
14 with two ends is significantly simpler than flat plates<sup>121</sup>. Thus, apart from directly  
15 weaving the device, FDSCs with “down to the wire”<sup>49</sup> technology can provide  
16 opportunities for 3D devices, light-harvesting designs, and integrating innovations.

17

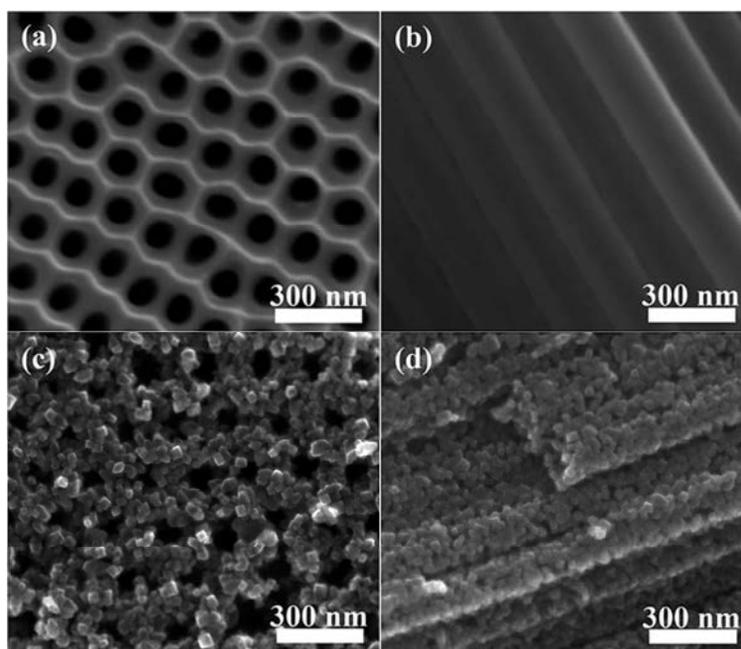
### 18 **3.2 Semiconductor Materials for Photoanodes**

19 TiO<sub>2</sub> nanoparticle with high specific surface area and good dye adsorption has been  
20 the basic material for porous fiber photoanode in early trials. Its suspension is a good  
21 candidate for dip-coating fiber substrates, with device efficiency of up to 7%<sup>25, 50</sup>.  
22 Considering the colloid preparation, in situ preparation of TiO<sub>2</sub> nanostructures on

1 substrates is more convenient for device fabrications.

2       Electrons are more mobile by several orders of magnitude in 1D nanomaterials than  
3 in traditional nanoparticles, which could contribute to efficient FDSCs.  
4 Electrochemical anodizing of Ti wire substrates can lead to Ti-supported TiO<sub>2</sub>  
5 nanotube arrays, and the corresponding FDSCs achieved an efficiency of up to 7%<sup>80</sup>,  
6 <sup>122</sup>. Electrolysis can be performed in large-scale preparation, and this process allows  
7 the fabrication of totally flexible devices. Surface treatment of TiO<sub>2</sub> nanotube arrays  
8 in niobium isopropoxide solution can weaken the electron recombination process and  
9 improve Voc and FF<sup>123</sup>. Transferring TiO<sub>2</sub> nanotubes to ordered hierarchical  
10 nanoparticles with NH<sub>4</sub>F solution may result in an efficiency of 8.6% as demonstrated  
11 in a flexible FDSC with 30% improvement from the untreated group (**Figure 15**)<sup>55</sup>.

12 Zhou et al.<sup>124</sup> synthesized TiO<sub>2</sub> nanowires array on Ti wire via Na<sub>2</sub>Ti<sub>2</sub>O<sub>5</sub>•3H<sub>2</sub>O NW  
13 transformation in acid. For FDSCs, a high efficiency of 5.4% was achieved because of  
14 the high Jsc. To simplify the preparation process, direct treatment of Na<sub>2</sub>Ti<sub>3</sub>O<sub>2</sub>  
15 network with TiCl<sub>4</sub> solution simultaneously exchanged Na<sup>+</sup> and formed a hierarchical  
16 structure<sup>125</sup>. Moreover, rutile TiO<sub>2</sub> NW or NR arrays were prepared on fiber substrates,  
17 but lower efficiency was achieved partly because of the suboptimal crystalline  
18 structure<sup>94, 95</sup>. After in situ preparations, an interstice between the Ti substrate and the  
19 oxidized layer is commonly observed. TiCl<sub>4</sub> post-treatment is advantageous in  
20 reducing the aforementioned defects and to the photoanode/electrolyte interface<sup>80, 126</sup>.



1

2 **Figure 15** TiO<sub>2</sub> with different nanostructures for FDSCs: SEM images of a smooth3 TiO<sub>2</sub> nanotube array (a and b) and a hierarchical TiO<sub>2</sub> nanotube array (c and d)<sup>55</sup>.

4 Reproduced from Ref. 55 with permission from the Royal Society of Chemistry.

5

6 For better adhesion between the porous layer and the substrate, Zou et al.<sup>53</sup> reported

7 facial synthesis via direct sintering of titanium tetraisopropoxide on titanium wire.

8 FDSCs based on the compact layer exhibited an efficiency of 5.3%, and the high *FFs*

9 indicated the good charge transport caused by the massive structure. To increase dye

10 adsorption, the TiO<sub>2</sub> nanoparticle layer was dip-coated, and the FDSCs based on the

11 bilayer structure achieved average efficiency of 7.2%. Compared with the device

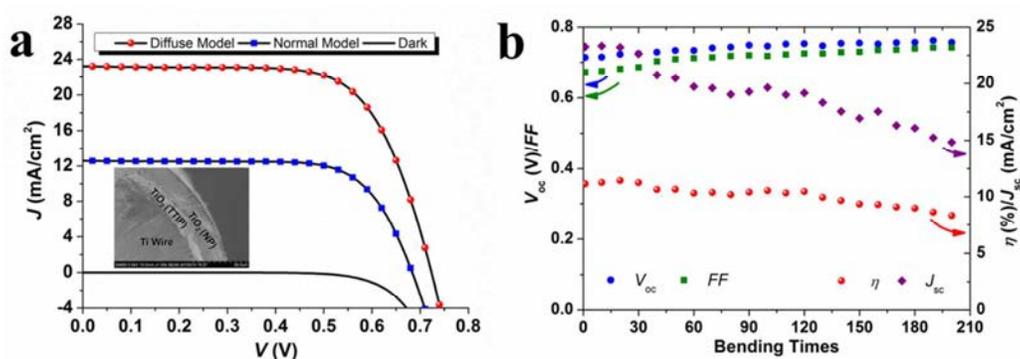
12 based on single nanoparticle layer, longer electron lifetime was obtained as a result of

13 the reduced interface charge recombination. With the TiO<sub>2</sub> bilayer, 9-cm long flexible

14 FDSC achieved an efficiency of 6.1% under AM 1.5 illumination and 11.4% with the

15 assistance of a piece of white paper, and 80% of the efficiency was maintained after

