

Review Article

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Advanced materials for flexible solar cell applications

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Abstract: The solar power is one of the most promising renewable energy resources, but the high cost and complicated preparation technology of solar cells become the bottleneck of the wide application in many fields. The most important parameter for solar cells is the conversion efficiency, while at the same time more efficient preparation technologies and flexible structures should also be taken under significant consideration [1]. Especially with the rapid development of wearable devices, people are looking forward to the applications of solar cell technology in various areas of life. In this article the flexible solar cells, which have gained increasing attention in the field of flexibility in recent years, are introduced. The latest progress in flexible solar cells materials and manufacturing technologies is overviewed. The advantages and disadvantages of different manufacturing processes are systematically discussed.

Keywords: Flexible materials; solar cells; fabricating technology

1 Introduction

In the field of large-area and green energy supply, solar cells have wide applications [2]. As the conversion efficiency increases and cost decreases, solar cell acquires more commercial applications. The concept of flexible solar cells appeared long time ago since a flexible structure facilitates the harvest of solar power on a large extent [3, 4]. Silicon solar cells have been extensively studied since early 1950s, and an increasing number of photovoltaic materials are investigated to improve cell perfor-

mances. Monocrystalline silicon and polycrystalline silicon are applied to solar cells in succession. Since 1970s the amorphous silicon solar cells were developed [5]. Compared with the crystalline silicon, amorphous silicon is much lighter and thinner. In recent years, a variety of solar cells have been designed: dye-sensitized solar cells (DSSCs), organic solar cells, Cu(In, Ga)Se₂ (CIGS) solar cells, perovskite solar cells, *etc.* [6]. To overcome the issue of flexibility, great efforts have been made to create different flexible component materials [7]. In particular, transparent electrode is an essential part for solar cells, and ITO conductive films are commonly used [8]. Despite flexible ITO glasses, flexible solar cells with opacity electrodes made of fibers were reported [9]. A significant progress has been achieved in manufacturing different kinds of flexible solar cells and their conversion efficiencies increase along with the technology advances.

2 Materials for flexible silicon solar cells

The silicon solar cells have been made huge progresses in these years, gaining the wildest applications in our lives. The abundant resources of silicon and the relative composite elements like boron, nitrogen enable the continuous production of such solar cells. Silicon solar cells are qualified for many operating situations for their stability, safety and outstanding service life, the flexibility of such solar cells explores the applications in more specific areas like buildings and vehicles. Presently there are three kinds of silicon solar cells including monocrystalline silicon, polycrystalline silicon and amorphous silicon.

Monocrystalline silicon solar cells possess the highest conversion efficiency up to 26% and develops fast these years. By adopting thin silicon layer to be the substrate of light-trapping layer, researchers have designed different structures of solar cells with decent properties. Fang *et al.* reported an ultrathin flexible solar cell with monocrystalline silicon substrate to support the silicon nanowires [10]. The cell also has a passivating layer of

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Al_2O_3 on the nanowires which enhances the light absorbing. Testing results show a conversion efficiency up to 16.61%. Lin *et al.* also utilized a 30 m thick silicon to be the flexible substrate [11]. The substrate is made by etching in a NaOH solution until the silicon chip is fully bendable and silicon constructs a 3D structure through a process of surface texturization. McMahon *et al.* utilized a glass substrate with high transition temperature to be the flexible substrate [12]. They adopted an Al catalyst layer which is deposited on the soft glass to grow silicon nanowires, the Al layer is deposited by thermal evaporation. Then the (111) oriented monocrystalline silicon subsequently by a process of electron beam evaporation. The method provides a possible approach to effectively induce electrode materials to soft substrates. To form a flexible cell, researchers also changes the outside structure of solar cells. A flexible monocrystalline silicon band is fabricated which indicates faster silicon production and solves the inconvenience of silicon ingots.

