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Research status of large-area flexible organic solar cells

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Abstract: Solar cell technology, as the hottest clean energy technology nowadays, can convert the abundant solar energy on earth into electricity. As the latest development direction of solar cell technology, large-area F-OSCs have a broad application prospect. Compared with traditional inorganic solar cells, large-area flexible organic solar cells(F-OSCs) have various advantages including flexibility, low cost and low weight. Because of its unparalleled advantages, the research field of large-area F-OSCs has achieved rapid development in recent years. This review shows representative results in the field through different aspects of the structure, fabrication process and applications of large-area F-OSCs. Also this review summarizes the problems that still exist in various aspects of large-area F-OSCs. Nowadays, the main problems that need to be solved for large-area F-OSCs are 1) increasing the power conversion efficiency(PCE) of the cells 2) improving the stability of the devices in the natural environment and 3) reducing the cost of manufacturing. Finally, an outlook on the future widespread application of large-area F-OSCs is presented.

Keywords: Organic solar cells, Large-area devices, Flexible devices, Power conversion efficiency.

1. Introduction

In 1986 Kodak's tang team prepared a double-layer organic solar cell(OSC) with copper phthalocyanine and perylene tetracarboxylic derivative as materials, achieving 1% PCE and achieving a breakthrough in OSCs [1]. After 35 years of continuous research, the PCE of small-area OSC devices has exceeded 19% in 2021 [2]. While the development of small-area rigid OSCs has been very rapid, the growth of large-area F-OSCs is still relatively backward.

Most solar cells on the market today are silicon-based inorganic solar cells based on rigid substrates. Compared with rigid silicon-based inorganic solar cells, F-OSCs have the advantages of flexibility, foldability, low cost, and also light weight. However, F-OSCs nowadays generally face the problem of lower power conversion efficiency (PCE) than their counterparts with rigid indium tin oxide (ITO) electrodes. Also the bending and tensile properties of the material are another important issue for F-OSCs. With the breakthrough in device power conversion efficiency for rigid small-area OSCs, researchers have been able to redirect their research in directions other than improving PCE. New technologies that have been applied to rigid OSC devices to improve PCE, such as novel donor molecules, acceptor molecules, transport layers and coating processes are also applicable to large-area flexible OSC devices. In 2020, Zhixiang Wei's group successfully made the device PCE of large-area F-OSCs maintain 98% of the PCE of small-area rigid devices [3]. Nowadays, the research direction of large-area F-OSCs focuses on 1) developing flexible substrates with good bending performance and high light transmission at the same time; 2) improving the light transmission and electrical conductivity of FTE; 3) improving the manufacturing process applicable to large-area flexible devices.

2. Device structure

The more typical large-area flexible organic solar cell structure consists of top electrode, transport layer, photosensitive layer, transport layer, and flexible transparent conductive electrode (FTE).

2.1. Flexible substrates

Substrates for F-OSCs are now widely available in polyethylene naphthalate (PEN), polyethylene terephthalate (PET), and polyimide (PI). PEN and PET films have both solvent resistance and transparency, two excellent properties. PI substrates have better high-temperature stability and photostability and can also be used to make flexible substrates for F-OSCs, but PI materials also have the problem of being opaque in the visible wavelength region, and functional units need to be added to turn PI into colorless. At the same time a variety of ultra-thin substrates of different materials can be used in F-OSCs to improve the light trapping ability and foldability of the devices.

2.2. Photoactive layer

The photosensitive layer is the most important part of the OSC. The photosensitive layer directly absorbs photons and converts them into electrical energy. In OSCs, a thickness of about 100 nm is optimal for the photosensitive layer. Nowadays, polymers are commonly used as donor molecules and non-fullerene materials as acceptor molecules, and the donor molecules are mixed with the acceptor molecules to form bulk heterojunction (BHJ) photoactive layers. The development of Y6 and its derivatives in the receptor molecule has led to increase in the efficiency of OSCs, which is now capable of achieving a PCE of 19%. In 2021 Jianhui Hou's group achieved a maximum PCE of 19% in a single-junction OSC by combining material design with a ternary blending strategy [2]. A variety of new donor materials have been reported so far. All of these materials have good photovoltaic properties, such as, D18 and its derivatives, BTR-Cl, etc.

