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Design and Preparation of Bending-Resistant Flexible All-Solid Dye-Sensitized Solar Cells

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Abstract: All-solid-state flexible dye-sensitized solar cells will not only expand the application scenarios of solar cells but also significantly extend the lifetime of solar cells. However, improving their bending-resistant ability is still a great challenge. In this study, a bending-resistant flexible all-solid dye-sensitized solar cell was designed and prepared. Firstly, for the preparation of TiO₂ photoanode, the traditional nano-sized film has been replaced by dual-porous film with both nano and submicron pores, which can not only benefit the filling of the electrolyte but also supply the space for stress release. Secondly, for the filling of the Poly(vinylidene fluoride)/Poly(ethylene oxide)-based electrolyte, the solvent is removed by a vacuum method, and the electrolyte fibers forming in the submicron pores also show the potential for stress release. Lastly, combined with the advantages of the dual-porous TiO₂ film and the fast evaporation of the polymer electrolyte, the conversion efficiency of the solar cells remains constant after the 20,000 bending times. The study supplies a demonstration for the development of all-solid-state flexible dye-sensitized solar cells.

Keywords: dye-sensitized solar cell; bending-resistant; dual-porous structure; all solid state



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1. Introduction

Dye-sensitized solar cells (DSCs) offer a technically and economically alternative concept to current p-n junction photovoltaic devices [1–4]. Different from the semiconductor in p-n junction cells, which performs both light absorption and carrier transport, the light is absorbed by the sensitizer on the surface of the wide band gap semiconductor in DSC, and the charge is separated at the interface between the sensitizer and the semiconductor, and then the carrier flows to the external circuit through the conduction band of the semiconductor. This innovative structure enables the independent development of photosensitizers and semiconductor materials [5] and even provides a prototype basis for quantum dot solar cells and perovskite solar cells [6,7].

The flexibility of DSCs further reduces the manufacturing cost and shows the advantages of bend resistance and suitability for rollable production. Several stable methods have been proposed for the low-temperature preparation of the TiO₂ semiconductor mesoporous layers, such as low-temperature spin coating, low-temperature hydrothermal method, film transfer method, mechanical film pressing method, and vacuum cold spraying method [8,9], and some bending-resistant data of the TiO₂ films have also been reported. Miyasaka et al. [10] prepared the TiO₂ film through electrophoretic deposition, and the curvature of the TiO₂ film was 0.2 mm^{−1}. Murakami et al. [11] prepared the TiO₂ film through chemical vapor deposition, and the bending radius of the TiO₂ film was 5 mm. Kijitori et al. [12] and Chen et al. [13] prepared the TiO₂ film using a low-temperature sintering method and mechanical film compression method separately, and both of the TiO₂ films showed a

good bending-resistant ability. However, the above original research only described the bending-resistant ability of TiO_2 thin films, and the bending-resistant ability of the whole solar cells was not involved. The flexible DSC as a stacking system is composed of several functional layers, and when it is under an action of bending behavior, in addition to the neutral layer, most of the layers will produce bending stress due to the bending strain. According to the conventional conclusions of engineering mechanics, the different areas within each functional layer along the stress-loading direction will show different strain states, and every strain state may have a serious effect on the performance of the solar cells. Then, the bending behavior of the whole cell becomes increasingly important because it is highly realistic. Dai et al. [14] prepared the TiO_2 film by cold isostatic pressing method, and after the TiO_2 film was assembled into solar cells, the TiO_2 film did not crack after 500 bending times. He et al. [15,16] established a standard to trace the bending-resistant ability of the solar cells, including bending modes, bending radius, and bending times, focusing on establishing the relationship between the structural change and performance evolution of the solar cells under various bending actions. Their research group pointed out that there was a critical bending radius, and when the bending radius was larger than the critical bending radius, the photovoltaic performance of the solar cells improved; otherwise, when the bending radius was smaller than the critical bending radius, the photovoltaic performance of the solar cells decreased with an increase in bending times due to the cracking and spalling of TiO_2 films. The above achievements in liquid solar cells have laid a good foundation for the research of all-solid-state solar cells.

