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A series of conducting gel electrolytes for quasi-solid-state quantum dot-sensitized solar cells with boosted electron transfer processes

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ABSTRACT

To pursue electron-generation stability with no sacrifice of photovoltaic performance has been a persistent objective for all kinds of solar cells. Here, we demonstrate the experimental realization of this objective by quasi-solid-state quantum dot-sensitized solar cells from a series of conducting gel electrolytes composed of polyacrylamide (PAAm) matrix and conductive polymers [polyaniline (PANI), polypyrrole (PPy) or polythiophene (PT)]. The reduction of S_x^{2-} occurred in both interface and three dimensional framework of conducting gel electrolyte as a result of the electrical conduction of PANi, PPy and PT toward refluxed electrons from external circuit to Pt electrode. The resulting solar cells can yield the solar-to-electrical conversion efficiency of 2.33%, 2.25% and 1.80% for PANi, PPy and PT based gel electrolytes, respectively. Those solar cells possessed much higher efficiency than that of 1.74% based on pure PAAm gel electrolyte owing to the enhanced kinetics for $S_x^{2-} \leftrightarrow S^{2-}$ conversion. More importantly, the stability of quasi-solid-state solar cell is significantly advanced, arising from the localization of liquid electrolyte into the three dimensional framework and therefore reduced leakage and volatilization.

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1. Introduction

Quantum dots, a class of nano-scale materials, possess a series of merits, such as the tunable band gap [1], high absorption coefficient [2], and multiple exciton generation [3]. Based on their extraordinary material properties, along with the prospect of cost effective precursors and simple versatile synthesis methodology, quantum dot-sensitized solar cells (QDSSCs) have drawn considerable attentions in recent years. Meanwhile, QDSSCs have been regarded as impressive candidates for next generation photovoltaics owing to their extra-high theoretical power conversion efficiency, stemming from the traditional dye-sensitized solar cells [4–8]. Up to now, enormous efforts have been made to explore advanced quantum dots as sensitizers such as CdS, CdSe, In_2S_3 as well as alloy quantum dots to enhance the overall power output. The recorded solar-to-electrical energy conversion efficiency of QDSSC has been raised to 11.6% based on Zn-Cu-In-Se quantum dots [9–12]. However, apart from the efficiency, stable electricity generation under persistent solar irradiation is another crucial parameter to evaluate the QDSSCs performance [13]. Unfortunately, the volatilization and leakage of liquid electrolyte (S^{2-}/S_x^{2-} redox couple) are always the challenging issues for commercial application

of QDSSCs. Therefore, how to address these problems has been a persistent objective to break the impasse of quantum dots based photovoltaic device [14].

Quasi-solid-state or full-solid-state electrolytes are preferred candidates in highly stable devices according to previous reports [15,16]. For solid-state electrolyte, sluggish charge transfer kinetics at the counter electrode/electrolyte as well as the photoanode/electrolyte interfaces will accelerate the recombination reaction, significantly reducing the overall device performance [17,18]. In contrast, quasi-solid-state gel electrolyte shows promising characteristics with comparable ionic conductivity of liquid electrolyte [19–21]. For example, Yu et al. have incorporated polyacrylamide (PAAm) polymer matrix into QDSSCs, yielding power conversion efficiency as high as 4% for CdS/CdSe co-sensitized solar cells [22]. Apart from the polymer-based gel electrolyte, low molecular mass gelators including dextran [23], 12-hydroxystearic acid [24] and natural polysaccharide Konjac glucomannan [25] have been employed to convert liquid electrolyte into quasi-solid-state species, which shows enhanced stability and corresponding lower conversion efficiency, mainly attributed to the resistance between polymer matrix and two electrodes. In order to address the above-mentioned issue, conductive additives such as graphene and conducting polymer have been incorporated into the quasi-solid-state electrolyte to enhance the power output of dye-sensitized solar

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46 cells, demonstrating the promising application in photovoltaic field
 47 [26,27]. Herein, taking all aspects into consideration, we developed
 48 a series of conducting gel electrolytes by incorporating polyaniline
 49 (PANI), polypyrrole (PPy) or polythiophene (PT) into the PAAm ma-
 50 trix, yielding the enhanced solar-to-electrical energy conversion ef-
 51 ficiencies as well as the stability comparing to pure PAAm gel elec-
 52 trolyte by boosting the electron transfer. Finally, the conversion ef-
 53 ficiency of the PAAm-PANI based quasi-solid-state device can be up
 54 to 2.33%. This can be explained by the expansion of the reduction
 55 reaction ($S_x^{2-} \leftrightarrow S^{2-}$) sites into the interior of conducting gel elec-
 56 trolyte comprising the PANi, PPy or PT chains.