1 bending 180 times (**Figure 16**)<sup>56</sup>. Furthermore, for FDSCs, SnO<sub>2</sub>-TiO<sub>2</sub> bilayer and  
 2 trilayer structures were constructed via thermal calcination<sup>127</sup>. The  $J_{sc}$  enhancement  
 3 derived by the SnO<sub>2</sub>-TiO<sub>2</sub> junction and the recombination shielding effect of the  
 4 compact TiO<sub>2</sub> film synergistically contributed to the high efficiency of 5.7%<sup>127</sup>.  
 5 Additionally, TiO<sub>2</sub> micron-cone array via electrochemical reactions was used as the  
 6 frame and electron transfer channel in fiber photoanodes. The TiO<sub>2</sub> multilayer  
 7 structure composed of compact, light scattering, and porous layers was introduced  
 8 into the fiber photoanode (**Figure 17**), and the fiber devices attained photoelectric  
 9 conversion efficiencies of 7.6% to 8.1%<sup>54</sup>. The high performance implied the  
 10 importance of designing multifunctional photoanodes. Nanoporous TiO<sub>2</sub> network as  
 11 the inner layer contributed to better connection of the nanoparticle layer and Ti  
 12 substrate, thus improving the electron transport rate and prolonging electron  
 13 lifetime<sup>126</sup>. Aerosol spray pyrolysis was used for TiO<sub>2</sub>-ZnO-based FDSCs, and  
 14 computational fluid dynamic simulation could exhibit the flow field near the fiber  
 15 substrate<sup>128</sup>.



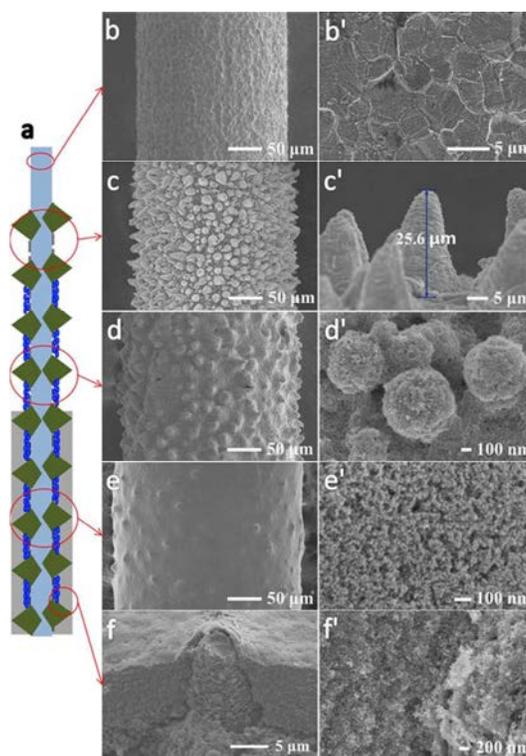
16

17 **Figure 16** (a)  $J$ - $V$  curves of the flexible FDSC (Length: 9.0 cm; photoanode diameter:

18 280  $\mu$ m). The device was tested with a normal black background (normal model) and

1 with a sheet of A4 white paper as a diffuse plate (diffuse model). Inset: Ti supported  
2 TiO<sub>2</sub> bilayer structure. (d) Trend of photovoltaic parameters ( $V_{oc}$ ,  $FF$ ,  $J_{sc}$ , and  $\eta$ )  
3 during the bending-recover cycles and the bending radius is approximately 3.0 cm<sup>56</sup>.  
4 Reproduced from Ref. 56 with permission from the Royal Society of Chemistry.

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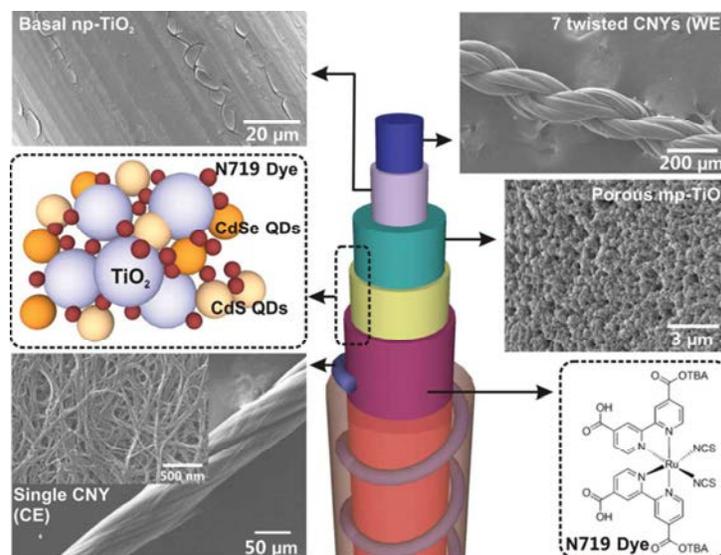
7 **Figure 17** SEM images of the novel photoanode components.(a) the schematic  
8 drawing of the whole photoanode, (b) Ti wire treated by an acid mixture,(c)Ti/TiO<sub>2</sub>  
9 micron-cone array, (d) light scattering layer, (e) porous layer, (f) cross section of the  
10 electrode; (b'), (c'), (d'), (e'), and (f') are the enlarged figures of (b), (c), (d), (e), and (f),  
11 respectively<sup>54</sup>. Reproduced from Ref. 54 with permission, Copyright © 2014  
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13

1 Recently, N3 and N719 are the most widely used dyes for FDSCs. To avoid the use  
2 of rare metal Ru, CdS and CdSe quantum dots (QDs) co-sensitized TiO<sub>2</sub> nanotubes for  
3 fiber solar cells obtained efficiency of up to 3.2% by optimizing the CdSe deposition  
4 time and the length of the nanotube<sup>64</sup>. A novel fiber solar cell with CdSe NW-grafted  
5 Ti wire achieved an efficiency of 2.9% in the S<sup>2-</sup>/S<sub>2</sub><sup>2-</sup> electrolyte<sup>129</sup>. The 1D solar cell  
6 on carbon fibers with vertical ZnO NWs and CdS QDs effectively absorbed visible  
7 light and converted it to electric energy<sup>130</sup>. Okoli and co-workers<sup>131</sup> coated  
8 QDs-incorporated TiO<sub>2</sub> on CNT microyarn and intertwined it with a carbon nanotube  
9 microyarn. This carbon-based fiber solar cell achieved an efficiency of 5.9% with  
10 long-term stability. Additionally, CdS, CdSe, and N719 were incorporated in fiber  
11 photoanode (**Figure 18**) to realize both multiple exciton generation effects and  
12 multiple electron transmission paths. An efficiency of up to 6.4% was achieved,  
13 which may have resulted from the special anode design<sup>78</sup>. Thus, novel sensitizers may  
14 offer an access for further development. Based on QD solar cells, these fiber devices  
15 do not contain any rare metal or volatile electrolyte, making them cheap candidates  
16 for applications. However, the toxic metal Cd is a drawback in this approach.