With the development of the low-temperature preparation method for the TiO_2 film and the evaluation mechanism of bending-resistant ability for the whole solar cells, the researchers put the solidization of the flexible DSCs into routine. In addition to the retaining advantages of low manufacturing costs, bend resistance, and roll-to-roll production, the flexible all-solid-state DSCs will significantly extend their lifetime due to the elimination of electrolyte leakage problems. Several systems of solid-state electrolyte materials with strong ion transport ability have been proposed, such as inorganic salt system, (Polyvinylidene fluoride)/(Polyethylene oxide) (PVDF/PEO) system, and spirodifluorene system [17–19]. Among them, PVDF/PEO-based solid-state electrolyte is low cost, and its unique long-chain plastic molecular structure shows inherent bending-resistant characteristics. More significantly, Li et al. [20] reported that when the PVDF/PEO-based electrolyte was infiltrated into the large pores, the electrolyte fibers appeared across large pores because of the quick removal of the solvent. The electrolyte fibers can not only promote I^-/I_3^- migration but can also be used as elastic material in TiO_2 ceramic framework to improve the bending-resistance ability. The research results of liquid DSCs show that the TiO_2 film stacked by submicron particles has a larger specific surface area and better light scattering, and the submicron pores in the structure will effectively promote the filling of polymer electrolyte [21] and accelerate the diffusion coefficient of electrolyte [22,23]. In this study, we try to replace the classical nano-porous TiO_2 films with dual-porous TiO_2 film, which contains both submicron pores and nanopores, and the solvent in the electrolyte is removed quickly by a vacuum drying method, which not only promotes the filling of polymer electrolyte but also provides the space for stress release. We attempt to reach a bending-resistant flexible all-solid dye-sensitized solar cells through the designed microstructure of photoanode including dual-porous TiO_2 film and the polymer electrolyte.

Therefore, the P25 and Polyethylene glycol (PEG) have been chosen to prepare the micron-sized composite clusters through composite perforation, grinding, screening, and high-temperature annealing, and then the composite clusters are stacked into dual-porous TiO_2 film through vacuum cold spraying (VCS). Subsequently, when the PVDF/PEO-based electrolyte solution is infiltrated into the dual-porous film, the solvent is quickly removed through vacuum drying. The combination of the dual-porous TiO_2 structure and the filling state of the PVDF/PEO electrolyte will effectively alleviate the stress concentration during the bending of the solar cells. At last, the photovoltaic performance and charge

transportation and recombination behavior of the solar cells during the bending process are systematically tracked to establish the correlation between structure and performance.

2. Materials and Methods

2.1. Materials

The main materials include P25 powder (Degussa, Munich, Germany) and SiO₂ powder (Degussa, Germany), ITO–PEN substrate (20 Ω·cm², Peccell, Yokohama, Japan), Di-tetrabutylammonium cis-bis(isothiocyanate)bis (2,2'-bipyridyl-4,4'-dicarboxylato) ruthenium(II) (N719, Solaronix, Aubonne, Switzerland), LiI and I₂ (ACROS, Geel, Belgium), N,N-Dimethylformamide (DMF, ACROS, Belgium), Acetonitrile (ACN, Kemiou, Tianjin, China), PEG ($M_w = 10,000$, ALDRICH, London, UK), PVDF ($M_w = 180,000$, ALDRICH, UK), and PEO ($M_w = 600,000$, ACROS, Belgium). These materials are not further purified during the experiment.

2.2. Preparation and Characterization of Dual-Porous TiO₂ Film

Firstly, a TiO₂ suspension is prepared by adding 50 g of P25 and 10 g of PEG into 250 mL of anhydrous ethanol and then stirring at 70 °C for 30 min. Secondly, TiO₂-PEG composite clusters with a PEG content of 16.7% are obtained through rotary evaporation to remove the solvent, followed by ball milling for 8 h to reduce the cluster size and sieving with an 800-mesh screen to remove the large clusters. Thirdly, the TiO₂-PEG composite powder was annealed at 450 °C for 30 min to remove PEG, and then the pure TiO₂ composite clusters with micron-scale size were received. Lastly, dual-porous TiO₂ films were prepared by VCS technology [24–26], and the typical spraying parameters had a scan rate of 25 mm·s^{−1}, spraying distance of 12 mm, scan interval of 5 mm, and a vacuum chamber pressure of 0.4 KPa. The large pores in the dual-porous TiO₂ film are characterized by electroless nickel plating. The morphology of TiO₂ films is characterized by a scanning electron microscope (SEM, TESCAN, Brno, Czech Republic).

2.3. Preparation and Filling of the PVDF/PEO-Based Electrolyte

Firstly, the polymer electrolyte can be obtained by dissolving 1.44 g of I₂, 9.52 g of LiI, 0.45 g of SiO₂, 3.36 g of PEO, and 5.05 g of PVDF into 150 mL of DMF/ACN, where the volume ratio of DMF and ACN is 7:3. Secondly, the dual-porous TiO₂ film is soaked in N719 dye solution with the concentration of 3×10^{-4} mol·l^{−1} and stored at 60 °C for 12 h, and then the unadsorbed dye is washed off using anhydrous ethanol. Lastly, the polymer electrolyte is dropped onto the dual-porous film and dried with a homemade vacuum machine under 0.01 MPa at 80 °C [15]. The morphology of TiO₂ film filled with polymer electrolytes is also characterized by SEM.

