Applications of Ferroelectric Materials in the Field of Photovoltaics

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Abstract To solve the severe energy shortages, a variety of photovoltaic devices such as silicon solar cells, inorganic thin film solar cells, perovskite solar cells, dye-sensitized solar cells, organic solar cells and quantum dot solar cells, have been developed for the utilization of solar energy. Major studies focus on how to improve the photovoltaic performance of these solar cells. Some of ferroelectric materials can be spontaneously polarized and have spontaneous polarization dipole moment inside at room temperature. Recently, some of ferroelectric materials are introduced in these solar cells due to their special properties. They can be used not only as light absorbing materials in ferroelectric solar cells, but also as functional materials due to their ferroelectric characteristic. The measurable macroscopic electric field can be formed inside ferroelectric materials film that can be used as built-in electric field for photovoltaic devices to separate electron-hole pairs and facilitate their transport. Therefore, the introduction of ferroelectric materials into several important solar cells can accelerate the transport rate of photogenerated electrons, thus the short-circuit photocurrent of the cells can be increased and the light-to-electric conversion efficiency is also effectively improved. In this minireview, we summarized the applications of ferroelectric materials in the field of photovoltaics to improve their photovoltaic performance.

Keywords ferroelectric materials, solar cells, spontaneous polarization, photovoltaic performance

Introduction

In recent decades, the demand of non-renewable fossil energy such as coal and oil has been quickly increased. The energy shortage has become an important factor restricting the economic development. Many countries are looking for new energy for the economic development. At this time, rich, clean and easy-to-obtain light energy shows its importance. Therefore, solar energy is regarded as the most promising energy source.^[1-4] At present, extensive researches are being carried out on the development of photovoltaic devices. The main types of solar photovoltaic devices include silicon solar cells, dye-sensitized solar cells, perovskite solar cells, organic thin film solar cells, copper indium gallium selenide solar cells etc. In the development of these photovoltaic devices, in order to prepare solar cells with lower cost and higher conversion efficiency, ferroelectric materials have attracted much attention due to their special photoelectric conversion mechanisms.^[5-7]

In 1920, French Valasek discovered the ferroelectric effect in the rochelle salt, which opened the curtain for the study of ferroelectric materials.^[8] The ferroelectric thin films have an improved light response speed and a larger photo-generated current, thus improving the power conversion efficiency (PCE) of ferroelectric photovoltaic device.^[9] Therefore, the research on the photovoltaic properties of ferroelectric materials has gradually shifted from single crystals and ceramics to ferroelectric thin films. There are some crystals with ferroelectricity, which can be divided into four categories from the crystal structure of ferroelectric materials containing hydrogen bonds such as potassium dihydrogen phosphate (KH_2PO_4), oxygen octahedrons, fluorine-containing octahedrons and other ionic groups. These four oxygen-containing octahedron ferroelectric materials are most used.

Among them, perovskite-type ferroelectric materials are more used and their structure is ABO_3 such as BaTiO_3 , BiFeO_3 , and PbTiO_3 . In addition, the photovoltaic properties of $\text{Pb}(\text{Zr,Ti})\text{O}_3$ ferroelectrics have also been mostly studied.

The properties of ferroelectric materials can be evaluated by the hysteresis loop (Figure 1). At the microscopic level, ferroelectric materials have many polarons with positive and negative centers that do not coincide, and polarons with the same direction gather together to form ferroelectric domains. When the voltage is applied, the domain of the polarization along the direction of the electric field is enlarged, and the polarization degree of the material becomes large as shown by the curve in the OA section. The polarization (P) increases with the electric field (E) to point B, and P changes linearly with E (BC segment). When the electric field strength decreases, P does not fall along the original curve, but decreases along the CBPr curve. When E is zero, the polarization value is the

Figure 1 Schematic diagram of hysteresis loop.

remnant polarization (P_r). As the material is graded to zero, the reverse electric field strength is called as the coercive field (E_c).