17 Photoanodes demonstrate great potential for further development because of their  
18 multiple functions of light harvesting, photo-electron generation, and charge transport.  
19 New structures, materials, and synthesis routes for fiber photoanodes are still  
20 necessary, where more improvement could be expected.





1

2 **Figure 18** The wire-shaped CdS, CdSe, and N719 co-sensitized solar cell prepared by  
 3 using a platinumized CNY as counter electrode, a braid of 7-twisted carbon nanotube  
 4 yarns with hybrid coating as the working electrode. Details are shown in SEM images  
 5 and sketch maps<sup>78</sup>. Reproduced from Ref. 78 with permission, Copyright © 2014  
 6 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

7

### 8 3.3 Catalytic Materials for Counter Electrodes (Cathodes)

9 Pt is widely used as a counter electrode for DSSCs because of its good conductivity  
 10 and good catalytic performance. Pt wire is the most common counter electrode for  
 11 efficient FDSCs, reaching an efficiency of > 8 %<sup>54, 55</sup>. For cost considerations, the  
 12 amount of Pt should be minimized. Decorating conductive fibers with Pt using  
 13 various techniques, such as magnetron sputtering, electrolytic deposition, chemical  
 14 reduction, and thermal pyrolysis, produced good results<sup>58, 90, 97, 98</sup>. Pt-decorated carbon  
 15 fibers have exhibited improved conductivity and catalytic activity toward  $I_3^-/I^-$  redox  
 16 couple than bare carbon fiber substrate, resulting in an FDSC efficiency of 5.8%<sup>90</sup>.

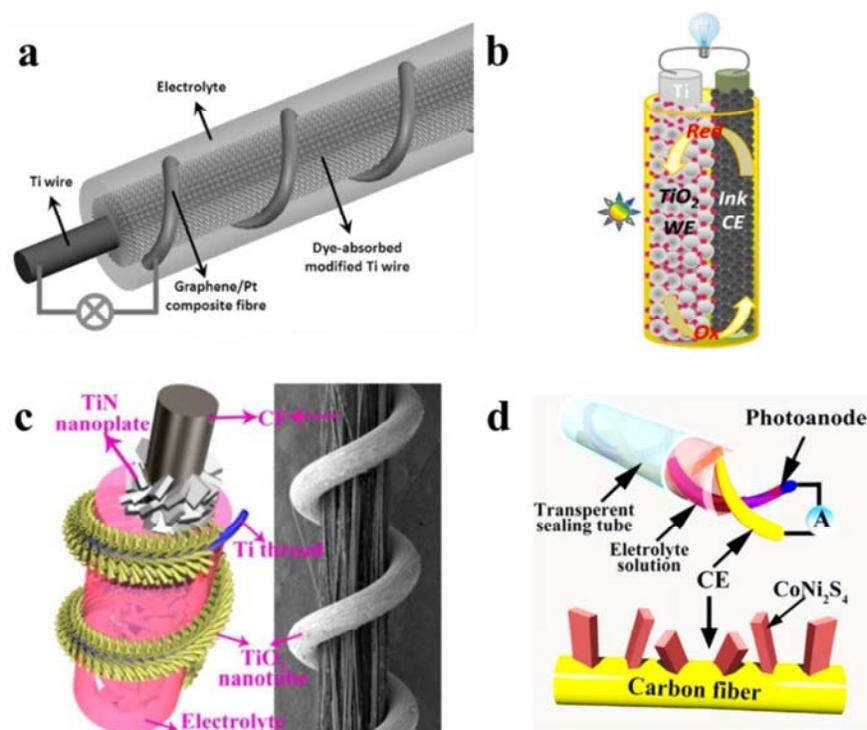
1 FDSC based on Pt-CNT yarn achieved an efficiency of 3.7%, and annealing further  
2 improved efficiency to 4.8% because of the enhanced charge transfer across the  
3 Pt/CNT interfaces<sup>98</sup>. Superparamagnetic Fe<sub>3</sub>O<sub>4</sub> and Ni nanoparticles were  
4 incorporated into CNT fibers for FDSCs, and they can stably attach to a magnet.  
5 Device efficiency of 6.0% was improved to 8.0% with Pt deposition<sup>132</sup>. Graphene/Pt  
6 composite fibers for FDSCs (**Figure 19a**) contributed to energy conversion efficiency  
7 of 8.5%<sup>58</sup>.

8 To avoid rare-earth metal Pt, carbon-based materials, conductive polymers, and  
9 inorganic nanocomplexes were used for efficient FDSCs. In the early attempts,  
10 commercial pen ink based on well-dispersed carbon nanoparticles was proved to be  
11 good catalytic material for I<sub>3</sub><sup>-</sup>/I<sup>-</sup> - and T<sub>2</sub>/T<sup>-</sup> - based electrolytes. Ink-coated SS wires  
12 and carbon fibers in parallel with the photoanodes were applied in FDSCs (**Figure**  
13 **19b**), and the optimal efficiency of 6.2% was achieved<sup>133</sup>. Nitrogen-doped graphene  
14 was also demonstrated as an efficient metal-free electro-catalyst for I<sub>3</sub><sup>-</sup>/I<sup>-</sup> redox  
15 couple.<sup>86</sup> Its pyridinic and quaternary nitrogen states especially provided active sites  
16 for promoting I<sub>3</sub><sup>-</sup> reduction. For FDSCs, the nitrogen-doped graphene catalyst  
17 prepared under 900 °C achieved an energy conversion efficiency of 5.4%, comparable  
18 with Pt-based device under similar conditions. Further study indicated that conductive  
19 polymer PEDOT: PSS composite-coated carbon fiber electrode showed catalytic  
20 performance and long-term stability that were superior to those of Pt-decorated  
21 carbon fiber<sup>96</sup>. As the coating time (layer thickness) increased, the improved  
22 conductivity and catalytic performance of the fiber electrode realized a device

1 efficiency of up to 5.6%. Polyaniline via in situ electrochemical polymerization  
2 displayed almost equivalent catalytic activity with SS sputtered with Pt, probably  
3 because of its porous characteristic and larger surface area. With this counter electrode,  
4 FDSCs achieved an efficiency of 5.4%<sup>69</sup>. Inorganic components are also good  
5 candidates for the fiber counter electrode. Porous, single crystalline titanium nitride  
6 (TiN) nanoplates were grown on carbon fibers via TiO<sub>2</sub> transfer<sup>59</sup>. Contrary to the  
7 typical design, the catalytic fiber was surrounded by helical photoanode (**Figure 19c**),  
8 and an efficiency of 7.2% was achieved in the FDSC with high  $J_{sc}$  of 19.35 mA/cm<sup>2</sup>.  
9 The intrinsic electrochemical properties, electrical conductivity of the TiN nanoplates,  
10 and their specific nanostructure (e.g., active sites, large macro-pores, tight fusion  
11 connectivity, and vertically aligned configuration) may function jointly for the  
12 photovoltaic performance. CoNiS<sub>2</sub> nanoribbon and NR on carbon fibers with good  
13 catalytic activity to I<sub>3</sub><sup>-</sup>/I<sup>-</sup> redox couple were applied to FDSCs (**Figure 19d**). The  
14 former achieved higher efficiency (7.0%) than the latter (4.1%). The distinct  
15 morphologies and exposed crystal facet are responsible for the significant distinction  
16 in performance<sup>134</sup>.