However, these materials are still less used in the processing of large-area devices. Meanwhile the donor materials and acceptor materials of OSCs change in the presence of UV light, water and oxygen in the air, often causing irreversible effects on device performance. Using the PI material introduced earlier, a Japanese team has developed a solution to filter UV light using PI as a substrate. The research team blocked 90% of the 350nm light that was in the UV range using a substrate made of 1.3μm PI material. Finally OSCs with 9% PCE using transparent PI substrates with photostability were made [4].

2.3. Transport layer

The role of the transport layer is to facilitate the transport and extraction of electrons and holes generated in the device. Improving the performance of the transport layer is also a key point to improve the efficiency of the device. The usual material used for the hole transport layer is transition metal oxide, in addition to which PEDOT: PSS is also commonly used. Transition metal oxides are specifically molybdenum oxide, nickel oxide and tungsten oxide, etc. The commonly used materials for the electron transport layer are metal oxides, such as titanium oxide, ZnO, SiO_2, etc. However, there are still shortcomings in the materials commonly used for the transport layer, so a variety of new solutions for transport layer materials have emerged in recent years. For example, to solve the hydrophilic problem of the hole transport layer material PEDOT: PSS, Yinhua Zhou's team used some special methods to reduce the acidity of PEDOT: PSS to improve the stability. A lower acidity was obtained by using perfluorosulfonic acid ionomer as counterion in the alcohol dispersion formulation. The 12.2 cm^2 large-area device fabricated with this transmission layer obtained a high stability of 83% original efficiency at the maximum power point after 1330h of continuous light exposure [5].

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2.4 Flexible transparent electrodes

An important reason limiting the growth of large-area F-OSCs at this stage lies in the flexible transparent electrodes. The light transmission of FTEs will directly affect the power conversion efficiency of OSCs. The higher the transmittance of FTE is, the more light will reach the active layer through the transparent material, which can make the light loss reduced. Another unavoidable problem of FTE is the high square resistance that leads to electrical losses. The high square resistance will be accumulated as the device area continues to expand, and the high square resistance becomes an important reason for the low PCE of large-area devices. Therefore, the future direction of research and development is to develop FTEs with low square resistance and high light transmission. ITO electrodes are now the most commonly used transparent electrodes for OSCs and possess superior performance. However, the disadvantages of poor ITO flexibility and high cost make it difficult to be applied to large-area F-OSCs. For the current situation, the technology for ITO applications is very mature. Electrodes made from ITO deposited on substrates commonly used for substrates are PEN and PET. Electrodes made in this way are more common in the market and can be purchased directly, so they are suitable for assessing the feasibility of other components of F-OSC. PEDOT: PSS is a conductive polymer in aqueous solution commonly used nowadays, and the conductivity of the solution varies depending on the formulation. PEDOT: PSS has adjustable work function and high optical transparency, so the product has been widely used commercially. PEDOT: PSS has been used in high-efficiency devices produced by several teams in recent years. Also PEDOT: PSS can be modified with other pairs of electrodes to improve device performance, which will be described later. However, the hydrophilic nature of PEDOT: PSS can affect the stability of the device in natural environments. In addition, there are also electrodes that take advantage of the high electrical conductivity of metals. Metallic grids electrodes are made of metal for conductivity, while using the gaps in the grid for light transmission. The metal possesses excellent electrical conductivity, making the metallic grids electrodes conductive as well. However, metallic grid electrodes are poorly compatible with plastic substrates and need to be modified to improve performance. For example, in 2019 Changqi Ma's group used PEDOT: PSS (E100) to modify Ag/Cu electrodes to achieve surface planarization. The PCE of 1 cm^2 large-area F-OSC device made with this electrode and NF3000-P:NF3000-N as active layer (NF3000-P as donor and NF3000-N as acceptor) reached 12.16% with no decrease in efficiency after 6200 hours of storage or 1000 times of bending [6]. Silver nanowire electrodes are also another excellent performance electrode with high light transmission and low square resistance, and good flexibility. However, there are still some problems with silver nanowire electrodes. If the thickness of AgNWs is increased in order to improve the conductivity, it will lead to the decrease of light transmission and bring the problem of surface roughness. The electrode made by Zhou Yinhua's group in 2021 used PET/Ag grid/AgNWs as the structure, and the electrode with this structure obtained extremely high transmittance. The team obtained AgNWs: PEI-Zn flexible transparent electrodes with low surface roughness by combining AgNWs with zinc-chelated polyethyleneimine(PEI-Zn), and also obtained good optoelectronic and mechanical properties. Large-area devices structured with PET/Ag grid/Ag NWs: PEI-Zn/AL/MoO_3/Ag fabricated by spin-coating and water transfer methods achieved excellent results of 13.1% PCE for 6 cm^2 and 12.6% PCE for 10 cm^2 [7]. The application of carbon nanomaterials such as graphene and carbon nanotubes in F-OSCs has also been extensively studied. The low contact resistance of carbon nanomaterials with organic materials, excellent transparency, and good electrical conductivity make them very promising for use as FTEs. Yang's team obtained excellent optoelectronic and mechanical properties by integrating PI directly on graphene to create the electrodes. The electrodes possess over 92% light transmission and a square resistance of 83Ω/sq. An F-OSC device with a PCE of 15.2% was made by this electrode [8]. Unfortunately, graphene electrodes have not yet been applied to large-area device fabrication. In summary, many breakthroughs in large-area F-OSCs have been obtained in the recent, but production issues and stability problems still limit the development of large-area F-OSCs. Also compared to small-area OSCs, large-area OSCs have been less studied. The efficiency of large-area devices has been lagging behind that of small-area devices. By devoting more research to large-area F-OSCs, It can be speculated that a solution suitable for large-area F-OSC may be available soon.