Polycrystalline silicon solar cells and amorphous silicon solar cells have a shortage in efficiency comparing with the monocrystalline silicon solar cells, but they have lower cost and the conditions of producing silicon is less rigor. Another superiority for monocrystalline silicon is the higher optical absorption coefficient which leads to better performance in weaker light and lower cost. Plentz *et al.* reported a flexible solar cell by rigor depositing silicon onto glass fiber fabrics [13]. With advantages of highly flexibility and thermal stability, the glass fiber is a promising alternative choice for replacing traditional glass or metal substrates. For the requirement of light absorbing, an aluminum-doped zinc-oxide layer is deposited as the transparent contact layer and the back titanium layer is reduced to semi-transparent for better illumination. The Ag nano particles efficiently reflect and scatter light back and correspondingly the current density increases to 19.5% higher. Águas *et al.* reported an application of cellulose paper in solar cell fabrication [14]. The paper with a hydrophilic mesoporous layer provides a wide, light and high thermal stability substrate for silicon solar cells. The comparison with solar cells deposited on glass shows that the paper not only forms a flexible packing of the cell but also has no effect on performance. Silicon solar cells are relative stable power systems with a solid structure, as different flexible composites are proposed and developed, the efficiency of the flexible cells increases gradually. Different scheme of silicon solar cells' connections like partitioning and welding also promote the multi-application of these cells. It is be expected that silicon solar cells will gain more applications in our lives with the realization of flexible technologies.

3 Materials for flexible DSSCs

In 1991, Grätzel *et al.* reported a new kind of solar cell: dye-sensitized solar cell (DSSC), opening up a new way for the explorations and utilizations of solar energy [15]. DSSCs have the advantages of abundant sources of raw materials, low cost and relatively simple manufacturing technique [16, 17]. With wider acceptance of material purity and relatively lower cost, the DSSCs show high competitiveness with the conventional solar cells. Till the development of DSSCs so far, the efficiency has reported up to be 13% [18, 19]. The DSSCs consist of a conductor, a dye photosensitizer, a nanoporous semiconductor membrane, counter electrodes and electrolyte. The photo anode and the counter electrode are the skeletal structure of DSSCs, thus flexible DSSCs require that both of the electrodes are flexible [20]. Usually conducting glasses, *e.g.* the indium doped tin oxide-coated (ITO) glass and the fluorine-doped tin oxide-coated (FTO) glass, are used to be the transparent conductor. However, the application of glasses is limited by their fragility and high weight. Actually, researchers have made efforts to deposit transparent conductive oxide on the flexible substrates. Up to now, the ITO-sputtered PEN (polyethylene naphthalate) and PET (polyethylene terephthalate) have been widely utilized in various photovoltaic devices due to their excellent electrical conductivity, light weight and good flexibility. Furthermore, by utilizing the roll-to-roll system in industry, it's possible to mass produce ITO/PET substrate with large scales. The polymeric electrode substrates, such as the ITO-sputtered PEN and PET, are suffered with mechanical or chemical changes when treated in a high process temperature. Lee *et al.* reported a flexible ITO-PEN substrate *via* electro spray deposition to prepare hierarchically structured TiO_2 on a flexible ITO-PEN under low temperatures (less than 150°C) [21]. The simple fabrication process formed porous membranes with large surface areas and after the post processing, the cell efficiency also increases to 5.57%.

ITO plays an important role in the most field of solar cells compositing, the lack of indium resources and the producing process aggravate the cost of solar cells and present a limit in large-scale production. Therefore, it demands to find alternative materials for replacing ITO. There have been several attempts to fabricate DSSCs by using fabric-based electrodes, Kavan *et al.* crossed the transparent PEN woven fabric and electrochemically platinumized tungsten wires in vertical directions to form a flexible cathode [22]. By contrary with the thermally platinumized FTO, it shows that the flexible electrode is enhanced in ohmic resistance. On the other hand, due to the transpar-