17 Given that the functional electrode provides the current cycle, the counter electrode  
18 is of equal importance in the photoanode. Given the relatively simple electrochemical  
19 process, this fiber electrode is a good platform for traditional and new material studies.  
20 In response to the reduction reaction, a catalyst is necessary to reduce the  
21 over-potential and charge transport resistance. The size/amount of counter electrode is  
22 relatively unrestricted in FDSCs, non-Pt counter electrodes can reasonably “win” over

1 Pt by larger amount and/or higher specific surface area, except for the catalytic  
 2 activity. Lighter weight, tighter connection, and smaller diameter are preferable for  
 3 portable/wearable applications. Thus, materials with excellent catalytic activity, high  
 4 specific surface area, long-term electrochemical stability, good adhesion, low cost,  
 5 and easy preparation can play remarkable roles for efficient FDSCs.



6  
 7 **Figure 19** FDSCs based on various catalytic materials. FDSCs with (a) spiral-wound  
 8 graphene/Pt composite fiber as counter electrode<sup>58</sup>. Reproduced from Ref. 58 with  
 9 permission, Copyright © 2013 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.  
 10 (b) Parallel fiber electrodes and the commercial fountain pen ink as catalyst on the  
 11 counter electrode<sup>133</sup>. Reproduced from Ref. 133 with permission from the Royal  
 12 Society of Chemistry. (c) TiN nanoplate-functionalized CFs were twisted with the  
 13 TiO<sub>2</sub> nanotube array-coated Ti thread for FDSCs<sup>59</sup>. Reproduced from Ref. 59 with  
 14 permission from the Royal Society of Chemistry. (d) FDSCs based on CoNi<sub>2</sub>S<sub>4</sub>

1 nanoribbon- and NR-modified carbon fibers<sup>134</sup>. Reproduced from Ref. 134 with  
2 permission, Copyright © 2014 Elsevier Ltd.

3

### 4 **3.3 Electrolyte**

#### 5 **3.3.1 Liquid electrolyte**

6 Liquid electrolyte based on  $I_3^-/I^-$  redox couple is the most used electrolyte for  
7 efficient FDSCs. The effects of  $I_2$  concentration on the device performance are caused  
8 by two contradictory factors: light harvesting and charge transportation. Optimized  $I_2$   
9 concentration would balance these factors. Higher  $I_2$  concentration indicates  
10 guaranteed charge transfer at counter electrode but more serious incident light loss.  
11 For concentrating systems, solar energy is excessive, and higher  $I_2$  concentration is  
12 preferable for enhanced charge transfer at the counter electrode<sup>68</sup>. When the  
13 electrolyte in FDSCs is refreshed<sup>135</sup>, the increased  $V_{oc}$  is mainly ascribed to the  
14 electron recombination resistance at the photoanode/electrolyte interface, in which  $Li^+$   
15 was proved to play an important role. The desorption of sensitizers and the increase in  
16 series resistance during the dynamic process result in decreased  $J_{sc}$  and  $\eta$ .

17 Non-iodine electrolyte thiolate/disulfide ( $T_2/T^-$ ) couple redox shows negligible  
18 absorption in the visible region, which would benefit light harvesting in FDSCs.  
19 Determining suitable catalytic materials for the redox couple is important toward the  
20 fabrication of an efficient device. Zou et al.<sup>133</sup> reported that pen ink/carbon fiber  
21 electrodes possess good catalytic activity toward  $T_2/T^-$  and contributes to achieve  
22 solar conversion efficiency of 4.0%, which is higher than that of Pt-based devices

1 (3.3%). Peng et al.<sup>75</sup> reported CNT counter electrode for flexible device with  
2 efficiency of 7.3%. Compared with core-sheath CNT/RGONR, CNT/RGO composite,  
3 and RGO fibers, the CNT fiber displays higher catalytic activity toward  
4 thiolate/disulfide couple redox and superior device performance<sup>93</sup>.

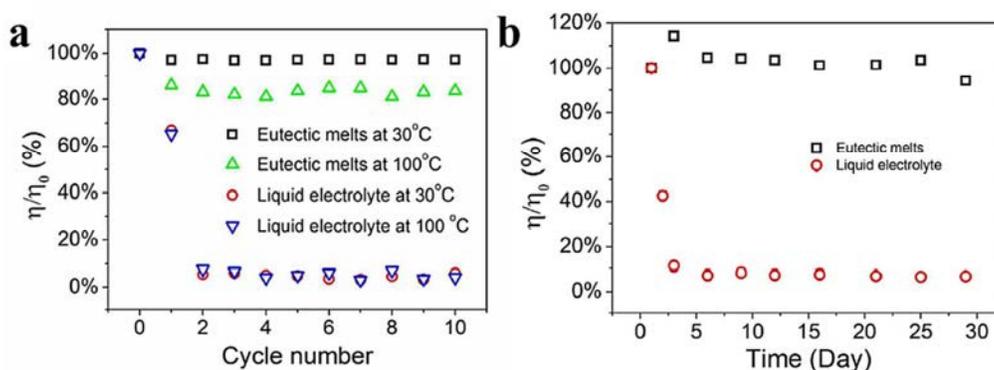
5 Conventional liquid-based electrolytes have contributed to the most efficient  
6 FDSCs, but leakage and volatilization of the organic solution are potential long-term  
7 problems. Additional sealing is used to resolve this drawback in FDSC archetypes<sup>136</sup>.  
8 In this case, volatile solvents would be avoided in electrolytes in future photovoltaic  
9 applications. However, the liquid electrolyte can guarantee moderate charge transport  
10 and light transparency, which are important in the development of highly efficient  
11 FDSCs. These characteristics are suitable supporting conditions for electrode material  
12 and interface studies.

