Polydopamine-Modified Electrospun Polyvinylidene Fluoride Nanofiber Based Flexible Polymer Gel Electrolyte for Highly Stable Dye-Sensitized Solar Cells

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ABSTRACT: To overcome the drawbacks of solvent evaporation and leakage from the liquid electrolyte in dye-sensitized solar cells (DSSCs), a polymer gel electrolyte (PGE), based on polydopamine (PDA)-modified electrospun polyvinylidene fluoride (PVDF) nanofibers, PDA@PVDF, was prepared and applied in quasi-solid DSSCs (QS-DSSCs). The PDA coating increased the wettability of the liquid electrolyte by improving the distribution of the ionic liquid electrolyte. This, in turn, enhanced the ion conductivity of the PGE as well as the stability of QS-DSSC. The PDA@PVDF nanofiber membrane exhibited a satisfactory ultimate tensile strength of 9.3 MPa with a failure elongation of 130%. Such high toughness provided excellent mechanical support for the PGE. Owing to the unique pore connected structure of nonwoven nanofibers as well as the reduced surface tension with the liquid electrolytes, the PGE-based QS-DSSC retained 95.7% of the short circuit current from the liquid-electrolyte-based DSSC and showed an energy conversion efficiency of 8.26%. No significant impacts were observed for the open-circuit voltage and fill factor. More importantly, the QS-DSSCs using PDA@PVDF-based PGE showed a significantly improved solar cell stability under both indoor and outdoor conditions.

1. INTRODUCTION

Dye-sensitized solar cells (DSSCs) have attracted much attention due to their low cost, easy design, and relatively high energy conversion efficiency.1−4 Recently, the energy conversion efficiency of DSSCs with liquid electrolytes was shown to be over 14%.5,6 However, the DSSCs using liquid electrolytes suffer from the problems of solvent evaporation and electrolyte leakage, which reduce the service life of DSSCs and limit their commercial application.7−9 To overcome these barriers, replacing the liquid electrolytes with quasi-solid or solid electrolytes was considered. In particular, the flexible polymer gel electrolyte (PGE) was favored due to its higher conductivity and energy conversion.10−12

PGE is a stable gel network constructed by filling a polymer matrix with a liquid electrolyte, which shows lower vapor pressure, lower contact resistance, and higher electrical conductivity with excellent thermal stability, mechanical stability, and durability. In recent years, numerous polymers have been prepared and applied as the PGE matrix for DSSCs, such as poly(ethylene oxide) (PEO), poly(vinyl pyrrolidone) (PVP), poly(dimethylsiloxane) (PDMS), and polyurethanes (PU).13−22 To obtain high ion conductivity, these polymers are often designed and fabricated into porous structures to provide more space for liquid electrolyte storage.23 For example, Manafi et al.24 fabricated a blend of porous poly(vinylidene fluoride-co-hexafluoropropylene) (PVDF-HFP) and poly(ethylene oxide) (PEO). When the PVDF-HFP/PEO blend ratio was 60/40 wt %, the best DSSC performance was achieved with the ionic conductivity of 2.07 × 10−3 S cm−1, open-circuit voltage of 0.685 V, short circuit current of 15.65 mA cm−2, filling factor of 60.4%, and photovoltaic conversion efficiency of 6.47%. Thomas et al.25 also prepared a novel porous honeycomb matrix with the blend of a polymethyl methacrylate, poly(3-decyl-1-vinyl imidazolium iodide), and polyindole (PMMA/PIL/PIN) matrix via the non-solvent-induced phase inversion method. Compared to the DSSC based on the liquid electrolyte, the DSSCs assembled using the polymer membrane had higher open-circuit voltage, short circuit current density, and power conversion efficiency (η = 5.96%) and superior stability (84% conversion efficiency retention after 250 h operation).

In recent years, a new type of porous material, electrospun nanofiber, has attracted much attention due to its easy...
manufacture, good mechanical strength, high porosity, and good specific surface area. The PGEs prepared by nanofibers showed high conductivity due to their unique nonwoven, interconnected matrix with large porosity, which is essential for high stability and high conversion efficiency of the DSSCs. Among the numerous electrospun nanofiber materials, the polyvinylidene fluoride (PVDF) has been widely used as the polymer matrix due to its high chemical inertness and good mechanical stability. However, due to the incompatibility between the PVDF and the liquid electrolyte, the reported photovoltaic performance based on PVDF PGE is not as promising as expected.