57 2. Experimental

58 2.1. Synthesis of PANi, PPy and PT

59 0.592 mL of aniline was dissolved in 20 mL of 1 M HCl aqueous
 60 solution to obtain a homogeneous mixture. 20 mL of 0.125 M am-
 61 monium peroxydisulfate (APS) aqueous solution was added drop-
 62 wwisely into the above mixture within 30 min. The polymerization
 63 reaction was carried out at 0°C. After 3 h, the resultant reactant
 64 was rinsed by 1 M HCl aqueous solution, filtrated, and finally vac-
 65 uum dried at 60°C for 24 h.

66 1 mL of pyrrole monomer was dripped in an aqueous solution
 67 containing 7.788 g of $FeCl_3 \cdot 6H_2O$ and 58 mL of deionized water. Under
 68 vigorous agitation, the polymerization reaction was carried out
 69 at 5°C for 24 h. After being rinsed by aqueous solution, filtrated,
 70 and finally vacuum dried at 60°C for 24 h, the resultant PPy pow-
 71 ders were obtained.

72 0.407 mL of 3,4-ethoxylenedioxy thiophene (EDOT) and 0.048 g
 73 of cetyltrimethylammonium bromide (CTAB) were dissolved in
 74 100 mL of 1 M HCl aqueous solution to obtain a homogeneous mix-
 75 ture. Then, 20 mL of 0.125 M APS aqueous solution was dripped in
 76 the above mixture within 30 min under 5°C for 24 h. The resultant
 77 reactant was rinsed by 1 M HCl aqueous solution, filtrated, and fi-
 78 nally vacuum dried at 60°C for another 24 h.

79 2.2. Synthesis of PAAm/conducting polymer composites

80 The PAAm matrix was synthesized according to an aqueous
 81 polymerization route. In details, 10 g of acrylamide monomer and
 82 0.5 g of PANi, PPy or PT were thoroughly dissolved in 15 mL of
 83 deionized water. After vigorous agitation, the solution was deg-
 84 gassed for 10 min and heated in a water bath of 80°C. Then,
 85 0.0015 g of N,N'-(methylene)bisacrylamide and 0.06 g of APS were
 86 added into the above mixture. With the proceeding of polymer-
 87 ization, the viscosity increased accordingly. When the viscosity of
 88 the PAAm prepolymers reached around 180 mPa s^{-1} , the reactants
 89 were poured into a Petri dish and cooled to room temperature
 90 with the formation of an elastic gel. After being rinsed with ex-
 91 cess deionized water, the samples were vacuum dried at 80°C for
 92 more than 12 h. The microporous matrices were prepared by im-
 93 mersing the dense composites into deionized water for 2 days to
 94 reach swelling equilibrium, and subsequently the swollen PAAm
 95 composite hydrogels were freeze-dried under vacuum for 48 h. The
 96 closed 3D framework of matrix will be open during the swelling
 97 process, whereas the freeze-dry technique can ensure the elimina-
 98 tion of water with no morphological variation.