### 13 3.3.2 Gel electrolyte

14 Gel electrolyte, which is easy to prepare and coat, may be more advantageous for  
15 practical applications. Polymer-based network can provide passageway for the charge  
16 transport between two fiber electrodes, and the device performance has been  
17 gradually enhanced in recent studies. Zou and Wu's group first reported a  
18 poly(ethylene glycol) gel containing  $I_3^-/I^-$  redox couple for quasi-solid FDSCs, and an  
19 efficiency of up to 1.5% was achieved for TNT-based device<sup>65</sup>. Kang et al.<sup>137</sup> prepared  
20 porous polyimide films as separator for FDSC using the phase inversion technique.  
21 Additionally, they prepared a series of porous polymer electrolyte membranes of  
22 PVdF-HFP with various P123 contents. The photovoltaic fibers exhibited the optimal

1 performance of 1.0%, and they were woven into mesh-like modules<sup>138</sup>. Although  
 2 reported as a solid<sup>106, 139, 140</sup>, treating the PVdF-HFP/3-MePRN/TBP composite as a  
 3 gel may be more advantageous. The refreshed efficiency of 6.4% (7.4% with a mirror)  
 4 and high  $V_{oc}$  up to 0.8 V indicate the interface corporation of fiber anode with the  
 5 electrolyte. Ionic salts possess good conductivity and low vapor pressure, and they  
 6 can also avoid solvents. Peng et al. claimed an efficiency of 2.6% with eutectic melts  
 7 of ionic salts, and the devices showed higher thermal stability and lifetime than liquid  
 8 based FDSCs (**Figure 20**)<sup>67, 141</sup>.

9 Thus, FDSCs based on gel electrolytes have achieved some progress, while further  
 10 improvements may be performed. Recent gels derived from the traditional DSSCs can  
 11 suffer from serious light absorption for FDSCs, which is responsible for the low  $J_{sc}$   
 12 and  $\eta$ . Exploring “more transparent” gels with good charge transport may be a  
 13 solution.



14  
 15 **Figure 20** (a)  $\eta/\eta_0$  values at 30 °C and 100 °C. FDSCs were repeatedly heated to 100  
 16 °C and then cooled down to 30 °C.  $\eta$  and  $\eta_0$  correspond to the energy conversion  
 17 efficiencies of the as-fabricated FDSC and the one after heating treatment. (b)  
 18 Comparison on the operation stability of FDSCs based on liquid electrolyte and

1 eutectic melts.  $\eta$  and  $\eta_0$  correspond to the energy conversion efficiencies of the  
2 as-fabricated FDSCs and the one in the following time during use. The devices were  
3 kept in dark at a temperature range of  $(25 \pm 5)$  °C and humidity range of  $(40 \pm$   
4  $30)\%$ <sup>141</sup>. Reproduced from Ref. 141 with permission from the Royal Society of  
5 Chemistry.

6

### 7 **3.3.3 Solid electrolyte**

8 Solid electrolyte for FDSCs involves stable, solvent-free, and transparent film with  
9 good charge transport. Suitable hole-transport materials can avoid liquid linkage  
10 problem and provide long-term stability. But alternative materials for this case are  
11 very limited and preparing this functional layer is quite challenging.

12 In 2008, Zou et al. first applied CuI as the solid electrolyte for flexible FDSC, but  
13 the efficiency was only 0.06%<sup>66</sup>. The defects in the CuI layer on the curved surface  
14 probably affected the carrier mobility and limited the photovoltaic performance. The  
15 TNT-based photoanode contributed to the higher efficiency of 0.21%, and the  
16 remarkably improved carrier-transfer is the possible reason<sup>65</sup>. Additionally, flexible  
17 solid FDSCs with efficiency of 1.38% were fabricated by employing annealed Ti  
18 substrate and polymer-templated photoanode. The passivation layer formed on the  
19 substrate reduced the Schottky barrier at the anode interface. The micron holes  
20 increased the filling of the CuI electrolyte, which resulted in improved electron  
21 lifetime<sup>57</sup>.

22 The solid FDSC presents a possible approach toward “dry” photovoltaic fiber as



1 well as inorganic, organic, and perovskite fiber solar cells. Given the aforementioned  
2 severe requirements, limited choices of materials are available. Even the famous hole  
3 transport material Spiro-OMeTAD was not reported for solid FDSCs, probably  
4 because of the difficulty in preparing a uniform film of this material on a curved  
5 surface and its long-term stability in the air. Novel hole transport layers may be the  
6 key point for efficient solid FDSCs.

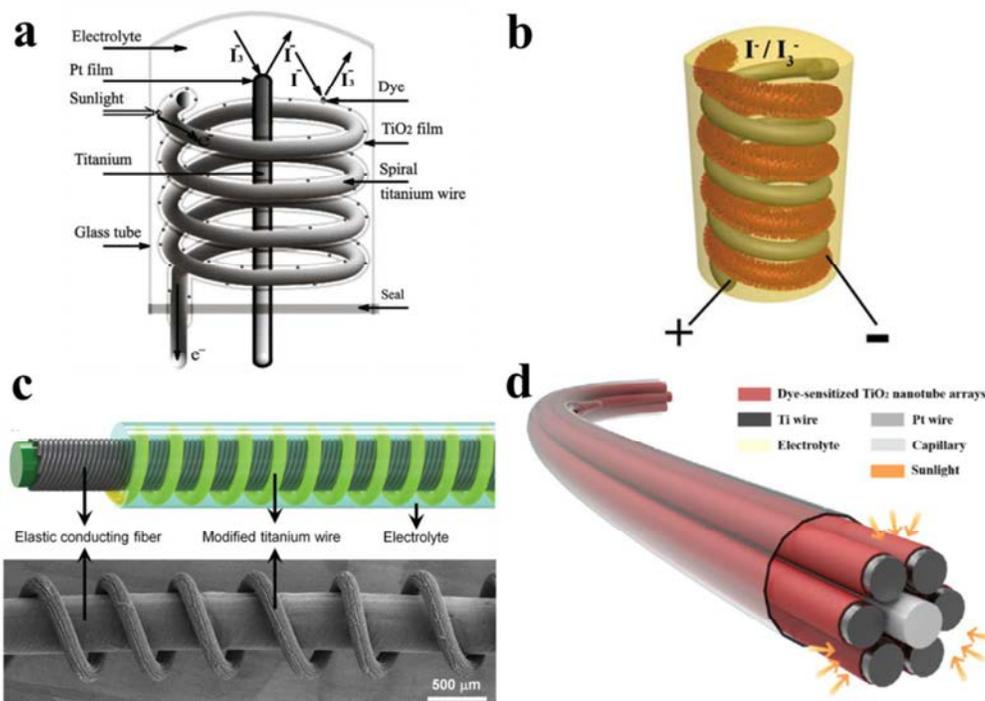
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#### 8 **4 3D Solar Devices with Light-harvesting Designs**

9 FDSCs can harvest solar energy from spatial sources because of the symmetrical  
10 structure of the fiber photoanodes. Special optical designs may be drawn on two  
11 levels: the individual fiber electrode and the fiber device itself. Light-harvesting  
12 designs were involved in power output maximization of a single device, as well as  
13 taking advantage of the active materials and fiber solar cell properties.