Therefore, in the current work, polyelectrolyte (PDA), a nature-inspired polymer that can be self-polymerized and coated on almost any surfaces, was used to coat the PVDF nanofibers to form PDA@PVDF nanofibers. The attached hydroxyl groups on the PDA improve the wettability of the ionic electrolyte on the PVDA nanofibers, which is critical for the best contact with the liquid electrolyte. It is expected to simultaneously enhance the conductivity and stability of DSSC by improving the compatibility between PVDF and liquid electrolytes. The microstructure, mechanical property, contact angle, thermal stability, and resistance of the prepared nanofibers and PGEs were evaluated. A quasi-solid DSSC (QS-DSSC) was fabricated from PDA@PVDF-based PGE. The PVDF-based QS-DSSC and liquid electrolyte DSSC were also fabricated for comparison.

2. RESULTS AND DISCUSSION

2.1. Morphologies and Contact Angles. The schematics and fabrication procedure of the hybrid PDA@PVDF nanostructure are illustrated in Figure 1, where the PDA coating layer was synthesized through a wet chemical method, resulting in a uniform PDA layer coated on the surface of PVDF to work as the framework for loading the ionic liquid electrolyte (ILEL). The SEM images of the electrospun PVDF nanofiber membrane and PDA@PVDF composite nanofiber membrane are shown in Figure 2a,b. The diameter of the PVDF nanofiber is about 900 nm. After the PDA coating, the surface of the nanofibers became smoother while maintaining the interconnected open pore structure of the PVDF nanofibers. There is no obvious increase in the diameter of the nanofibers since only a very thin layer of PDA was coated through in situ polymerization. These results indicate that the thin PDA coating does not change the porosity of the original nanofiber network. Such an interconnected microporous structure is essential for holding liquid electrolytes and allowing good ion transport within the membrane.

The insets in Figure 2 show the contact angle images of the corresponding membranes. Good wettability of a membrane with electrolyte is necessary for avoiding air bubbles with improved retention of the liquid electrolyte, which is important for the long-life service of a DSSC. From the insets in Figure 2, the contact angle between the nanofiber membranes and the liquid electrolyte was decreased from 127.0° to 81.0° after the PDA modification, which means that the liquid electrolyte wettability of the PDA@PVDF nanofibers was significantly improved.

2.2. Electrolyte Uptake. The electrolyte uptake as one of the indicators for the wettability of the membranes was measured and summarized in Table 1. The electrolyte uptake of the PDA@PVDF nanofiber membrane was 1100% with respect to the 760% for the uncoated PVDF nanofiber membrane. The increase in the electrolyte uptake capacity after the PDA modification is associated with the improved surface wettability as confirmed from the contact angle measurements. Meanwhile, the BET measurement revealed that the porous structures of the PVDF membrane were maintained after the PDA coating. The measured average pore diameter was 7.15 nm with a pore volume of 0.006 cm³·g⁻¹ at a specific surface area of 3.60 m²·g⁻¹. The improved wettability with a large void volume is essential for holding the liquid electrolyte with high electrolyte uptake, and guaranteeing high ion mobility within the polymer network. Hence, the PDA-modified PVDF nanofiber membrane is expected to offer better DSSC performance.

2.3. Thermal Stability. For the large-scale DSSCs, the PDA@PVDF composite membrane must be thermally stable in the devices since the solar cells are potentially exposed to high temperatures. From the TGA curves, as shown in Figure 3a, a one-step degradation was observed from both the PDA-coated and uncoated PVDF nanofibers at about 440 and 420 °C, respectively. These observations were in good agreement with the literature. The weight loss at high temperatures was due to the polymer’s thermal breakdown. The slightly higher PDA@PVDF decomposition temperature was due to the presence of the PDA decomposition signal at a higher temperature. Unfortunately, due to the broad width and significant overlap of the weight loss signals from PDA and PVDF, it was difficult to isolate the signals from them in the TGA spectra. However, the DSC spectra in Figure 3b offered better-resolved signals, and the peak contributed from the PDA was clearly resolved as an overlapping peak on the higher temperature than the DSC peak from the PVDF. The spectra confirmed that PDA is thermally more stable than PVDF. Hence, the coating PDA can slightly increase the thermal stability of the PVDF nanofibers.