2.4. Assemble of Solar Cells and Characterization of the Bending-Resistant Ability

The blank part of the ITO–PEN substrate is covered with the invisible tape leaving the 10 × 10 mm² area. The Pt metal is sputtered with an ion sputtering instrument (E-1045, HITACHI, Tokyo, Japan) for about 10 min to form the Pt counter electrode. A bending test machine [20,22] is used to perform a bending experiment, and the typical external bending mode has a bending radius of 8 mm and a bending speed of 100 times/min. The solar simulator (Solar 94023A, Newport, UK) and its supporting test system (Keithley 2400) are used to test the photovoltaic performance of the solar cells under the irradiation condition of AM 1.5 G. Electrochemical workstations (Solartron 1260 wideband frequency analyzer and Solartron 1287 constant potentiometer, AMETEK, Pittsburgh, PA, USA) are used to test the charge transportation and recombination behavior of the solar cells.

3. Results and Discussion

3.1. The Formation of the Submicron Pores in Dual-Porous Film

The morphology of the nano-sized P25 powder is shown in Figure 1a. When it is sprayed directly into TiO₂ film, it usually shows a pore size of ~30 nm based on the nitrogen

de-adsorption data [27], which not only harms the infiltration of the PVDF/PEO-based electrolyte but also hinders the stress release. According to the principle of graded assembly, the larger pores are reached using stacking powders with larger sizes. Therefore, a certain amount of PEG has been added into the nano-sized P25 powder by mixing materials to form the agglomerates, grinding to decrease the cluster size, screening to eliminate the larger cluster, and high-temperature annealing to remove PEO; then, composite TiO_2 clusters with an average size of $5\ \mu\text{m}$ were obtained, as shown in Figure 1b. When it is sprayed into dual-porous films, the cross-sectional view of the TiO_2 film is shown in Figure 1c. It can be seen that the thickness of the film is uniform, about $16.5\ \mu\text{m}$, and there is an obvious sense of the concavo-convex morphology based on the cross-sectional views of the film, which may be attributed to the submicron pores in the dual-porous film. A high magnification SEM morphology data of the dual-porous film is shown in Figure S1 in the Supporting Information; as can be seen, the nanopores in the cluster still exist, which will benefit the infiltration of the polymer electrolyte.