14 In a typical FDSC architecture, a Pt counter electrode twisted around the  
15 photoanode can hinder the incident light to the photoanode and the shading effect of  
16 electrolyte. Turning the two fibers around can avoid the problem and minimize the  
17 amount of Pt. Liu et al.<sup>142</sup> reported a mini 3D solar device (**Figure 21a**) utilizing a  
18 spiral-shaped Ti/TiO<sub>2</sub>/N719 photoanode around the counter electrode with an energy  
19 conversion efficiency of 4.1%. Under natural sunlight, the modules with tree-like  
20 structure exhibited an efficiency of up to 4.9% with 3D light utilization<sup>143</sup>. Moreover,  
21 DNA-like DSSCs with symmetrical double-helix fiber photoanode and cathode were  
22 investigated (**Figure 21b**)<sup>144, 145</sup>. With spiral wire photoanode, a novel stretchable

1 photovoltaic device was fabricated on elastic conductive rubber fibers twisted with  
2 CNT fiber (**Figure 21c**). The photovoltaic devices achieved an energy conversion  
3 efficiency of 7.1%, and the efficiency was well maintained under bending and  
4 stretching, which are important in portable electronic devices and facilities<sup>146</sup>. For 3D  
5 DSSC design, mesh-based photoanodes have also been applied to TCO-free cylinders  
6 with efficiency of 5.5%<sup>147, 148</sup>. These devices can capture light effectively from any  
7 direction to the device center. However, excess liquid electrolyte filled between  
8 electrodes in the sealing tube<sup>149</sup>. To reduce the amount of electrolyte, Hayase et al.<sup>150</sup>,  
9 <sup>151</sup> designed coil-type TCO-less cylindrical DSSCs based on metal wire-supported  
10 TiO<sub>2</sub> porous layer with an efficiency of 3.9% to 4.7%. A glass mesh was employed to  
11 avoid potential short-circuit between the two packing electrodes. TiO<sub>x</sub> layer formed  
12 on the wire, and the wire diameter affected the overall device performance.  
13 Considering the difficulty in preparing spiral dye-sensitized photoanodes and fixing  
14 electrodes, the above 3D architectures have not been paid much attention. However,  
15 opportunities are open for highly efficient and practical designs. Jia et al.<sup>52</sup> fabricated  
16 a flexible 3D solar device (**Figure 21d**) with multi-working electrodes and one  
17 counter fiber electrode, which further extended the 3D light-harvesting property of  
18 single fiber photoanode. The energy conversion efficiency increased gradually from  
19 2.8% to 6.6% as the number of photoanodes increased from 1 to 6. Interface  
20 modification with Nb<sub>2</sub>O<sub>5</sub> energy barrier refreshed the efficiency to 9.1%, which was  
21 retained about 93% under bending with different angles (1.7 cm-long device bended  
22 at 30°, 90° and 180°).



1

2 **Figure 21** Typical structures of 3D FDSCs. (a) DSSCs utilizing spiral-shaped fiber  
 3 photoanode around the counter electrode<sup>143</sup>. Reproduced from Ref. 143 with  
 4 permission, Copyright © 2009 Elsevier Ltd. (b) Symmetrical double-helix fiber  
 5 electrodes for DNA-like DSSC<sup>144</sup>. Reproduced from Ref. 144 with permission,  
 6 Copyright © 2009 Published by Elsevier B.V. (c) Stretchable photovoltaic device  
 7 based on spiral wire photoanode and elastic rubber fibers twisted with CNT fibers<sup>146</sup>.  
 8 Reproduced from Ref. 146 with permission, Copyright © 2014 WILEY-VCH Verlag  
 9 GmbH & Co. KGaA, Weinheim. (d) flexible 3D solar device with multi-working  
 10 electrodes and one counter fiber electrode<sup>52</sup>. Reproduced from Ref. 52 with  
 11 permission. Copyright © 2015 Elsevier Ltd.

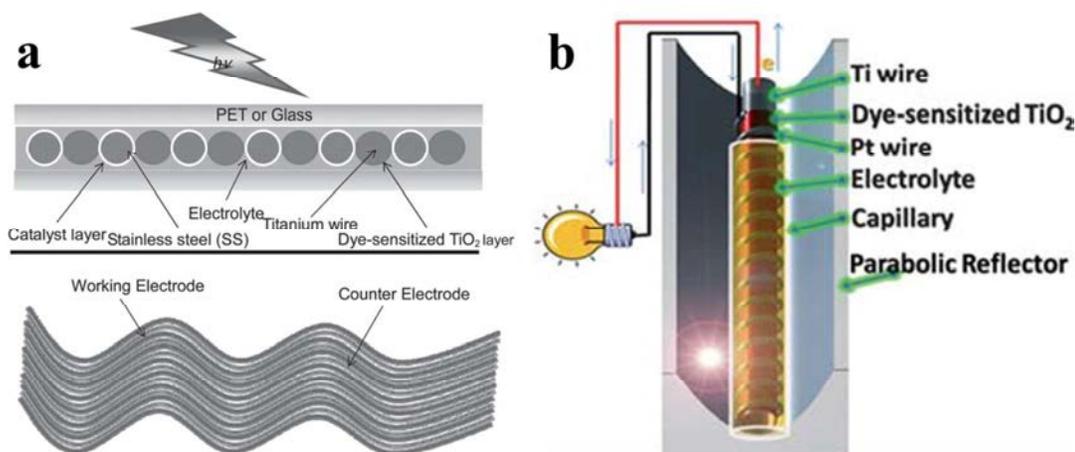
12

13 With efficient fiber electrodes, TCO-free, flexible, and bifacial DSSCs (**Figure 21a**)  
 14 were constructed with an efficiency of up to 2.4% ( $1.5 \text{ cm}^2$ ), which is quite different

1 from that of traditional sandwich-type flat-cell structure<sup>152</sup>. Similar solar devices were  
2 realized based on dye-sensitized ZnO arrays<sup>153</sup> and multi-color solar textile was  
3 fabricated with metal-free organic dyes<sup>154</sup>. A flexible polymer solar cell textile with  
4 similar light-harvesting function was also developed. Ti/TNT mesh, which was coated  
5 with P3HT:PCBM and PEDOT:PSS layers, was adhered to transparent CNT sheets on  
6 both sides. The solid, lightweight, and flexible device exhibited an energy conversion  
7 efficiency of 1.1%<sup>155</sup>.

8 Given the 3D light-collection property of fiber devices, the power output of FDSCs  
9 can be remarkably larger via catoptric and concentrated light. In 2011, Zou and  
10 co-workers<sup>25</sup> reported concentrating designs of FDSCs in conjunction with  
11 microgrooves (**Figure 22b**), including V-shaped, semi-ellipse, semicircle, and  
12 parabolic light condensers. In the parabolic groove, the converged light was  
13 theoretically perpendicularly to the FDSC and significantly increased the output  
14 power by fivefold. Similar result was confirmed by Jia et al<sup>125</sup>. To overcome the  
15 absorption of electrolyte, red-shift the incident light is a proper solution, but solar  
16 energy losses during the luminescence. Luminescent solar concentrators can  
17 down-shifting and concentrating light at the same time and could play a role in fiber  
18 solar designs. Integrating FDSCs and luminescent solar concentrators via grooves  
19 (**Figure 23**), semi-transparent and colorful waveguide solar devices were achieved,  
20 which may be used as solar windows<sup>68, 156</sup>. The effects of concentrator selection,  
21 device design, iodine concentration, illumination area, angle of inclination, and light  
22 intensity were analyzed. Maximum output power achieved an enhancement factor of

1 5.7 via efficient light collection over a large area.