2.4. Mechanical Properties. The tensile strength of the PVDF membrane and PDA@PVDF composite membranes were measured and shown in Figure 4. The ultimate tensile strength of the PDA@PVDF membrane was 9.2 MPa, which was lower than that of the PVDF membrane (13.5 MPa). However, its failure strain was 128%, which was higher than that of the PVDF membrane (102%). The results showed that the PDA coating lowered the ultimate tensile strength, which is related to the increased pore diameter of the PDA@PVDF microstructure. Meanwhile, the increased failure strain indicates a higher resistance to the stress failure due to the combination of PDA and PVDF that can effectively release the applied stress. Such mechanical performances could reduce the
rupture of the membrane in PGE and reduce the possible electrolyte leakage in the DSSCs.

2.5. Electrochemical Performances of DSSCs. To study the photoelectric performance of QS-DSSCs assembled with the PDA@PVDF nanofiber membrane, their photoelectric performance was compared with that of DSSCs assembled with ILEL (L-DSSC). Figure 5a shows the photocurrent density vs voltage ($J-V$) curves for DSSCs using the PVDF membrane, PDA@PVDF membrane, and ILEL. Their photovoltaic characteristics are summarized in Table 2.

Based on Figure 5a and Table 2, it can be seen that the solar cell with ILEL had the highest efficiency ($\eta = 9.3\%$) and short circuit current density ($J_{SC}$ 18.79 mA cm$^{-2}$). Its open-circuit voltage ($V_{OC}$) and fill factor (FF) were 0.73 V, and 68%, respectively. Although the efficiencies of the QS-DSSCs were lower than those of the L-DSSC, they were much higher than those of cells using other polymers. The reduced conversion efficiency of QS-DSSCs than that of L-DSSC is mainly due to the reduced mobility of the iodide ions within the nonwoven electrosprin fiber membrane between the TiO$_2$ photoanode and the Pt cathode with restricted dye regeneration kinetics, leading to the reduction of $J_{SC}$ and thus the photoelectric conversion efficiency. Notably, the QS-DSSCs based on the PVDF and PDA@PVDF membrane showed almost identical conversion efficiency, indicating that the addition of the PDA coating on PVDF had a negligible impact on the ionic transport property.

To understand the effect of polymer fiber on the matrix, the interfacial charge transfer resistances of the three types of DSSCs were investigated through the EIS measurements. The
Nyquist plots of the DSSCs with three different electrolyte systems and the equivalent circuit of the DSSCs are shown in Figure 5b. The $R_S$ represents the series resistance. The $R_{ct1}$ originates from the charge transfer resistance at the Pt/electrolyte interface. The $R_{ct2}$ corresponds to the charge transfer resistance at the TiO$_2$/electrolyte interface. These parameters obtained from the Nyquist analysis for the three types of fabricated DSSCs are summarized in Table 2.

In the Nyquist plots, the first small semicircle corresponds to the charge transfer resistance at the Pt/electrolyte interface ($R_{ct1}$), and the second bigger semicircle corresponds to the charge transfer resistance at the TiO$_2$/electrolyte interface ($R_{ct2}$). Normally, there is a third semicircle in the low-frequency region representing the ion diffusion resistance in the electrolyte. The absence of this feature indicates good ion mobility in DSSCs with both liquid and quasi-solid electrolytes. The series resistances of the QS-DSSCs were higher than those of the L-DSSC due to the introduction of the polymer fibrous membrane. Compared with the interface resistance of the DSSCs based on the pure PVDF membrane, the PDA coating slightly increased the charge transfer resistances at the Pt/electrolyte interface ($R_{ct1}$) and the TiO$_2$/electrolyte interface ($R_{ct2}$).