Ferroelectric materials are capable of spontaneous polarization and have a measurable macroscopic electric field inside that can be used as built-in electric field for photovoltaic devices to separate electron-hole pairs and facilitate their transmission. This polarized electric field is the driving force of photocurrent in ferroelectric photovoltaic devices. The dipoles arranged in the ferroelectric material provide a promising way to control the barrier height, the offset of the interface energy or the width of the depletion region, photovoltaic effect of ferroelectric materials (Figure 2).^[10–12] This is crucial for all of these ferroelectric materials used in photovoltaic devices. Most of ferroelectric materials not only have the ferroelectric properties, but also absorb light to generate electron-hole pairs.

Figure 2 The photovoltaic effect of ferroelectric materials.^[12]

Ferroelectric Solar Cells

The overall properties of ferroelectric materials mainly affect the separation of electron-holes and the arrangement of energy bands affecting solar cells.^[13] The ideal photovoltaic material should have a band gap of 1.0–1.8 eV that matches the solar spectrum. However, traditional ferroelectrics are usually insulators with large band gap, which has relatively limited light absorption and photocurrent. Therefore, they are not suitable for photovoltaic applications. Zhang *et al.*^[14] reported a new type of ferroelectric material, KBiFe_2O_5 , which has narrower bandgap $E_g = 1.6$ eV. Internal polarization helps to separate photogenerated carriers and enhances the photovoltaic effect, thus having stronger photoelectric activity. The structure of the photovoltaic device is metal electrode/ KBiFe_2O_5 /metal electrode (Figure 3). This ferroelectric material has higher light absorption leading to higher carrier concentration, which contributes to the conversion of solar energy. Thus, open-circuit photovoltage (V_{oc}) and short-circuit photocurrent (J_{sc}) are 8.8 V and 15 mA/cm^2 , respectively.

Figure 3 Schematic diagram of the mechanism of photovoltaic device with the structure of metal electrode/ KBiFe_2O_5 /metal electrode.^[14]

Most of researches on ferroelectric thin film solar cells focus on the preparation of ferroelectric material thin films. In 2007, Pintilie *et al.*^[15] mainly studied the photovoltaic properties of a sandwich structure of metal/ferroelectric thin films/metal with $\text{Pb}(\text{Zr,Ti})\text{O}_3$ (PZT) as the ferroelectric layer, and the photoelectric properties conducted by PZT thin films was improved by adjusting the ratio of Zr and Ti. The Schottky potential existed on the surface of the PZT electrode, so that photocurrent signal could be detected. Then, Cao *et al.*^[16] studied ferroelectric thin film solar cell with the ITO/PZT/Pt structure. In order to improve the photoelectric performance, the n -type cuprous oxide (Cu_2O) layer was inserted between the PZT thin film and cathode Pt. The n - n heterojunction of $\text{Cu}_2\text{O}/\text{PZT}$ and the Schottky barrier of PZT/ITO provided favorable energy level alignment for efficient extraction of electrons on the electrode. The solar cell achieved J_{sc} of 4.80 mA/cm^2 , and the PCE is increased by 72 times to 0.57%. The ferroelectric solar cells with the structure of glass/ITO/PZT/ a -Si/Ag was prepared.^[17] It was found that the solar cell with such a structure facilitated the transmission of electrons photogenerated by ultraviolet visible light, thereby larger J_{sc} and PCE were obtained.^[17] The band gap of PZT ferroelectric film is 3.6 eV, a Si film with a narrow band gap (about 1.8 eV). Although the difference of band gaps between the Si film and the PZT film is large, the Si/PZT heterojunction can effectively enhance the photovoltaic output of solar cells based on ferroelectric PZT films.^[17] It has also been found that perovskite-structured methylammonium trihalide thin films has ferroelectric properties, and a novel ferroelectric solar cell was developed based on this structural thin film. The cell structure is $\text{TiO}_2/\text{MAPbI}_{3-x}\text{Cl}_x/\text{Au}$, which achieved a PCE of 6.7%. The output direction of V_{oc} and J_{sc} of the cell changed according to the polarization direction of the ferroelectric thin film. In addition, the maximum V_{oc} obtained by the cell depended on the coercive field of the ferroelectric materials.^[18]