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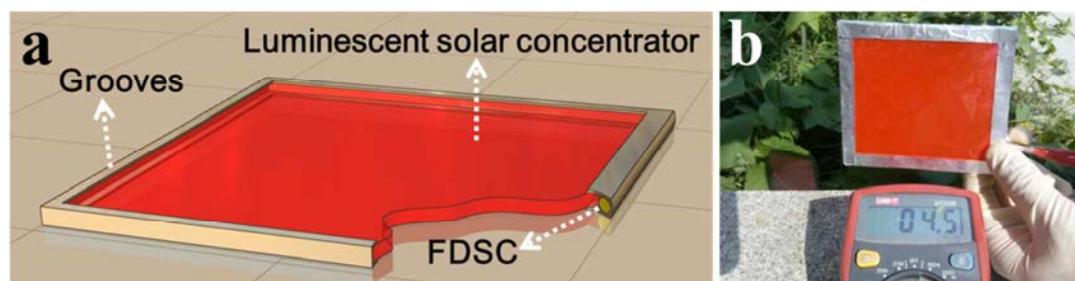
3 **Figure 22** (a) Cross section (up) and sketch (down) of the flexible bifacial DSSC<sup>152</sup>.

4 Reproduced from Ref. 152 with permission, Copyright © 2012 WILEY-VCH Verlag

5 GmbH & Co. KGaA, Weinheim. (b) FDSCs in conjunction with microgrooves<sup>25</sup>.

6 Reproduced from Ref. 25 with permission from the Royal Society of Chemistry.

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8

9 **Figure 23** (a) The scheme of waveguide solar devices integrating FDSCs and

10 luminescent solar concentrators via grooves<sup>68</sup>. Reproduced from Ref. 68 with

11 permission from the Royal Society of Chemistry. (b) Waveguide devices with nearly

12 90° tilted angle achieved  $I_{sc}$  of 4.5mA under natural light illumination (14:11-14:17

13 on September 11th, 2013)<sup>156</sup>. Reproduced from Ref. 156 with permission, Copyright

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## 1 5 Fiber Hybrid Energy Systems for Flexible Electronics

2 Solar energy is widely distributed and environmentally friendly, but the weather  
3 and night can break its continuous supply. For flexible energy management,  
4 combining solar energy conversion with electrical power storage is a reliable  
5 approach. The power fibers are lightweight, flexible, and scalable, and they can be  
6 easily integrated into electronic textiles. Integrated fiber-shaped energy devices can  
7 serve as a solar-powered system for portable microelectronics, which significantly  
8 improved the original scope of photovoltaic cells and resulted in extensive  
9 opportunities for interdisciplinary research.

10 Considering the wire shape, integration of fiber solar cells with fiber shaped  
11 supercapacitors (FSCs) has attracted much attention<sup>157</sup>. In 2011, Wang and  
12 co-workers<sup>26</sup> considered the integration of multiple energy harvesters and a storage  
13 device along a single fiber. In their design, a nanogenerator, DSSC, and  
14 supercapacitor were built on a flexible PMMA fiber coated with thin Au film (**Figure**  
15 **24a**). However, these devices were characterized separately, and the incorporation of  
16 multi-energy conversion approaches was not presented. Zou et al.<sup>69</sup> developed an  
17 integrated power fiber with overall efficiency of 2.1% that incorporated FDSCs and  
18 FSCs (**Figure 24b**). The polyaniline-coated SS wires acted as the counter electrode of  
19 FDSCs (5.4%) and symmetrical electrodes of faradic supercapacitors (19 mF/cm<sup>2</sup>).  
20 Peng et al.<sup>70</sup> applied CNT fiber for FDSC (2.2%) and double-layer supercapacitors  
21 (0.6 mF/cm<sup>2</sup>), and the integrated system exhibited an overall efficiency of 1.5%.  
22 These trials successfully used the catalytic and electrochemical properties of

1 multi-functional materials, and attempts toward gel/solid single integrated power  
2 fibers are promising. Ti wire-supported titania nanotubes and aligned CNT sheet are  
3 two electrodes for both FDSCs and supercapacitors<sup>158, 159</sup>. These bi-functional devices  
4 achieved maximal solar energy conversion efficiency of 6.58% and specific length  
5 capacitance of 85.03 mF/cm, respectively<sup>158</sup>. With solid electrolyte, the photoelectric  
6 conversion and energy storage efficiencies have reached 2.73% and 75.7%,  
7 respectively<sup>159</sup>. The overall device efficiency was maintained by 88% after 1000  
8 bending cycles and by 90% after 1000 hours. With similar design, organic fiber solar  
9 cells have also been utilized to the integrated system (**Figure 24c**)<sup>160</sup>. The organic  
10 solar cell based on P3HT: PCBM and PEDOT: PSS layers achieved an efficiency of  
11 1.0% and an asymmetrical supercapacitor energy storage efficiency of 65.6%. The all  
12 solid-state and self-powered fiber showed an entire solar-electrical conversion  
13 efficiency of 0.82%.

14 In addition to the above linear localized power fibers, coaxially integrated power  
15 fibers were also designed for multi-step energy conversion. A coaxial energy fiber  
16 integrating 3D solar cell in its sheath and CNT double-layer supercapacitor in the core  
17 was developed (**Figure 24d**)<sup>161</sup>. This energy fiber exhibited a total efficiency of  
18 energy conversion and storage of 1.83%. The coaxial structure and aligned  
19 nanostructure at the interface of the electrode enabled the device to be robust to  
20 bending and stretching. These characteristics facilitate the practical application of  
21 these devices on wearable electronics.

