



Graphene tailored gel electrolytes for quasi-solid-state quantum dot-sensitized solar cells



Xiao Jin ^{a, b}, Chun Chang ^b, Zhongping Chen ^b, Qinghua Li ^{a, b, *}

^a School of Physics Science and Technology, Lingnan Normal University, Zhanjiang 524048, PR China

^b Jiangxi Engineering Laboratory for Optoelectronics Testing Technology, Nanchang Hangkong University, Nanchang 330063, PR China

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ABSTRACT

Quantum dot-sensitized solar cells (QDSCs) are promising for future renewable sources. By addressing the issues on liquid-electrolyte leakage and sluggish charge transportation, a class of graphene oxide tailored polyacrylamide (PAAm/GO) gel electrolytes are synthesized for quasi-solid-state QDSC applications. The conducting pathways with interconnected GO species can catalyze reduction reaction from S_n^{2-} to S^{2-} and therefore shorten charge-transfer length. Upon illumination by simulated sunlight (AM1.5, 100 mW cm^{-2}), the optimized QDSC with PAAm/0.9% GO yields a maximized power conversion efficiency of 4.10% in comparison with 1.78% for the device free of GO. Arising from the good retention of liquid electrolyte by three-dimensional PAAm/GO framework, the final quasi-solid-state QDSCs present good long-term stability.

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1. Introduction

With the development of human society, energy crisis has been paid growing attentions. One of the solutions to this impasse is to replace fossil energies by renewable sources such as solar cells. The mesoscopic structured photovoltaics are considered as promising candidates because of simple fabrication processes, high efficiency and scalable materials and technologies. However, a remaining issue for all of these solar cells with mesoscopic TiO_2 is either liquid electrolyte leakage for dye-sensitized solar cells and quantum dot-sensitized solar cells (QDSCs) [1–4] or humidity attack for perovskite solar cells. The relatively high photoelectric power conversion efficiency in theory is featured on QDSCs for the third generation photovoltaics. A state-of-the-art QDSC comprises of a quantum dot-sensitized photoanode, a liquid electrolyte having S/S_n^{2-} redox couples and a counter electrode [5]. Earlier studies on liquid-junction QDSCs are mainly focusing on the synthesis of new quantum dots with less surface defects, tunable bandgap, high molar extinction coefficient [6] and high absorption coefficient [7]. The photoelectric conversion efficiency is still markedly increased by using narrow bandgap dualistic quantum dots such as CdS, CdSe,

PbS [10], PbSe [11], ternary alloy quantum dots such as CuInS_2 [12], CuInSe_2 [13], CuInTe_2 [14] as well as composite quantum dots such as CdS/CdSe [8,9]. The high efficiencies arise from the rapid charge transfer processes within liquid system, however the liquid electrolyte always attack sealing film to leak from sandwiched solar cells, which has been an unsolved but crucial problem for their future commercialization [15].

To address this issue, polymer gel electrolyte is always utilized as a placeholder for liquid redox couples to increase long-term stability of QDSCs [16,17]. The use of quasi-solid-state gel electrolytes creates associated problems: reduced charge-transfer rate and sluggish electrocatalytic behaviors at counter electrode/gel electrolyte interfaces [18]. In the current work, we would like to build electron-conducting gel electrolytes by incorporating graphene oxide (GO) into three-dimensional polyacrylamide (PAAm) framework, leading to the formation of interconnected conducting channels. These peculiar pathways can conduct electrons from counter electrode to conducting PAAm/GO electrolyte, expanding catalytic area and shortening charge-transfer length [19].

2. Experimental

2.1. Preparation of GO powders

GO powders were prepared according to the following

* Corresponding author. School of Physics Science and Technology, Lingnan Normal University, Zhanjiang 524048, PR China.

E-mail address: qhli@hqu.edu.cn (Q. Li).

procedures. In details, 1 g of graphite powders, 0.5 g of NaNO_3 and 23 mL of concentrated sulphuric acid solution (95 wt%) were placed in an ice-bath. After agitation for 1 h, 3 g of mineral chameleon particles were added into above-mentioned reagent and kept agitation for another 2 h. Subsequently, the reactor was transferred into 35°C water bath and mixed with 86 mL of deionized water. 4.5 mL of 30% hydrogen peroxide aqueous solution was dropped until no bubbles. The corresponding dark brown solution was repeatedly centrifuged at a speed of 8500 rpm for 3 min and rinsed with deionized water to control pH of solution change at 3–4. Finally, the resultant GO solution was transferred into a Petri dish in lyophilizer and freeze dried more than 48 h to obtain anhydrous GO powders.

2.2. Synthesis of PAAm/GO gel electrolytes

The PAAm/GO matrix was synthesized by an aqueous solution polymerization method. In details, 10 g of acrylamide monomer and stoichiometric GO powders (The GO dosages in total gel electrolyte were tuned at 0, 0.5, 0.7, 0.9 and 1.1 wt%) were dispersed in 15 mL of deionized water. By heating the reagent to 80°C in a water bath with agitation, 1 mL of 1.5 mg mL^{-1} crosslinking agent from N,N' -(methylene)bisacrylamide and 1 mL of initiator from 0.225 g mL^{-1} ammonium persulfate were dropped. Under agitation for several minutes, the reagent became a viscous system because of formation of branching polymer backbones. Subsequently, the reagent was transferred into a Petri dish and cooled to room temperature for persistent polymerization to form completed three-dimensional PAAm framework. After drying at 60°C for more than 24 h, then final PAAm/GO matrix was obtained.