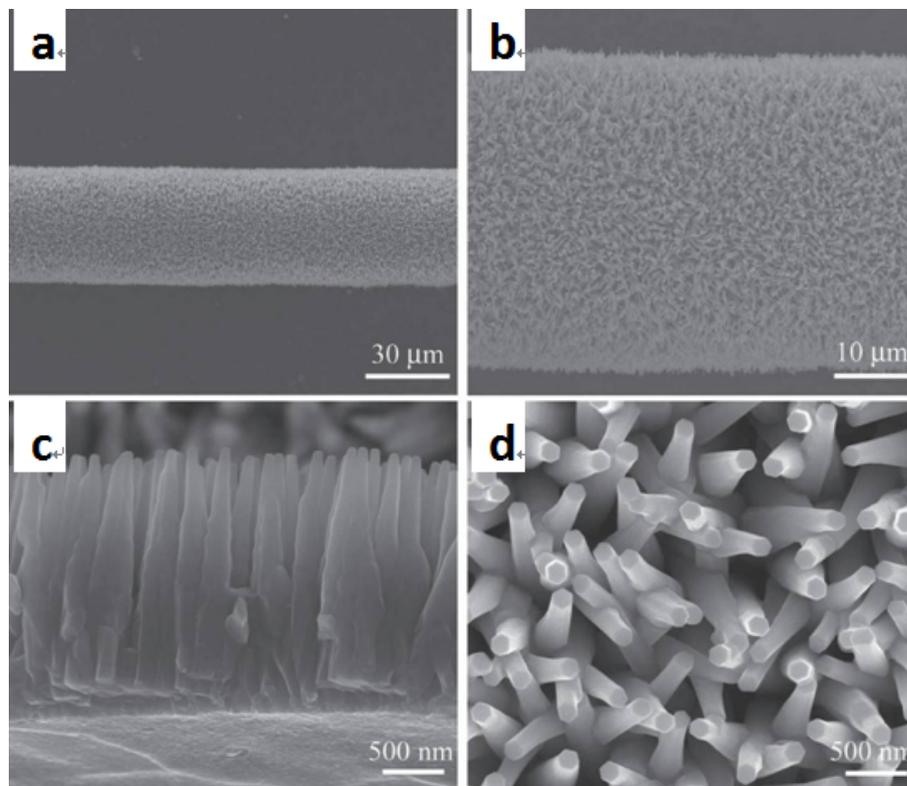


Figure 1: (a) SEM image of ZnO nanowire arrays grown on stainless-steel microwires. (b) Magnified SEM image of a wire section, uniformly covered with high-density ZnO nanowires. (c) Cross-section and (d) top-view SEM images showing the well-aligned, high-density ZnO nanowire arrays grown on the stainless-steel microwire.

ent PEN fiber and TiO_2 film, the electrode shows better optical transparency. The electrode exhibits good stability and storage capacity at open circuit for almost a month. Wang *et al.* let the ZnO nanowire arrays radially grown on wires of stainless steel, Au, Ag, and Cu as the working electrodes, while Pt wires to be the counter electrodes [23]. Figure 1 shows the SEM photo of ZnO nanowire arrays from different angles. Figures 1(a) and 1(b) show that the ZnO nanowire is along the whole length of the wire of Fe. Figures 1(c) and 1(d) clearly exhibit the fully uniformity of the nanowires. The double-wire DSSCs possess merits of very good flexibility and high transparency via a simple, facile, and controllable way of disposition. Corning[®] produced a type of flexible glass called Willows[®] Glass whose thickness was only 100 μm , significantly increases the substrate flexibility and reduces the rigidity with bend stress less than 100 MPa at a bend radius of 5 cm [24, 25]. Such novel flexible glass has been applied in flexible applications as well as other solar cell devices, showing the potential for mass manufacturing of flexible devices. Sheehan *et al.* tested the DSSCs fabricated with the Willows Glass, the results demonstrate that flexible Willow Glass substrates with higher thermal stability owns the potential in roll-to-

roll manufacturing for application in solar cells [20]. Meanwhile by comparing with the commercial ITO glasses, flexible glass substrates fabricated DSSC generates competitive efficiency with the conventional DSSCs.