Ferroelectric-Semiconductor Photovoltaics

Since the cell prepared by the ferroelectric materials could obtain large V_{oc} , the cell prepared by the semiconductor materials could obtain large J_{sc} , the combination of two materials has attracted extensive research interest. The ferroelectric BaTiO_3 thin films with tens of nanometers thickness could be used in the solar cell to have photovoltaic response, and the bulk photovoltaic effect measurement of the film reveals an increase in the photonic field and PCE, so BaTiO_3 was received extensive attention.^[19] Wang *et al.*^[20] studied ferroelectric material BaTiO_3 that was introduced into Si semiconductor material. In this solar cell, BaTiO_3 with field effect could separate carriers generated by silicon substrate to generate electricity. In addition, theoretical studies have shown that the electric field generated by the polarization of BaTiO_3 could diffuse into the silicon base, and the adjustment of relevant parameters could optimize the electric field, thereby improving the photoelectric performance of the solar cell.

Immediately after, Liu *et al.*^[21] studied the use of conventional semiconductor material silicon with good capability of carrier transport as the light absorbing layer. They deposited a layer of BaTiO_3 on the semiconductor to provide an external electric field for separating carriers generated by the Si (Figure 4). This ferroelectric material in silicon solar cells without p - n junction was not used for light absorption and provided only external electric field. The anode and the cathode on the semiconductor layer after the BaTiO_3 polarization collected electrons and holes, respectively. The electrons are combined with the holes at the cathode through the external circuit to form complete loop.

Figure 4 Structure diagram of the Si/BaTiO₃ solar cell.^[21]

Dye-Sensitized Solar Cells Based on Ferroelectric Materials

Dye-sensitized solar cells (DSSCs) have attracted much attention due to their low cost, no pollution to the environment, extensive materials to fabricate solar cell. In 1999, Grätzel *et al.*^[22] synthesized mesoporous nanoparticles SiTiO₃ by improving the reported method and prepared thin films deposited on conductive glass in dye-sensitized solar cells. It was found that V_{oc} of the cell based on SiTiO₃ is 100 mV higher than that of the cell based on TiO₂. For lower J_{sc} and IPCE, the surface of SrTiO₃ selectively changed first by pH treatment, thereby enhancing the adsorption of dyes and improving the J_{sc} and IPCE, which provided basis for further optimizing the performance of dye-sensitized solar cells.

BaTiO₃ is a ferroelectric material that has been widely used in cells for several years. Zhong *et al.*^[23] mainly studied the application of BaTiO₃/TiO₂ (4) electrode in dye-sensitized solar cells. It was found that DSSCs with BaTiO₃/TiO₂ (4) electrode has higher PCE than that based on the conventional TiO₂ electrode. Because BaTiO₃ is a high dielectric ferroelectric material (although it has the similar structure with TiO₂), the electron lifetime of DSSCs with BaTiO₃/TiO₂ (4) electrode was significantly improved, and the recombination reaction at the TiO₂ photoelectrode/electrolyte interface was reduced. The interface charge recombination was reduced, so that solar cells with better photovoltaic performance were obtained.