PAAm/GO gel electrolyte was prepared by immersing anhydrous PAAm/GO matrix into a liquid electrolyte consisting of 1 M sulphur and 1 M sodium sulfide aqueous solution for three days to reach swelling equilibrium.

2.3. Preparation of CdS/CdSe quantum dots sensitized TiO_2 photoanodes

TiO_2 colloid prepared by a classical sol-hydrothermal method [20] was coated onto freshly cleaned FTO glass substrate ($12\ \Omega\ \text{square}^{-1}$, $2 \times 2\ \text{cm}^2$ in size) by a doctor-blade method. The area and thickness of colloidal TiO_2 layer were controlled at $0.5 \times 0.5\ \text{cm}^2$ and $10\ \mu\text{m}$, respectively. Subsequently, the colloidal TiO_2 films were calcined at 450°C for 30 min to obtain a mesoscopic TiO_2 film.

CdS quantum dots were adsorbed on mesoscopic TiO_2 film by a successive ionic layer adsorption and reaction (SILAR) method according to previous report [21]. In details, FTO glass supported TiO_2 film was soaked by 0.1 M cadmium nitrate ethanol solution and 0.1 M sodium sulfide methanol solution. Anhydrous ethanol and anhydrous methanol were used to rinse to remove weakly adsorbed quantum dots. CdS quantum dot sensitized photoanode was obtained by repeating the procedures for 12 times. To synthesize CdS/CdSe co-sensitized TiO_2 photoanode, 0.1 M Se powders and 0.2 M sodium sulfide aqueous solution were refluxed at 120°C for 2 h to obtain Na_2SeSO_3 solution. Then, 25 mM cadmium acetate, 3 mL ammonia (NH_4OH) and 10 mL of Na_2SeSO_3 were mixed in 20 mL deionized water. Subsequently, CdS quantum dot sensitized photoanode was soaked into abovementioned mixture and heated at 90°C for 30 min to obtain CdS/CdSe sensitized TiO_2 photoanode.

2.4. Synthesis of CoSe alloy counter electrodes

CoSe alloy counter electrodes were prepared according to similar processes in previous report [22]. In details, 0.01 g of Se powders and 0.0238 g of $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$ were dissolved in 27.5 mL of deionized water, which was agitated to form a homogeneous phase. Then, 7.5 mL of hydrazine hydrate (85 wt%) was added and kept for agitation for 10 min, and subsequently transferred into a Teflon-lined autoclave. Meanwhile, cleaned FTO glass with size of $1.5 \times 2\ \text{cm}^2$ was placed into autoclave with FTO layer downward. After reaction for 12 h at 120°C , the CoSe alloy counter electrode was rinsed with deionized water and vacuum dried at 60°C .

2.5. Assembly of quasi-solid-state QDSCs

The PAAm/GO gel electrolyte was cut into a piece with thickness of around $500\ \mu\text{m}$, and was sandwiched between a CdS/CdSe quantum dot sensitized TiO_2 photoanode and a CoSe alloy counter electrode. The final solar cell architecture was shown in Fig. 1, upon irradiation from photoanode side, electricity was generated to external circuit.

2.6. Measurements and characterizations

The current density-voltage curves of QDSCs were recorded on an electrochemical workstation (CHI600E, Shanghai Chenhua Device Company, PR China) upon irradiation with a sunlight simulator (short arc xenon lamp/mercury lamp light source CHF-XM-500W,

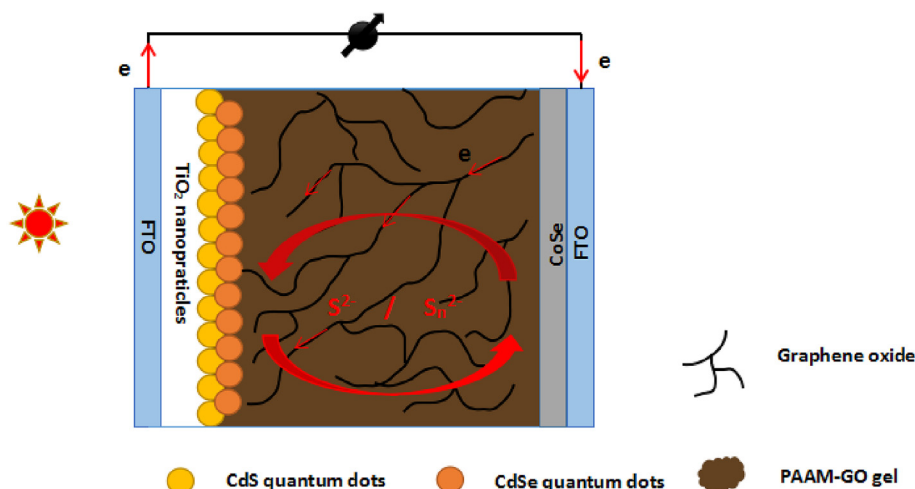


Fig. 1. Schematic structure of the QDSC with PAAm/GO gel electrolyte and redox processes.

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