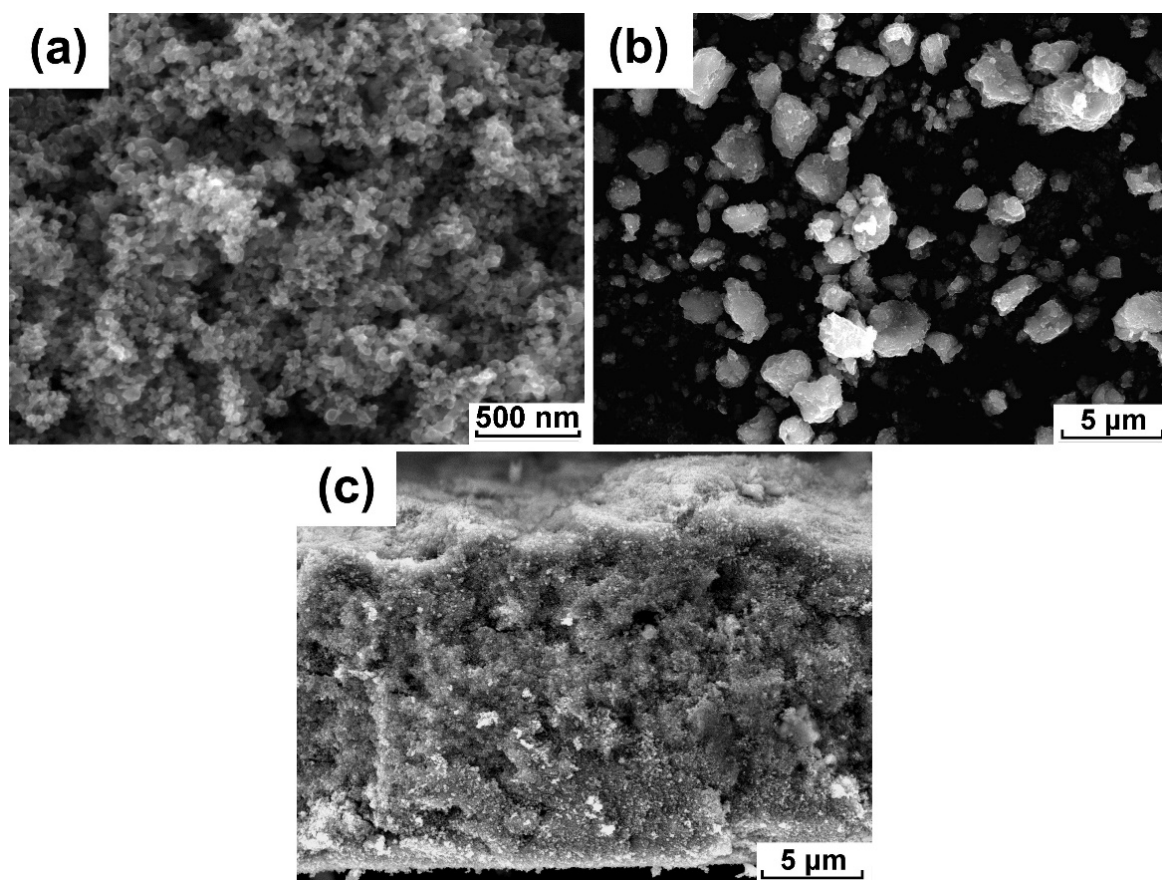


Figure 1. The microstructure of the powder and composite film: (a) the pristine P25 powder, (b) the composite cluster after annealing, and (c) the cross-sectional view of the dual-porous film.

To verify that the concavo-convex morphology is caused by submicron pores in the film, the electroplate nickel method is used to fill the nickel into submicron pores. Based on the distribution position and particle size of the nickel particles in the dual-porous film, the submicron pores can be forecasted. The cross-sectional view of the dual-porous film after electroplate nickel is shown in Figure 2. It can be seen that white nickel particles appear in the entire dual-porous film, and the size of most nickel particles is in the submicron scale. Therefore, it can be concluded that the dual-porous film contains both submicron pores and nanopores.

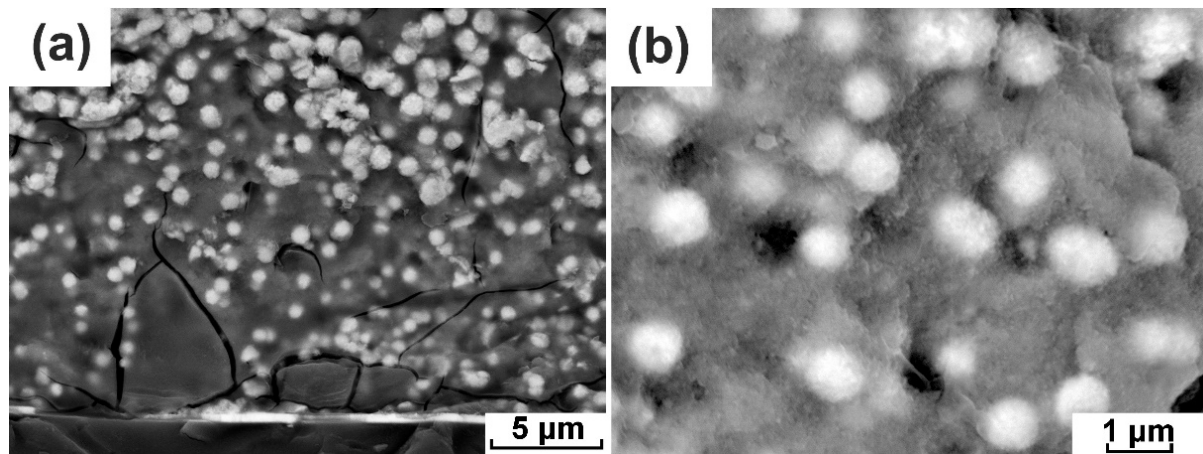


Figure 2. The cross-sectional view of the dual-porous film after electroplate nickel: (a) distribution and (b) particle size.

3.2. The Formation of PVDF/PEO Electrolyte Fibers in Submicron Pores

After the polymer electrolyte is infiltrated into the dual-porous TiO_2 film by the solution method, the solvent is quickly removed under vacuum conditions. The dual-porous TiO_2 film filled by polymer electrolyte was observed as shown in Figure 3; as it can be seen, the electrolyte is uniformly coated on the surface of the TiO_2 powder. In addition, there are electrolyte fibers in the submicron pores. The diameter of electrolyte fibers is about 100 nm and the length is in the micron scale, and it is expected to be used as a toughening material in the ceramic TiO_2 film to improve its bending resistance.