Yu *et al.*^[24] synthesized pure phase perovskite-type SrTiO₃, CaTiO₃ and BaTiO₃ powders by the solid phase reaction. The thin films were prepared to be used in DSSCs. It was found that the cells prepared by the three products obtained higher V_{oc} than the cell based on the pure TiO₂. The higher V_{oc} is due to the fact that the difference between the Fermi level of the oxide and the redox potential of the electrolyte could be larger than that for TiO₂. From another aspect, they prepared TiO₂/SrTiO₃ and TiO₂/BaTiO₃ composite electrodes for DSSCs. When the doping of SrTiO₃ and BaTiO₃ reached 30%, the highest PCE was achieved, which is more efficient than the cell constructed by pure TiO₂. In 2015, Yu *et al.*^[25] continued to study DSSCs constructed with TiO₂ as the first layer, SrTiO₃ or BaTiO₃ as the second mesoporous double-layer electrode. The electrode constructed by the mesoporous structure of SrTiO₃ and TiO₂ could increase the adsorption amount of dye. Since SrTiO₃ or BaTiO₃ was deposited on the upper layer of TiO₂ (Figure 5), the electron-hole recombination at the interface between the first layer and the second layer could be suppressed. In addition, the conduction band of SrTiO₃ and BaTiO₃ is more negative than that of TiO₂, and the matching of band structure improved the electron lifetime. Both of these aspects could improve V_{oc} and PCE of the solar cell.

Figure 5 Schematic illustration of the double-layer electrode.^[25]

Xie *et al.*^[26] fabricated composite structure of BaTiO₃ ferroelectric material and TiO₂ as photoanode, and the remnant polarization of the composite thin film was 105 $\mu\text{C}/\text{cm}^2$, indicating that BaTiO₃/TiO₂ (4) still has very good ferroelectric performance. In the composite structure, BaTiO₃ provided polarized electric field, and TiO₂ encapsulated in the outer layer received electrons generated by the light-exciting dye and provided the path of electron transport. The polarization of the BaTiO₃ thin films enhanced the adsorption interaction of dye in the cell. The cell has J_{sc} of 14.44 mA/cm^2 and PCE of 7.29%. The report of this work led to the study of the use of ferroelectric materials in photoelectric devices to improve their performance and deepen the understanding of charge transport and photovoltage generation mechanisms.

Perovskite Solar Cells Based on Ferroelectric Materials

BiFeO₃ is a typical ferroelectric thin film material, and BiFeO₃ crystal has photoelectric and photoconductive properties when it is in contact with gold electrode. It has been reported that polycrystalline BiFeO₃ ferroelectric thin film material was deposited on conductive glass substrate by the chemical solvent DMF modification method, and the modified BiFeO₃ thin films as light absorbing material could absorb more visible light and has narrower band gap.^[27,28] Liao *et al.*^[29] found that (benzylammonium)₂PbCl₄ is a ferroelectric and has ferroelectric properties. At the same time, different metal cations and halides in the perovskite could be adjusted to change the band gap for using in the photovoltaic field. It provided new methods and ideas for the development of new ferroelectric semiconductors. In 2015, Chen *et al.*^[30] proved that CH₃NH₃PbI₃ has ferroelectric properties. In the solar cell composed of perovskite as light absorbing material, when positive electrode electric field was applied on the electrode, the arrangement of ferroelectric domains was arranged in the preferred direction to make the separation and rush through of photogenerated electrons, moreover, the cell obtained the highest PCE and J_{sc} under the condition of +2 V polarization. Subsequent control experiments further showed that the polarization and scanning direction also showed significant impact on the photovoltaic performance of 0.25BaTiO₃—0.75BiFeO₃ ferroelectric solar cells. This indicated the prevalence and universality of this behavior in solar cells based on ferroelectric materials. The application of ferroelectric materials provides a new perspective for adjusting the working mechanism and photovoltaic performance of perovskite solar cells.