The electrolyte between the two electrodes has significant effects on the photoelectric performance and operate stability of flexible DSSCs. Conventionally high efficient DSSCs commonly used liquid electrolyte with low viscosity organic solvents, when applied in flexible DSSCs, the photo anode without high-temperature treatment cannot withstand long-term soaking. In addition, the risk of hard to encapsulate, easy to leak out and dissolving the polymer substrate are also difficult to resolve. Hence the gel or all-solid-state electrolyte becomes an important research direction for preparing stable, efficient and flexible DSSCs. Hoang *et al.* added lab-made platinum nanoparticles to improve the charge transfer of the gel electrolyte (based on polyethylene oxide (PEO)) [26]. The nano Pt serving as the catalyst to the gel electrolyte enhances the photovoltaic performance of DSSCs and improves the inside charge transfer between the redox layer and the oxidized dyes. The defect of the gel materials which limits its employment is that they don't have specific forms. When the

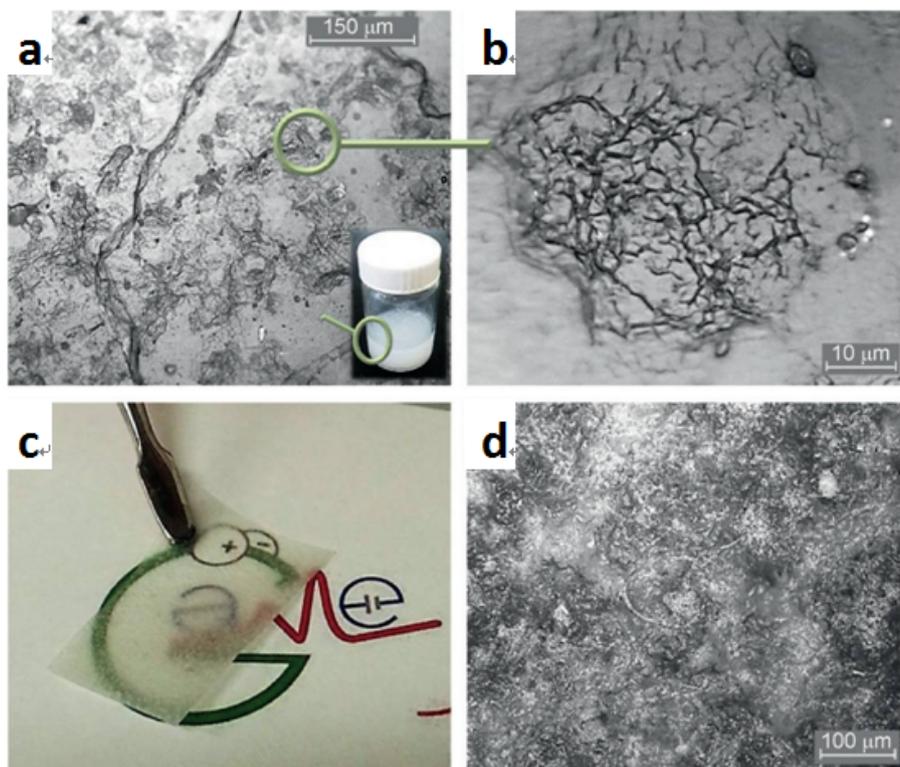


Figure 2: (a, b) Optical microscope analysis of the NFC dried network of fibers; (c) appearance of a 30 wt% NFC-based PEM; (d) a 30 wt% NFC-based PEM observed with an optical microscope.

flexible cells are bent, the two electrodes will possibly come into contact, causing problems like short out. Polymer electrolyte membranes (PEMs) are good alternative materials to improve the gels' defect, since they are self-standing with specific form which is more convenient and appropriate for the preparation of solid electrolytes for flexible DSSCs. Cellulose nanofibers, as introduced earlier, are explored to prepare PEMs. The high sunlight conversion efficiency of 7.03% is achieved at simulated light intensities of 1.0. Furthermore, the cellulose nanofibers also improve the long-term stability of the cells and excellent durability of over 95% retentive efficiency after treated by extreme aging conditions. Chiappone *et al.* selected Bisphenol A ethoxylate dimethacrylate (BEMA) and poly (ethylene glycol) methyl ether methacrylate (PEGMA) mixing with NFC solutions to prepare PEMs [27]. Figure 2(a) and 2(b) show the networks of the fibers and Figure 2(c) and 2(d) show the appearance of a stable PEM formed by 30 wt% NFCs under optical microscope. By adding the NFCs the film appears to be flexible but not rigid, free-standing and forms a stable network after drying which means great mechanical property. The addition of NFC enhances the stability of the devices and remains more than 95% of the efficiency under the over operating conditions.