![Figure 5](https://doi.org/10.1021/acsomega.1c03232)

**Table 2. Photovoltaic Parameters of the DSSCs with Quasi-solid-state Electrolytes and Ionic Liquid Electrolyte and $R_S$, $R_{ct1}$, and $R_{ct2}$ Values Calculated from the EIS Data**

| samples   | $\eta$ (%) | $V_{OC}$ (V) | $J_{SC}$ (mA cm$^{-2}$) | FF (%) | $R_S$ | $R_{ct1}$ | $R_{ct2}$ |
|-----------|------------|--------------|--------------------------|--------|------|-----------|-----------|
| PVDF      | 8.36       | 0.73         | 17.79                    | 64     | 29.49| 7.232     | 87.84     |
| PDA@PVDF  | 8.26       | 0.72         | 17.95                    | 64     | 20.40| 9.325     | 88.53     |
| ILEL      | 9.30       | 0.73         | 18.76                    | 68     | 16.95| 8.225     | 74.33     |

![Figure 6](https://doi.org/10.1021/acsomega.1c03232)

**Figure 6.** (a) $V_{OC}$, (b) $J_{SC}$, (c) FF, and (d) $\eta$ stability of DSSC devices under indoor conditions.
interface ($R_{int}$) while significantly decreasing the series resistance ($R_s$).

2.6. Stability of DSSCs. The advantage of using a quasi-solid electrolyte reinforced with a PDA-modified PVDF nanofiber membrane is the improvement in the service lifetime of the solar cell by stabilizing the ionic liquid electrolyte. Figures 6 and 7 show the long-term performance of the four key parameters ($V_{OC}$, $J_{SC}$, FF, and $\eta$) of the cells under indoor and outdoor conditions, respectively.

From Figure 6a,c, the $V_{OC}$ and FF of the solar cells with a different type of electrolyte (ILEL, PVDF, and PDA@PVDF) showed a relatively stable performance. However, the $J_{SC}$ was gradually degraded as shown in Figure 6b, although, for the cells with ILEL- and PVDF-based electrolytes, the degradation was much faster than that of the solar cell with the PDA@PVDF electrolyte. Normally, the $V_{OC}$ is determined by the redox potentials, while the $J_{SC}$ could be affected by the internal resistance near the electrodes and within the electrolyte. Therefore, the slower degradation of the $J_{SC}$ in the PDA@PVDF cell is a good indication of the improved interface stability due to the synergetic effects of the PVDF nanofiber network and its improved compatibility with the electrolyte by PDA coating. It also confirmed that only using the pristine PVDF membrane without PDA coating is insufficient in maintaining cell stability. The degradation of the $J_{SC}$ was also manifested through the degradation in the photoelectric efficiency, $\eta$, shown in Figure 6d. After 390 h of continuous operation, the efficiency of the ILEL showed a 19.4% degradation and PVD showed a 18.5% degradation, while, in contrast, the PDA@PVDF only had a 1.4% degradation.

To evaluate the operational feasibility under real weather conditions, the photovoltaic parameters of DSSCs were tracked under the outdoor condition, as shown in Figure 7. Since the outdoor condition can accelerate the degradation, we used the factor of failure time to evaluate the weather resistance of DSSCs. The DSSC with ILEL failed at the operation time of 305 h, while the DSSC with PVDF failed at 350 h. The DSSC with PDA@PVDF had the longest operating life of 410 h, showing an improvement of 17.1 and 34.4% compared with ILEL and PVDF, respectively. It is expected that under outdoor conditions, the solar cells are subject to large temperature variation and moisture exposure, which can cause the failure of the cells. The results indicate that the quasi-solid electrolyte based on PDA@PVDF nanofiber membrane is more resilient to the environment changes with the best stability.

3. CONCLUSIONS

Using the PDA-modified nonwoven PVDF nanofiber membrane, the high uptake of electrolyte was achieved, which is crucial for minimizing the performance reduction in PGE-based QS-DSSC. There was no significant reduction in DSSC performance when the PDA@PVDF nanofiber membrane was used. More importantly, such a solar cell exhibited the longest outdoor operation life in comparison with the DSSC using ILEL or the unmodified PVDF nanofiber membrane. Such good stability originates from the high thermal stability, good mechanical strength, and tensile strength of the PDA@PVDF nanofiber membrane. Therefore, the PDA@PVDF composite membrane is a suitable material to stabilize the performance of DSSCs.
4. EXPERIMENTAL SECTION

4.1. Materials and Chemicals. Polyvinylidene fluoride (PVDF) (Kynar HSV 900, Arkema, France), N,N-dimethylformamide (DMF), and acetone were purchased from Sinopharm Chemical Reagent Corp. Dopamine hydrochloride (A.R.) was purchased from Acros Organics Inc., Belgium. Trihydroxymethyl aminomethane (Tris, A.R.) was purchased from Shanghai Ruji Biotech Co. Ltd., China. The commercial ionic liquid electrolyte (ILEL, DMIII, I2 Li, TBP, acetonitrile, NJU-AN-I), titania (TiO2) photoanode, Pt counter electrode, and ruthenium dye (N719) were purchased from Kunshan Sunlait Corp.