Morris *et al.*^[31] used transient absorption spectroscopy (TAS) to study the carrier kinetics of BaTiO₃ ferroelectric materials. In

the absence of applied electric field, the dipoles presenting inside of BaTiO₃ could prevent electron-hole recombination and obtained longer lifetimes carriers, which proved that ferroelectric materials are indeed important for improving the PCE of solar photovoltaic devices. Okamoto *et al.*^[32] studied the application of BaTiO₃/TiO₂ as double mesoporous electron transport layer in perovskite solar cells. By comparing the effects of different thicknesses of BaTiO₃ thin films on the photovoltaic performance of cells, it was found that PCE increased from 9.89% of the TiO₂ mesoporous monolayer to 12.4%, when two layers of BaTiO₃ were spin-coated to form the structure (Figure 6). The crystal size of CH₃NH₃PbI₃ on the double mesoporous layer is usually larger, while the larger CH₃NH₃PbI₃ crystal could absorb light and improve the photocurrent, thereby enhancing the photovoltaic performance of the solar cell.

Figure 6 *J-V* curves of solar cells with the structure diagram as shown in inset.^[32]

Organic Solar Thin Film Solar Cells Based on Ferroelectric Materials

Yang *et al.*^[33] embedded poly(vinylidene fluoride- trifluoroethylene) (P(VDF-TrFE)) in organic solar cell (substrate/ITO/PEDOT:PSS/P₃HT/P(VDF-TrFE)/PCBM/Al). The polarizing layer film could adjust the LUMO difference between the donor and the acceptor and reduce the LUMO energy level difference to optimum value, which is beneficial to electron transport. After the ferroelectric thin film was forward polarized, V_{oc} and J_{sc} of the cell were 0.67 V and 9.0 mA/cm², respectively. Therefore, both V_{oc} and J_{sc} of the cell could be improved.

Then, Asadi *et al.*^[34] studied the organic solar cells doped with ferroelectric thin films, and compared the cells of P(VDF-TrFE), TrFE, and LiF. After a series of optimized tests, it was found that all three materials contained fluoride. It was speculated that this substance should play an important role in cell performance. Through this study, it was learned that the introduction of P(VDF-TrFE) ferroelectric materials into the solar cells was mainly due to the fluoride inside the materials. Xiao *et al.*^[35] prepared P(VDF-TrFE) nanoparticles by the solution chemistry and applied them to ferroelectric organic photovoltaic devices. The cell structure is ITO/PEDOT:PSS/PCDTBT:PC70BM/P(VDF-TrFE)/Al. By adjusting the size of P(VDF-TrFE) NPs and the crystallinity of the crystal to control the coverage of the intermediate layer of the ferroelectric thin films, PCE of the cell could be improved. After P(VDF-TrFE) thin film was forward polarized, the photovoltaic performance parameters of the device were greatly improved. Moreover, V_{oc} , J_{sc} and PCE of the cell were 0.91 V, 11.3 mA/cm² and 6.64%, respectively. The applications of ferroelectric materials provide a new perspective for adjusting the working mechanism and photovoltaic performance of perovskite solar cells.

Conclusions and Perspectives

Ferroelectric materials have attracted extensive attention due to their special photoelectric conversion mechanism. Since the ferroelectric solar cells show large V_{oc} and the semiconductor solar cells usually give large J_{sc} , the combination of the two materials will attract extensive research interest. Some of ferroelectric materials have the properties of semiconductors, so they can be used as light absorbing materials in ferroelectric solar cells to generate electron-hole pairs. At the same time, some of ferroelectrics can be spontaneously polarized and make measurable macroscopic electric field inside, which can separate electron-hole pairs and promote their transmission. These ferroelectric materials can accelerate the transport rate of electron-holes, therefore, ferroelectrics will be widely introduced as different functional structures in semiconductor solar cells to improve the photoelectric performance of these cells. In addition, in perovskite ferroelectric materials, different metal cations and halides could be adjusted to change their band gap for using in the photovoltaic field. It provided new methods and ideas for the development of new ferroelectric semiconductors used in solar cells. Rational introduction of ferroelectric material structure in solar cells opens a new way to further improve their photovoltaic performance.

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Conflict of Interest

The authors declare no conflict of interest.

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