4 Materials for flexible perovskite solar cells

The perovskite solar cells were first put forward in 2009. The photoelectric transformation efficiency was only 3.8% at that time [28]. Zhou *et al.* promoted the efficiency to 16.6% on average, with the highest efficiency of 19.3% in 2014 [29]. And the efficiency has been rapidly risen up to 20.1% at present [30]. The speed of development of this technology is unprecedented and is worth high attention. The light absorption layer material of is $\text{CH}_3\text{NH}_3\text{PbX}_3$ (X: I^- , Br^- or Cl^-), the material is typically perovskite structure, and was first used as a novel dye in DSSCs. The cell structure was similar to typical DSSCs until Kim *et al.* reported an all-solid-state perovskite solar cell in 2012 [31]. They employed perovskite nanoparticles as light harvesters, and with standard AM-1.5 sunlight such cells generated large photocurrents exceeding 17 mA/cm^2 . Research on flexible perovskite solar cells subsequently made a significant progresses in the recent years. The difficulties for flexible perovskite solar cells to overcome include: flexible materials for both sides of substrates and alternative electrode materials to take place of Au or Ag to be the counter electrode. Das *et al.* prepared a flexible perovskite solar cell

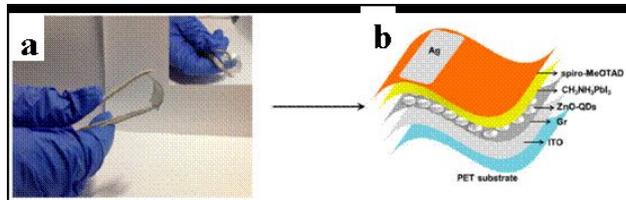


Figure 3: (a) Photograph of ITO-PET/Gr, (b) a schematic illustration of the fabricated ITO-PET/Gr/ZnO-QDs(AP jet)/CH₃NH₃PbI₃/spiro-MeOTAD/Ag flexible perovskite solar cell.

with high performance by a ultrasonic spray coating process [32]. With the PET substrate, the cell was bendable and the conversion efficiency was as high as 13%. A problem for flexible perovskite solar cells is that the high temperature requirement for producing the TiO₂ layer which would limit the wider application of the device. You *et al.* prepared a high efficiency and flexibility perovskite solar cell using polyethylene terephthalate/ITO to be the flexible substrate [33]. The component layers were processed in solution form under 120°C and a 9.2% efficiency was achieved. Similarly, in order to solve the low-temperature preparation problems, Dkhissi *et al.* reported a method to fabricate planar perovskite solar cells with controlling the entire process under the temperature of 150°C [34]. By employing p-type poly (3,4-ethylenedioxythiophene):poly (styrenesulfonate) (PEDOT:PSS) to replace TiO₂ making up as the blocking layer, the process temperature is able to control and ensure a high efficiency of 12.3%.

ZnO provided an alternative electron-transport layer material which is much thinner as reported by Liu *et al.* in 2013 [35]. Without the sintering process, the temperature can be reduced. Solar cells based on the design showed nice flexibility and exhibited conversion efficiency as high as 15.7%. Ameen *et al.* used ZnO quantum dots treated by the atmospheric plasma jet as the electron transport layer and ITO-PET as the substrates to assemble highly flexible perovskite solar cells [36]. In Figure ?? the ITO-PET/Gr thin film substrate and the fabricated solar cells are clearly showed. Figure 3(a) shows good flexibility of the substrate and Figure 3(b) demonstrates the cell structure. The ZnO quantum dots are coated on the ITO/Gr substrate by the atmospheric plasma jet method. Analysis shows that the jet-treated thin film fabricates the perovskite solar cell with enhanced photo current density. Additionally, the fabrication process with simplified procedures, long-term stability and flexible structure reveals the high developing potentials of such novel perovskite solar cells.