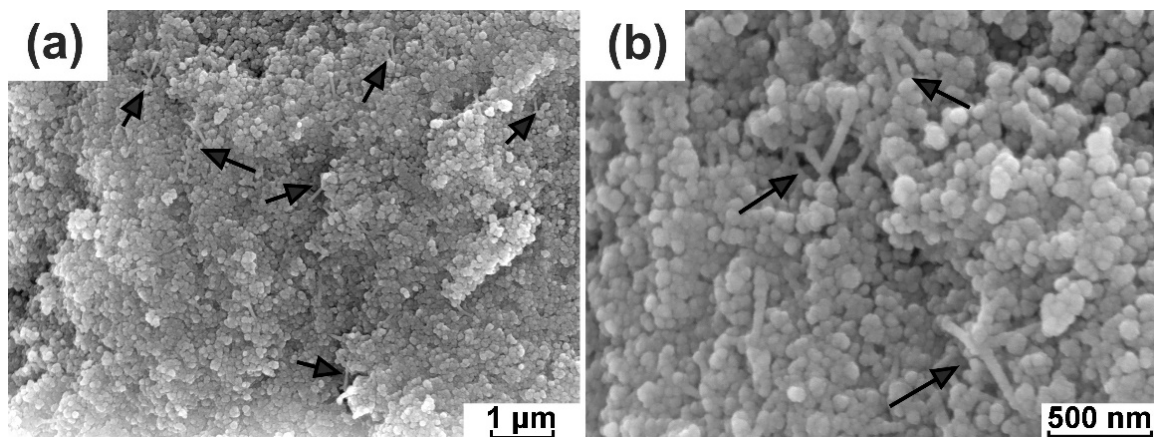


Figure 3. The cross-sectional view of the dual-porous film after being filled with PVDF/PEO-based electrolyte: (a) low magnification and (b) high magnification, and the obvious electrolyte fibers are marked with black arrows.

The formation reasons for the observed electrolyte fibers are shown in Figure 4. Firstly, when the PVDF/PEO-based electrolyte is dropped onto the dual-porous TiO_2 film, the submicron pores are infiltrated with the electrolyte solution (Figure 4a). Secondly, during the drying process, along with the evaporation of the solvent, the PVDF/PEO solute gradually precipitates and shrinks to the surface of the TiO_2 film to relieve the pressure of saturation (Figure 4b). Thirdly, due to the rapid solvent evaporation through the vacuum drying method, part of the solute does not have enough time to diffuse to the TiO_2 surface but is solidified in the submicron pore as a fiber, and then the electrolyte fibers across the submicron pores are formed (Figure 4c).

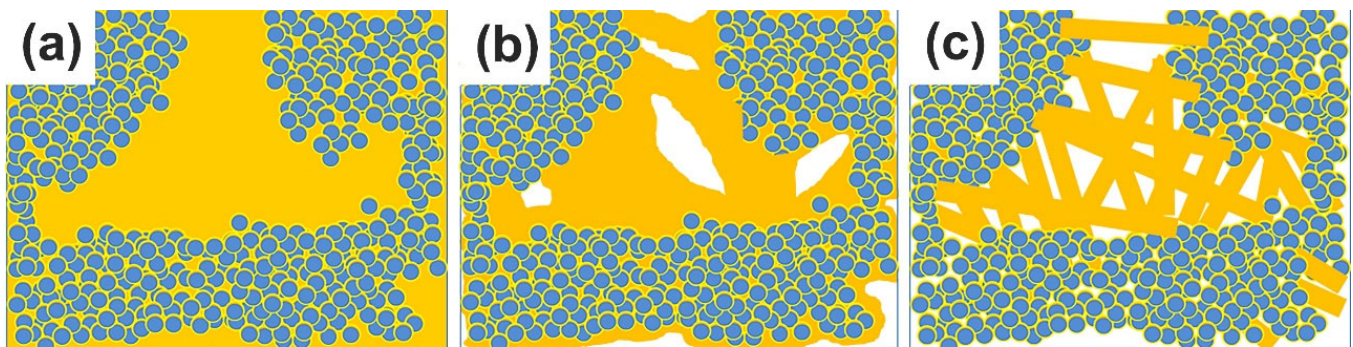


Figure 4. The formation process of the electrolyte fiber on dual-porous film: (a) the electrolyte solution fully infiltrates the submicron pore, (b) the PVDF/PEO solute shrank to the TiO_2 film along with the evaporation of the solvent, and (c) the electrolyte undergoes in-situ solidification into a fiber because of the fast evaporation of the solvent. The blue particles represent TiO_2 and the electrolyte filling in the TiO_2 film is represented by yellow.