4.2. Preparation of PDA@PVDF Nanofibers and PGEs. Electrospun PVDF membranes were prepared according to our previous work. Briefly, the electrospun PVDF membrane was firstly immersed in ethanol for 30 min to remove some impurities. The cleaned membrane was then transferred into a Tris–HCl buffer solution of 3,4-dihydroxy phenethylamine (pH = 8.5) before adding 2 g of dopamine. The pH 8.5 is required for the self-polymerization of dopamine. After stirring vigorously for 24 h, the membrane was taken out and washed with deionized water to remove the residual dopamine and detached dark brown polydopamine particles before being dried in vacuum at 60 °C.

The electrospun PVDF nanofiber membrane and PDA@PVDF composite membrane were immersed in the ILEL solution for about 30 min to ensure sufficient electrolyte infiltration of the nanofiber membranes. The excess solution on the membrane surface was carefully wiped off with the filter paper, resulting in the quasi-solid-state electrolyte.

4.3. Fabrication of DSSC Devices. The DSSC was assembled in a classic sandwich structure consisting of a photosensitive photoanode, electrolyte, and cathode. In this study, the commercial photoanode was subsequently immersed in an ethyl alcohol solution of N719 dye of 0.3 mM at room temperature for 24 h. The surface of the photoanode was briefly rinsed with anhydrous ethanol to surface deposits of N719. The commercial Pt counter electrode was used as the cathode. The ionic liquid was used as the electrolyte, and the quasi-solid electrolyte was prepared in step 2.2. The DSSCs were assembled using the hot-press method. To compare the device performance, DSSCs were also fabricated with ILEL, with and without PVDF. Each fabricated cell has an active area of 0.196 cm2.

4.4. Measurements. The surface morphologies of the membranes were investigated by a field emission scanning electron microscope (FE-SEM, TESCAN MIRA3). The uptake of the electrolyte by the membranes was measured by the weight change before and after absorbing liquid electrolyte using the following equation:

\[
\text{Uptake(\%)} = \left(\frac{W - W_0}{W}\right) \times 100\%
\]

where \(W_0\) and \(W\) are the weights of the membrane before and after absorbing the liquid electrolyte.

Brunauer–Emmett–Teller (BET) surface area analysis was used to measure the specific surface areas and pore structures of the nanofiber membranes using the low temperature (77 K) nitrogen adsorption isotherms measured over a wide range of relative pressure from 0.02 to 1. The contact angle measurements were carried out by a contact angle measuring system (SL200C, USA KINO Industry) at room temperature. The thermal stability of the electrospun nanofiber membranes was measured using a thermal gravimetric analyzer (TGA-STA 499F Instrument) under nitrogen flow; the samples were heated from 50 to 1000 °C at a heating rate of 10 °C min\(^{-1}\). The thermal behavior of the membranes was also analyzed using a differential scanning calorimeter (DSC-200 F3 Instrument). The mechanical properties of the films were tested using a digital tensile instrument (AGS-X, Shimadzu, Japan). The tensile strength was measured at a tensile increasing rate of 10 mm/min with a standard sample dimension of 20 mm.

The photovoltaic characteristics of the DSSCs were measured by using a solar simulator (150 W simulator, Newport, Oriel 9408, 3A) under AM1.5 and 100 mW/cm² of light intensity. The incident light intensities were calibrated by using a reference cell. Electrochemical impedance spectroscopy (EIS) measurements were tested by a Zahner PP211 coupled with an IM6 electrochemical workstation in the frequency range of 10 µHz to 100 kHz and using an AC voltage signal of ±20 V. These measurements were carried out under the illumination of 100 mW/cm² using the same solar simulator. To further evaluate the stability of the DSSCs based on the PDA@PVDF electrospun nanofiber membrane quasi-solid-state electrolyte and ionic liquid electrolyte, the cells were placed in indoor and outdoor conditions (the aging box was used to simulate the harsh outdoor environment; temperature at 55 °C, humidity of 50%, and irradiance of 1000 W/m²), and the electrochemical performance of the DSSCs was characterized regularly.

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Notes

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