5 Materials for Cu(In, Ga)Se₂ solar cells

Copper indium gallium selenide thin film solar cells (CIGSS) is quickly developed after the 1980s among the various types of solar cells. With the advantages of high efficiency, low cost and flexible possible, CIGSSs are considered to be the best potential choice of the next solar cell generation. The highest conversion efficiency of CIGSSs reported so far is 20.8%, with the deposition on a rigid glass substrate [37]. Comparing with traditional CIGSSs, the flexible CIGSSs are lighter, foldable and can be applied to uneven surface [38, 39]. Flexible substrate CIGSSs also open up more application possibilities in the ground and space, for instance, the transport, installation, maintenance and disassembly. Methods like evaporation, sputtering and deposition are suitable for flexible electrode producing and how to better coordinate the connection of cells also becomes the key points. The choice of flexible substrate should meet the requirements of good thermal stability, which is able to bear the high temperature environment when producing the absorption layer, chemical stability without reacting with Se, and adaptability of vacuum and suitable for coiling. Flexible foils are the most commonly used flexible substrates, they are demonstrated with cost-effectiveness and versatility [40, 41]. In particular, stainless steel is one of the most promising materials as a flexible substrate. Many reports demonstrate the possibilities of the deposition of CIGSSs on stainless steel with varied methods. In attempt to increase the efficiency, Liu *et al.* reported a flexible CIGSSs on stainless steel with a special Ti/TiN composite structure [42]. The efficiency of 9.1% is demonstrated as a consequence of the unique structure for the Fe ion diffusion barrier. Broadband nanostructured antireflection coatings are applied in flexible CIGS solar cell as reported by Pethuraja *et al.* [43]. In addition, combined flexible foils can also be applied as the supporting layer, and the addition of Mo and Cr on stainless steel deliver the highest conversion efficiency of 9.88% [44]. And CIGSSs on stainless steel with deposited Na/Mo layer exhibit the highest efficiency of 15.04%. Polymer substrates, on the other hand, are another interesting materials, which also have been developed intensively [45, 46]. Li *et al.* applied the co-evaporation process to deposit CIGSSs on polyimide, obtaining a conversion efficiency of 7% [47]. Figure 4 presents the SEM images of the CIGSSs cross-sections made by the modified method and the traditional method, respectively. With the modified three-stage co-evaporation process, the grain size of CIGS becomes significantly larger than the traditional process. The combi-

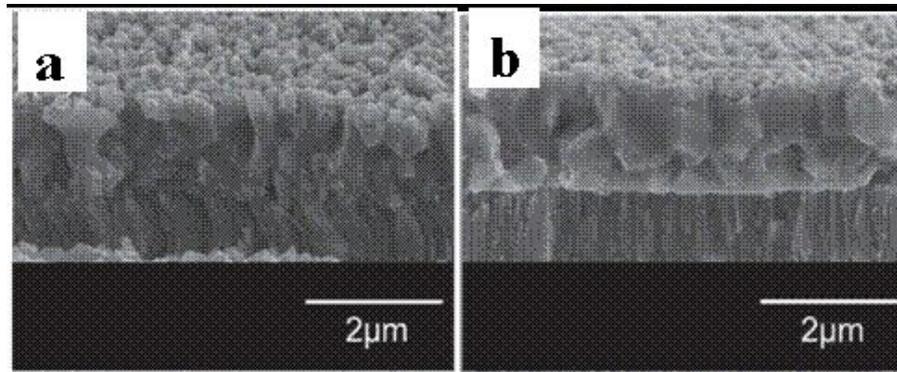


Figure 4: SEM images of CIGS thin films grown with (a) traditional process and (b) new process.

nation of flexible foils and polymers is also found with potential to serve as a promising substrate. Yu *et al.* deposited and laser scribed a Mo thin film on the polyimide substrate to assess the capability of scribing on the combined film [48]. Although many of CIGS solar cells with flexible substrates are demonstrated with high efficiency of ~10%, it still remains to be developed.

6 Conclusion and future outlook

Unlike solar cells based on rigid slabs or filmy cells on the glass substrates, the most important traits of flexible solar cells are of light weight, shatter-resistant and they exhibit high specific power. The technology of flexible solar cells is also reposed on the flexible substrates, for instance: the stainless steel or polymers [49, 50]. As most of the solar cells have requirements of high transparency of the electrodes, researchers are searching for flexible transparent materials to replace the hard glasses in the part of light blocking layer. The flexible structure of solar cells provides opportunities to continuous and mass production of power supply in the future. It can be expected that flexible energy applications will eventually change the energy structure profoundly and transform our life enormously.

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