3.3. The Bending-Resistant Ability of the Flexible All-Solid-State Dye-Sensitized Solar Cell

After the flexible all-solid-state DSCs are assembled, the bending-resistant ability of the solar cells is tested. The short-circuit current density (J_{sc}), open-circuit voltage (V_{oc}), fill factor (FF), and conversion efficiency (η) of five cells with the same bending process are detected. Figure 5 shows the photovoltaic performance of the solar cells at the recovery state after 0, 1000, 2000, 5000, 10,000, and 20,000 bending times, and the bending radius is chosen as 8 mm during the whole testing process. It can be seen that the original performance data are V_{oc} of 0.57 V, J_{sc} of $2.81 \text{ mA}\cdot\text{cm}^{-2}$, FF of 0.30, and η of 0.49%; and during 20,000 bending times, the J_{sc} , V_{oc} , FF, and η all show a certain amount of change because of the change in the inner structure. When the parameters in J_{sc} , V_{oc} , FF, and η are separately shown according to the bending times in Figure 6, it can be seen more intuitively that the V_{oc} and FF of the solar cells increase slightly, while the J_{sc} of the solar cells continue to decrease with an increase in bending times. At last, the η almost remains constant during the 20,000 bending processes. Compared with the liquid solar cells that have been reported, the flexible all-solid DSCs show good bending-resistant ability.

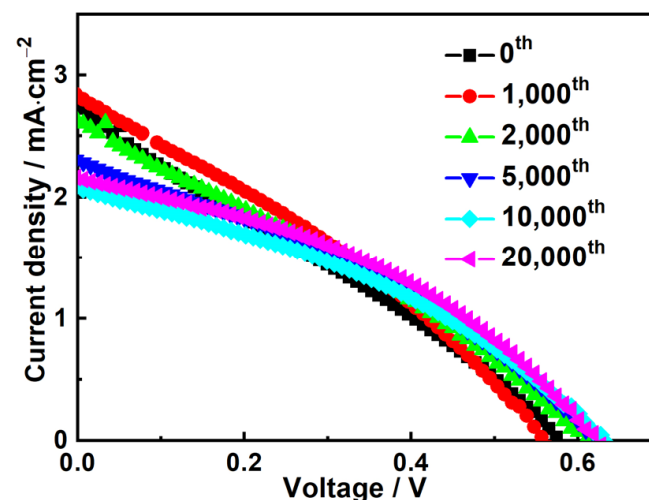


Figure 5. The photovoltaic performance of the flexible all-solid-state solar cell under the bending times of 0th, 1000th, 2000th, 5000th, 10,000th, and 20,000th.