99 2.3. Synthesis of conducting quasi-solid-state gel electrolyte

100 To synthesize the conducting quasi-solid-state gel electrolyte,
 101 the freeze-dried PAAm composite hydrogels were immersed into
 102 the liquid-state electrolyte including 1 M S, 1 M Na_2S , and conduct-
 103 ing polymer (PANi, PPy or PT) with a concentration of 1.38 g mL^{-1}
 104 for two days until their swelling equilibrium. The finally obtained

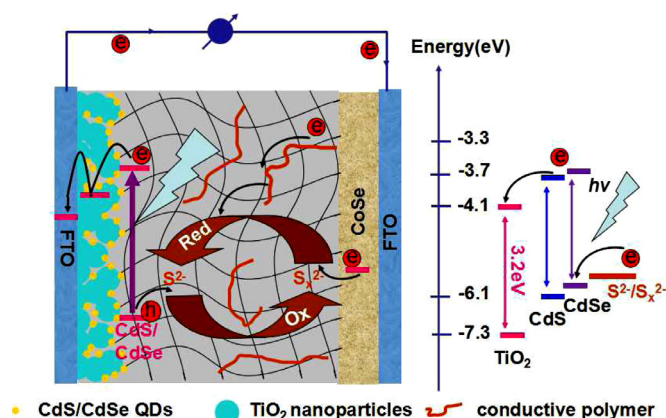


Fig. 1. The schematic diagram of a quasi-solid-state QDSSC device from a CdS/CdSe-sensitized TiO_2 photoanode and a conducting gel electrolyte.

105 electrolytes were labeled as the PAAm-PANI, PAAm-PPy and PAAm-PT
 106 gel electrolytes, respectively.

107 2.4. Preparation of CoSe alloy counter electrodes

108 The CoSe alloy counter electrode was synthesized following the
 109 previous reports [28]. In details, 0.01 g of Se powders and 0.0238 g
 110 of $CoCl_2 \cdot 6H_2O$ were mixed in 27.5 mL of deionized water by vig-
 111 orous agitation to obtain a homogeneous mixture. Subsequently,
 112 7.5 mL of hydrazine hydrate (85 wt%) was added dropwisely into
 113 the above solution. After vigorous agitating for 10 min, the mixture
 114 was transferred into a 50 mL of Teflon-lined autoclave, in which
 115 the cleaned FTO glass substrate with FTO layer downward was im-
 116 mersed. After reacting at 120°C for 12 h, the FTO substrate was
 117 rinsed by deionized water and vacuum dried at 50°C.

118 2.5. Assembly of quasi-solid-state QDSSCs

119 Before assembling solar cells, a layer of TiO_2 nanocrystal with
 120 the thickness of $10 \mu\text{m}$ and the active area of $5 \text{ mm} \times 5 \text{ mm}$ was
 121 firstly deposited on the surface of fresh cleaned FTO glass substrate
 122 ($12 \Omega \text{ cm}^{-2}$) with doctor-blade method according to previous re-
 123 ports [29]. Subsequently, the as-prepared anode film was sintered
 124 at 450°C for 30 min in air to form a mesoporous construction.

125 For fabricating the sensitized photoanode, the above-mentioned
 126 TiO_2 was alternatively soaked in 0.1 M $Cd(NO_3)_2$ ethanol solution
 127 for 1.5 min and 0.1 M Na_2S methanol solution for 1.5 min. By re-
 128 peating these cycles for 12 times to obtain CdS sensitized TiO_2
 129 anodes. The CdSe deposition after CdS coating was performed by
 130 chemical bath deposition (CBD) method. Briefly, the CdS-coated
 131 TiO_2 film was immersed into a solution containing 25 mM cad-
 132 mium acetate dihydrate, 20 mM sodium sulfite (Na_2SO_3), 10 mM
 133 selenium (Se) and 3 mL of a NH_4OH solution for 1 h. Finally, the
 134 quasi-solid-state QDSSC was fabricated by sandwiching the gel
 135 electrolytes into the gap between CdS/CdSe sensitized photoanode
 136 and CoSe counter electrode, as shown in Fig. 1.

137 2.6. The characterizations

138 The surface morphologies of 3D polymer matrixes were
 139 recorded via a SEM-4800-field-emission scanning electron micro-
 140 scope. The FTIR spectra were recorded on a PerkinElmer spectrum
 141 1760 FTIR spectrometer. The ionic conductivities of gel electrolytes
 142 were measured using a pocket conductivity meter (DSSJ-308A, Le-
 143 icip Instrument) by filling the gel electrolytes into the interspace be-
 144 tween two electrodes, which was calibrated with 0.01 M KCl aque-
 145 ous solution prior to experiment.

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