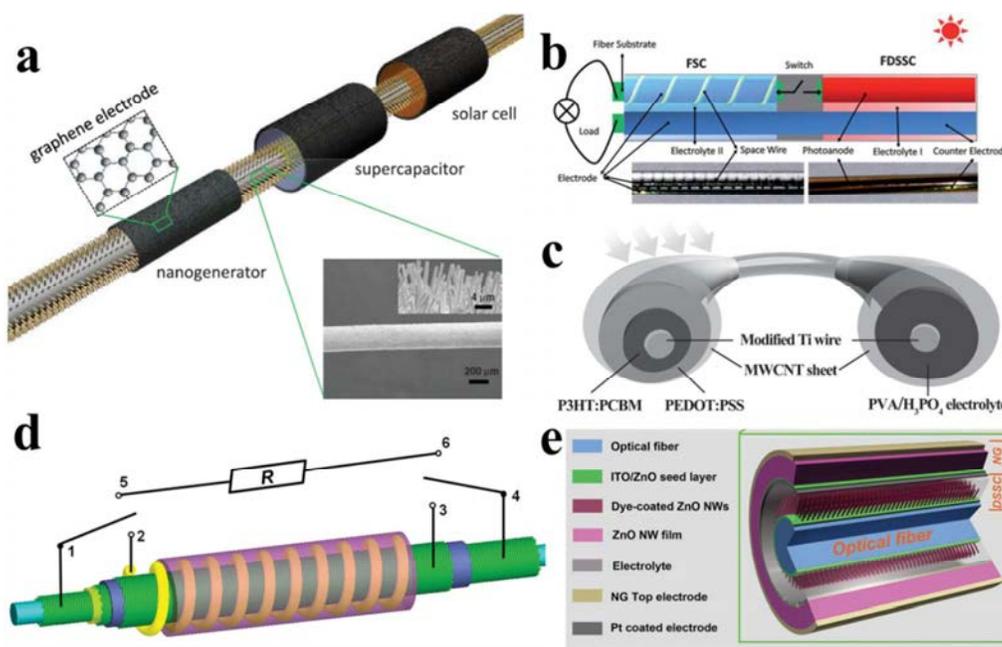
22 Although characterized with high power output, FSCs with low energy density

1 would become saturated after solar-charging for only several minutes. Thus, a fiber  
2 rechargeable battery may be more suitable for long-term energy storage. Zou et al.  
3 developed a flexible fiber-shaped zinc bromide battery with capacity of up to  
4 19 mA h/mL and energy efficiency of 69%. This battery exhibited good cycle stability,  
5 linear capacity, and linear energy density. The design included flexible conductive  
6 electrodes without additional fragmenting coating layers. This characteristic enhanced  
7 the device flexibility and allowed the fiber battery to be easily integrated into the cloth  
8 for wearable electronics. Solar-charging the fiber battery with flexible fiber-based  
9 solar modules resulted in an overall efficiency of 3.4% for the hybrid energy system<sup>56</sup>.

10 Fiber solar cells have been integrated to other hybrid energy systems. Wang et al.<sup>162</sup>  
11 demonstrated an optical fiber-based coaxially structured 3D compact hybrid cell  
12 (**Figure 24e**), in which the DSSC core harvested solar energy, while the piezoelectric  
13 nanogenerator shell harvested mechanical energy. The flexible device can work  
14 simultaneously or independently, and the output of the hybrid cell ( $\sim 7.65 \mu\text{A}$  current  
15 and 3.3 V voltage) is strong enough to power nanodevices or even personal  
16 electronics.

17 Still, recent hybrid energy systems are in the preliminary stage. More concept than  
18 design art is expected, especially device scale, performance, repeatability, and cost, et  
19 al. As the evolution of device physics, multi-functional energy devices are on the way.





1  
2 **Figure 24** (a) Fiber-based multi-energy device composed of a nanogenerator, solar  
3 cell, and supercapacitor. ZnO NWs were grown on the Au-coated flexible plastic wire  
4 and Cu mesh with conductive graphene as electrodes<sup>26</sup>. Reproduced from Ref. 26 with  
5 permission, Copyright © 2011 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.  
6 (b) Integrated power fiber consisting of an FDSC and FSC<sup>69</sup>. Reproduced from Ref.  
7 69 with permission from the Royal Society of Chemistry. (c) All solid-state, coaxial,  
8 and integrated fiber devices based on organic fiber solar cell and supercapacitor<sup>160</sup>.  
9 Reproduced from Ref. 160 with permission, Copyright © 2013 WILEY-VCH Verlag  
10 GmbH & Co. KGaA, Weinheim. (d) Self-powered, elastic energy fiber with 3D solar  
11 cell in its sheath and CNT double-layer supercapacitor in the core<sup>161</sup>. Reproduced  
12 from Ref. 161 with permission, Copyright © 2014 WILEY-VCH Verlag GmbH & Co.  
13 KGaA, Weinheim. (e) The 3D hybrid cell with an optical FDSC, which harvests solar  
14 energy, and a piezoelectric nanogenerator shell, which harvests mechanical energy<sup>162</sup>.  
15 Reproduced from Ref. 162 with permission, Copyright © 2012 WILEY-VCH Verlag

1 GmbH & Co. KGaA, Weinheim.

2

### 3 **Summary and Perspectives**

4 Various fiber/wire-shaped solar cells have achieved great progress in recent years.  
5 Especially, the energy conversion efficiency of FDSCs has remarkably and rapidly  
6 increased from  $< 0.5\%$  to  $> 9\%$ . Moreover, ultra-long, flexible, stable, and lightweight  
7 devices have been reported as potential flexible/wearable power sources. However,  
8 these notable features were realized in different systems. For example, high  
9 efficiencies appeared without long-term stability and large size. Lightweight and  
10 stable devices were reported with relatively low efficiency or power output. Future  
11 explorations should be aimed at optimizing the device parameters and features  
12 synergistically.

13 Despite promising device performance, the liquid electrolyte is the potential  
14 drawback for FDSCs. Opportunities are open for quasi-solid and solid FDSCs, as well  
15 as other inorganic, organic and hybrid photovoltaic fibers. We believe that perovskite  
16 solar cells, which can be constructed via solution process, can be a suitable candidate  
17 for efficient solid photovoltaic fibers. Further material science, interface engineering  
18 and structural designs can promote perovskite solar fibers to higher levels, such as  
19 efficient perovskite solar wires in tens of centimeters, self-standing devices with  
20 harmless/protective packages, and solar textiles for wearable power sources. Beyond  
21 material science and fiber architectures, other considerations, such as technical  
22 solutions to controllable electrode disposition and efficient manufacture/integration,

1 may be considered for actual solar textiles, which were discussed by Krebs et al<sup>163</sup>.

2 But the particularity of fiber solar cells might bring new concept to the technical field.

3 The properties of fiber devices have been gradually recognized and utilized to  
4 optical designs for high power output. Replacement of rare-metal materials is another  
5 concern in previous studies, and satisfying results have been achieved. Novel  
6 materials and micro/nanostructure construction are the keys for these breakthroughs,  
7 and the foundation for further innovations has been established. The structure of fiber  
8 devices has also been extensively developed to flexible or even elastic 3D solar  
9 devices and integrated energy systems. Although previous studies only provided the  
10 initial ideas, they facilitated the breakthrough for novel solar architecture and  
11 portable/wearable electronics of the future.

12 We are optimistic that new materials, preparation technology, and interface  
13 engineering can make a difference in the future, and photovoltaic fiber will find its  
14 role as a solar technology. Further studies on fiber innovation may also result in  
15 higher efficiency, longer lifetime, harmless devices, practical solar modules, and  
16 solar-driven electronics, among others.

17

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7



8

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24

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26

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28

29 solar cells in China.

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## Flexible Fiber/Wire Shaped Solar Cells in Progress: Properties, Materials, and Designs

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Development of various fiber/wire-shaped solar cells, conventional materials, device properties, innovative designs, and integrated power systems are reviewed.