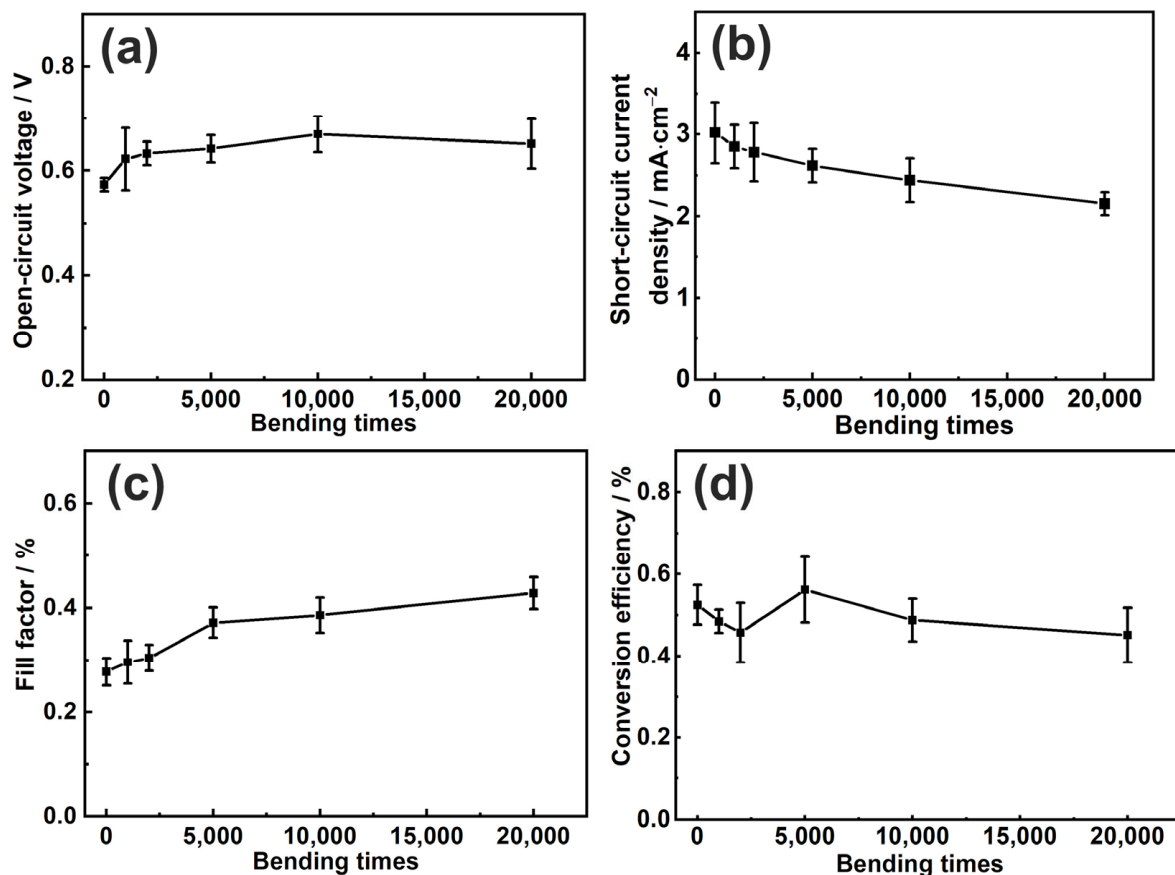


Figure 6. The variation tendency of the photovoltaic parameters for an average of 5 solar cells with increasing bending times: (a) V_{oc} , (b) J_{sc} , (c) FF, and (d) η , where V_{oc} is short for open-circuit voltage; J_{sc} is short for short-circuit current density; FF is short for fill factor, and η is short for conversion efficiency.

To find out why there is a certain amount of change for the photovoltaic parameters during the bending processes, the electrochemical impedance spectroscopy (EIS) of the solar cell with increasing bending times is tested at the V_{oc} under illumination. Figure 7 shows the Nyquist plots of the flexible all-solid-state DSCs under the conditions of 0, 1000, and 20,000 bending times, respectively. There are three semicircles in the impedance curves from the high-frequency zone (10^5 – 10^3 Hz) to the middle-frequency zone (10^3 – 10^1 Hz) and the low-frequency zone (10^1 – 10^{-1} Hz) from left to right; the catalytic reduction ability of Pt electrode is represented by the semicircle at the high-frequency zone, the electron transportation and recombination ability of $\text{TiO}_2/\text{N719}/\text{electrolyte}$ are represented by the semicircle at the middle-frequency zone, and the charge diffusion ability of I^-/I_3^- for the electrolyte is represented by the semicircle at the low-frequency zone [28,29]. It can be seen from Figure 7 that with an increase in bending times, there is no significant change for the semicircles at the high-frequency and middle-frequency zones, whereas the semicircle at the low-frequency zone shows an obvious change.

A generalized equivalent circuit model [30–32] shown in the inset of Figure 7 is employed to fit the EIS spectrum, the detailed data are shown in Table 1, where the R_{pt} represents the catalytic resistance of the Pt electrode; R_t represents the transport resistance of electrons in the TiO_2 film; R_{ct} represents the charge recombination resistance at the $\text{TiO}_2/\text{N719}/\text{electrolyte}$ interface, and R_w represents the diffusion resistance of I^-/I_3^- in the polymer electrolyte. It can be seen with increasing bending times that R_{pt} , R_t , and R_{ct} are almost constant; however, the charge diffusion resistance R_w increases from 1280 Ω to 1500 Ω , which indicates that the structure of Pt electrode, TiO_2 film, and the $\text{TiO}_2/\text{N719}/\text{electrolyte}$ is stable, whereas the structure of the polymer electrolyte may be

deteriorated. That is to say, during the bending process, the photoanode of the solar cells is stable due to the increasing bending-resistant ability coming from the designed dual-porous TiO_2 film and polymer electrolyte filling state. Though bending causes changes in the electrolyte, based on the research on liquid flexible DSCs, we will try to manipulate the neural layer at the electrolyte region to achieve bending resistance for the entire structure in the future.