3. Manufacturing

The active layer, interfacial layer and electrodes of large-area F-OSCs have specific requirements for fabrication methods. Also if different materials are used, different processing methods are required. Most of the traditional laboratory fabrication methods are done by spin-coating, which cannot be applied large-scale production. Therefore for large-area F-OSCs, finding those manufacturing methods that can be transferred to R2R manufacturing while meeting different manufacturing requirements is a key point. For large-area F-OSCs, as the area continues to expand, a large number of defects will appear in the active layer of the film, making the device efficiency significantly reduced. Therefore for large-area devices the process needs to be optimized. Nowadays, for manufacturing large-area active layers, squeegee coating, gravure printing, inkjet printing, and the most commonly used method is slot die coating [9].

For large-area F-OSCs electrodes, different fabrication methods need to be determined depending on the material. For metal or metal oxide thin film electrodes, metallic grid electrodes, the balance of conductivity and transparency is very important. Nowadays, mature ITO materials are generally fabricated using 0D coating technology. Screen printing techniques are commonly used for electrodes made of metallic materials, and the two-dimensional layer technique allows the electrodes to obtain sufficient transparency. For silver nanowire electrodes, screen printing, inkjet printing, or gravure printing techniques can be utilized. For materials such as molybdenum oxide, ZnO, and titanium oxide, which are commonly used as transport layer materials, smooth films can be fabricated by 0D controlled spraying. It is also possible to print ZnO films using inkjet printing technology, but the higher film roughness can cause a decrease in the PCE of the device.

4. Application of large-area OSC

Although there are still unavoidable problems in efficiency and manufacturing of large-area F-OSCs, many practical applications of large-area F-OSCs are now emerging. In the application of medical devices, flexible photovoltaic devices can supply energy to various medical devices. Especially for some implantable devices, self-powered implantable devices can reduce the surgical risks associated with the power supply batteries of implantable devices [10]. In everyday life, in 2019 E.G. Jeong's team developed a SiO_2-polymer composite overlay to serve as an encapsulation barrier and fabricated a photovoltaic module photovoltaic module: including a textile-based polymer solar cell (PSC), light-emitting diode, and encapsulation barrier. The photovoltaic module maintained stable characteristics even after 20 cycles of ten minutes per wash. Even after bending and washing, it did not show any performance degradation within 30 days [11]. This research provides an excellent solution for water-washable, wearable optoelectronic devices. In agriculture, in 2020 Brendan T. O'Connor and team proposed a greenhouse with integrated translucent OSC for net-zero energy consumption [12]. Also in 2020, Ziyi Ge and his team reported the use of F-OSC as a roof in a simulated greenhouse at laboratory scale. After nine days the mung bean leaves in the greenhouse showed the same growth rate as those under natural light [13]. These two research results show the potential of large-area OSCs in agriculture. Back in 2015, at the Milan Expo, a solar tree was demonstrated. Although the PCE of the device is relatively low, 8.5% for 0.04 cm^2, 7.2% for 1 cm^2, and 4% for 250 cm^2 [14]. However, this research has opened the world's eyes to the feasibility of F-OSC system that can be processed on a large scale.

5. Challenges and future outlook

With global warming and the shortage of fossil fuels, which are important issues for all countries, the use and development of new energy sources has been greatly promoted. After 37 years of reaserch, since 1986, the PCE of OSCs has gained tremendous progress, increasing from 1% to 19%, which is a very exciting result. For large-area F-OSCs, many breakthroughs have been made in recent years. Yinhua Zhou and his team fabricated large-area flexible devices with PET/Ag lattices/AgNWs structures as electrodes, and PBDB-T-2F: Y6: PC71BM as active layers. The device, after a rational modular design, 54cm^2 obtained an efficiency of up to 13% through a large-area flexible device consisting of 9 sub-cells. The report also demonstrates the application of using flexible large-area OSC for charging cell phones

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[7]. This research also provides an opportunity to demonstrate a completely new solution for the production of large-area devices. By modularizing F-OSC devices, large-area devices can be made by stitching the modules together. This avoids the difficulties associated with direct manufacturing of very large area devices. Through this paper for the current stage of the emergence of several OSCs in real-world applications can be seen, although the overall efficiency of large-area devices compared to small-area single-section OSCs is still relatively backward, but the future direction of OSCs in daily life applications still belong to large-area devices. The large-scale production and commercial application of large-area flexible OSCs still has great difficulties at this stage. These difficulties will be summarized in the rest of this paper, and the authors hope that they will be useful for future research.

5.1. Efficiency issues

The PCE of large-area F-OSCs has gained large numbers of breakthroughs, but there is still a big gap compared with inorganic chalcogenide solar cells and inorganic silicon-based solar cells. The lower PCE will make OSCs much less economical. With the introduction of each part of F-OSCs in the previous section, the following points can be summarized. If efficiency of large-area F-OSCs should be improved, researchers need to: 1) The material of the active layer needs to be further improved, and the photosensitivity of the active layer also needs to be further enhanced. 2) enhance the ability of ETL/HTL to transfer electrons and holes, while optimizing the surfaces where the transport layer joins other layers to reduce power losses. 3) improve the efficiency of the device by studying transparent flexible electrodes with higher light transmission and conductivity. At the same time, future FTEs need to meet the characteristics of smooth surface and good flexibility at the same time. 4) improve light transmission, mechanical properties and thermal stability of flexible substrates, so that the substrate can adapt to different manufacturing methods. 5) optimize the manufacturing method of large-area organic material films and reduce the defects in large-area films to suppress efficiency losses still occur during the manufacturing of large-area devices.

5.2. Stability issues

The acceptor and donor molecules at this stage and a variety of other organic materials are susceptible to natural environmental influences. A variety of factors such as ultraviolet light under natural light conditions, water in the air, and oxygen can lead to the denaturation of organic materials. Avoiding this deficiency by changing the properties of chemical substances or blocking the influence of natural environment through proper design would be an excellent method to improve the stability of OSCs. Also for large-area flexible OSCs, the mechanical stability of the material is important. Whether the material can withstand multiple bending without loss of performance will affect the application prospects of F-OSCs.