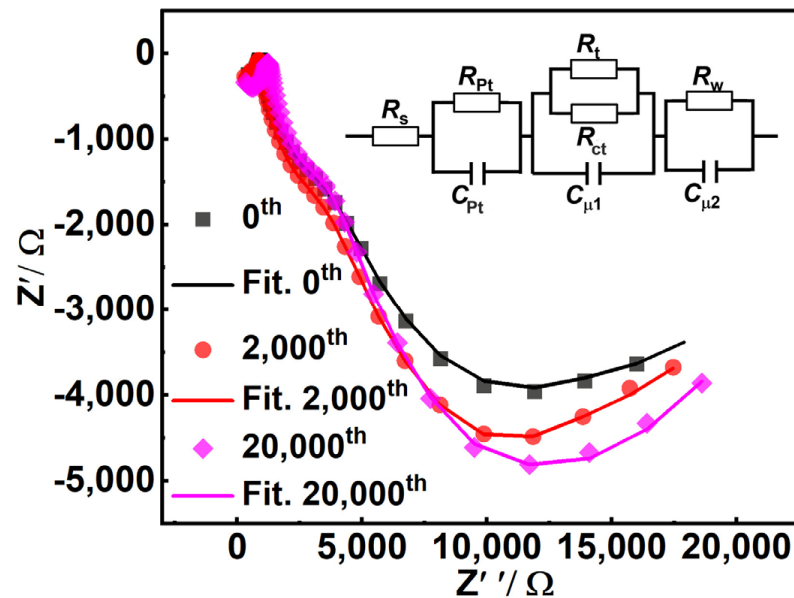


Figure 7. The typical Nyquist plots of the flexible all-solid-state DSCs under bending times of 0, 2000, and 20,000 at V_{oc} under illumination. The dots are the pristine data, and the solid lines are the fitting results based on the equivalent circuit model shown in the inset. In the equivalent circuit model, R_s is the series resistance including the sheet resistance of the whole cell; R_{Pt} and C_{Pt} are the charge transfer resistance and electrical double-layer capacitance at the Pt/electrolyte interface; R_t is the transport resistance of the electrons in TiO_2 film, and R_{ct} is the charge transfer resistance of electrons at the TiO_2 /dye/electrolyte interface; $C_{\mu 1}$ is the chemical capacitance of TiO_2 film; R_w and $C_{\mu 2}$ are the charge diffusion resistance and chemical capacitance of the electrolyte.

Table 1. The detailed Nyquist plot data of the flexible all-solid-state DSC after bending times of 0, 2000, and 20,000.

Bending Times	R_t/Ω	$R_{ct}/\times 10^3 \Omega$	$R_w/\times 10^3 \Omega$
0	10.7	1.79	1.28
1000	11.9	1.80	1.35
20,000	11.0	1.79	1.50

4. Conclusions

In this study, the dual-porous TiO_2 film is obtained by spraying micron-sized composite clusters to replace the traditional nano-porous film, and besides the nanopores, the nickel-plating results show that there are submicron pores. After the polymer electrolyte is infiltrated into the dual-porous film, the solvent is removed quickly by vacuum drying, and then the PVDF/PEO-based electrolyte is not only coated on the surface of the TiO_2 particles but also forms fibers in the submicron pores. When the designed photoanodes, including the dual-porous TiO_2 film and the polymer electrolyte filling state, are assembled into flexible all-solid-state DSCs, the solar cells show a bending-resistant ability during the bending process. The total conversion efficiency of the designed solar cells is almost constant during the 20,000 bending times because of the improved stress release ability, and the EIS results show that the change in the electrolyte structure may result in a change

in individual photovoltaic parameters. The research illustrates that increasing the stress release space (submicron pores) and introducing elastic material (polymer electrolyte and electrolyte fiber) in the TiO₂ film improves the bending-resistant ability of the photoanode of the all-solid-state DSCs.

Supplementary Materials: The following are available online at <https://www.mdpi.com/article/10.3390/coatings14040504/s1>, Figure S1: The high magnification SEM morphology data of the dual-porous film.

Author Contributions: Conceptualization, Y.L. and W.-W.D.; methodology, Y.L.; validation, Y.L., Y.-X.H. and W.-W.D.; formal analysis, Y.-X.H. and L.L.; investigation, Y.-X.H. and L.L.; resources, Y.L. and W.-W.D.; data curation, L.L., J.-H.C. and X.G.; writing—original draft preparation, Y.L.; writing—review and editing, Y.L. and W.-W.D. All authors have read and agreed to the published version of the manuscript.

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