5.3. Cost issues

Low cost is also one of the advantages of large-area F-OSCs, but the manufacturing cost of large-area F-OSCs is still high in the context of large-area OSCs that cannot be manufactured on a large scale at this stage. The complex molecular synthesis and strict experimental conditions make the cost of various organic raw materials actually not low. At the same time, if the stability issues mentioned above cannot be solved, the commercial prospects of large-area F-OSCs remain poor.

6. Conclusion

To sum up, although large-area flexible organic solar cells are still not available for application in reality, there are research teams trying to solve the various problems mentioned above. Although the performance of today's OSCs still does not meet all requirements, four very exciting applications have been described in this paper. OSCs have shown many different applications in a lot of fields, such as medical, agricultural, and daily life. It is believed that in the near future, the problems of stability, cost and efficiency of large-area F-OSCs will be overcome in the future, at that time, large-area F-OSCs will be used more and more in various fields

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References

[1] Tang C W. Two-layer organic photovoltaic cell[J]. Applied physics letters, 1986, 48(2): 183-185.

[2] Cui Y, Xu Y, Yao H, et al. Single-junction organic photovoltaic cell with 19% efficiency[J]. Advanced Materials, 2021, 33(41): 2102420.

[3] Wang G, Zhang J, Yang C, et al. Synergistic optimization enables large-area flexible organic solar cells to maintain over 98% PCE of the small-area rigid devices[J]. Advanced Materials, 2020, 32(49): 2005153.

[4] Kimura H, Fukuda K, Jinno H, et al. High operation stability of ultra flexible organic solar cells with ultraviolet-filtering substrates[J]. Advanced Materials, 2019, 31(19): 1808033.

[5] Jiang Y, Dong X, Sun L, et al. An alcohol-dispersed conducting polymer complex for fully printable organic solar cells with improved stability[J]. Nature Energy, 2022, 7(4): 352-359.

[6] Han Y, Chen X, Wei J, et al. Efficiency above 12% for 1 cm^2 flexible organic solar cells with Ag/Cu grid transparent conducting electrode[J]. Advanced Science, 2019, 6(22): 1901490.

[7] Yang Y, Xu B, Hou J. Solution-processed silver nanowire as flexible transparent electrodes in organic solar cells[J]. Chinese Journal of Chemistry, 2021, 39(8): 2315-2329.

[8] Koo D, Jung S, Seo J, et al. Flexible organic solar cells over 15% efficiency with polyimide-integrated graphene electrodes[J]. Joule, 2020, 4(5): 1021-1034.

[9] Yang F, Huang Y, Li Y, et al. Large-Area Flexible Organic Solar Cells. npj Flexible Electron. 2021, 5, 30[J].

[10] Hashemi S A, Ramakrishna S, Aberle A G. Recent progress in flexible–wearable solar cells for self-powered electronic devices[J]. Energy & Environmental Science, 2020, 13(3): 685-743.

[11] Jeong E G, Jeon Y, Cho S H, et al. Textile-based washable polymer solar cells for optoelectronic modules: toward self-powered smart clothing[J]. Energy & Environmental Science, 2019, 12(6): 1878-1889.

[12] Ravishankar E, Booth R E, Saravitz C, et al. Achieving net zero energy greenhouses by integrating semitransparent organic solar cells[J]. Joule, 2020, 4(2): 490-506.

[13] Song W, Fanady B, Peng R, et al. Foldable semitransparent organic solar cells for photovoltaic and photosynthesis[J]. Advanced energy materials, 2020, 10(15): 2000136.

[14] Berny S, Blouin N, Distler A, et al. Solar trees: first large-scale demonstration of fully solution coated, semitransparent, flexible organic photovoltaic modules[J]. Advanced Science, 2016, 3(5): 1500342.