



# High efficiency quasi-solid state dye-sensitized solar cells based on a novel mixed-plasticizer modified polymer electrolyte



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## ABSTRACT

Quasi-solid-state dye-sensitized solar cells (DSSCs) fabricated with the mixed-plasticizer (MP) modified polymer electrolyte are reported in this paper. The mixture of hydroxyethyl methacrylate (HEMA) and ethylene glycol (EG) as plasticizer are added into the original composite polymer electrolyte (CPE) based on poly(ethylene oxide)/poly(vinylidene fluoride-hexafluoropropylene) (PEO/P(VDF-HFP)) and KI/I<sub>2</sub>. The olefinic bonds in HEMA and hydroxyl bonds in EG provide strong molecular polarity to effectively reduce the crystallinity of the polymer electrolyte, thus largely improve the ionic conductivity of the CPE. On account of MP, the decrease of crystallinity provides a better photovoltaic performance, the best photon-to-current conversion efficiency is 6.79% with a short current density J<sub>sc</sub> of 15.23 mA cm<sup>-2</sup> under AM 1.5 illumination. Fourier transforms infrared (FT-IR), differential scanning calorimetry (DSC), ionic conductivity, electrochemical impedance spectroscopy are test to analyze superior property of DSSCs assembled with MP-modified CPEs. It shows that the performance of the CPEs can be largely improved by MP.

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## 1. Introduction

Dye sensitized solar cell (DSSC) has attracted enough attention for several decades since the first report written by O'Regan and Gratzel in 1991 because of its low-cost and simple fabrication process. The highest solar cell efficiency has reached 13% [1] by now with liquid electrolyte. However, liquid electrolyte has several shortages such as volatility, packaging problem and toxicity [2,3]. Due to above shortages, the study on DSSCs with long-term stability and high efficiency is a hot research direction.

Solid or quasi-solid state dye-sensitized solar cells [4] are good choice to overcome the shortages that liquid electrolyte possesses. A large number of researches have been investigated about using small molecule gelata [5], plasticizer [6], inorganic nanoparticle [7] modified polymer electrolyte and conducting polymer [8] to manufacture solid or quasi-solid electrolyte.

For polymer electrolyte, excellent film-forming property, good elasticity and light weight provide a significant advantage. However, there are still several difficulties to settle. For example, the ionic

conductivity in liquid electrolyte is still higher than that in solid and quasi-solid electrolyte [9]; the permeation with TiO<sub>2</sub> nanocrystalline is weak [10]; the interfacial electron recombination is serious [11].

In order to improve the ionic conductivity in solid and quasi-solid electrolyte, plasticizer is often added into the polymer electrolyte system. It has been inspected that many papers have shown the improvement of the DSSC performance using electrolytes that combine PEO/P(VDF-HFP) with several different types of materials, such as cross-linking agent [12], inorganic nanoparticles [13], organic small molecular materials [14] and plasticizer [15]. Plasticizer can effectively reduce the degree of crystallinity and improve the electrical conductivity, improving short circuit current consequently. As is known, single plasticizers are studied with CPEs in many reported researches. However, there is no work on adding mixed-plasticizer (MP). It is induced that the internal coupling in MP may lead to a higher current increase compared with single plasticizer. Therefore we report a high efficiency quasi-solid-state DSSC assembled with the MP of HEMA and EG as the plasticizer in this experiment. HEMA and EG provide strong molecular polarity to effectively reduce degree of crystallinity in CPEs. The reliability of ionic conductivity and photovoltaic performance on the HEMA with EG concentration in CPEs were also investigated. The CPEs employs KI and I<sub>2</sub> as redox couple, a polymer composite of PEO/P(VDF-HFP) as the polymer matrix, glycol dimethyl ether(DME)and propylene carbonate(PC)as the solvent. It was found that the addition of HEMA

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and EG into the CPEs caused significant increases in the short circuit current density ( $J_{sc}$ ) and achieved an optimal photon to current conversion efficiency up to 6.79%, compared with 6.11% of original DSSC.

## 2. Experimental part

### 2.1. Materials

Poly(ethylene oxide) (PEO,  $M_w = 2 \times 10^6 \text{ g mol}^{-1}$ , Aldrich), Poly(vinylidene fluoride-hexafluoropropylene) (P(VDF-HFP),  $M_w = 4.77 \times 10^5 \text{ g mol}^{-1}$ , ElfAtochem), potassium iodide (KI, 98.5%, SCRC, China), iodine ( $I_2$ , 99.8%, Beijing, China), propylene carbonate (PC, 99.9%, SCRC, China), 1,2-dimethoxyethane (DME, 99.0%, SCRC, China),  $TiO_2$  nanoparticles (P25, 20–30 nm, Degussa AG, Germany) and  $SiO_2$  nanoparticles (aerosil 200, 12–15 nm, Degussa AG, Germany), dye (N719, Ruthenium 535 bis-TBA, Suzhou, China), hydroxyethyl methylacrylate (HEMA, Aldrich), ethylene glycol (EG, Aldrich). All chemicals referred above are stored in vacuum desiccators before use and used without further purification.

### 2.2. Synthesis of polymer electrolyte

1 g high molecular polymer compound PEO/P(VDF-HFP) (weight ratio of 2:3) and 30 g organic solvent PC/DME (volume ratio of 7:3) were blended at  $80^\circ\text{C}$  for about 4 h. 0.1 g  $SiO_2$  nanoparticles were added after the polymer compound dissolved completely in organic solvent. Then 1.25 g KI and 0.19 g  $I_2$  were added to the slurry; continuous stirring was performed until the redox couple was well mixed with the polymer slurry. Then the CPE was divided into five with the same mass.

After that, CPEs were obtained with different concentrations (0 g, 0.10 g, 0.15 g, 0.20 g, 0.25 g) of the MP, which combined EG with HEMA (mass ratio 1:2). The materials were stirred until they were mixed uniformly.

All of the CPEs with different concentrations of MP were spread on the dye-sensitized  $TiO_2$  photoanodes. Then the dye-sensitized  $TiO_2$  photoanodes with the different CPEs were put into the oven at  $50^\circ\text{C}$  to evaporate the solvent.

### 2.3. DSC and FT-IR studies

Differential scanning calorimetry (DSC) thermograms of the polymer electrolyte membranes was measured at the heating rate

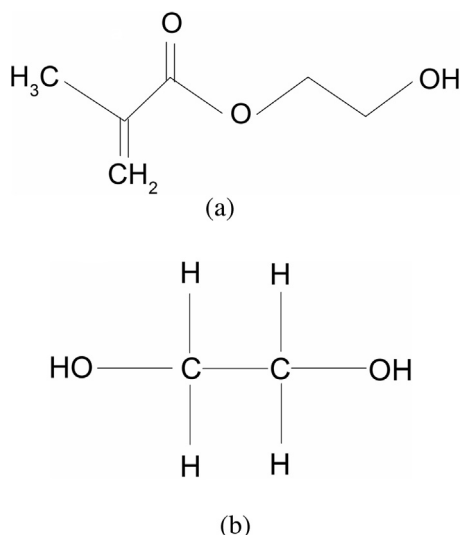


Fig. 1. The chemical construction of HEMA (a) and EG (b).

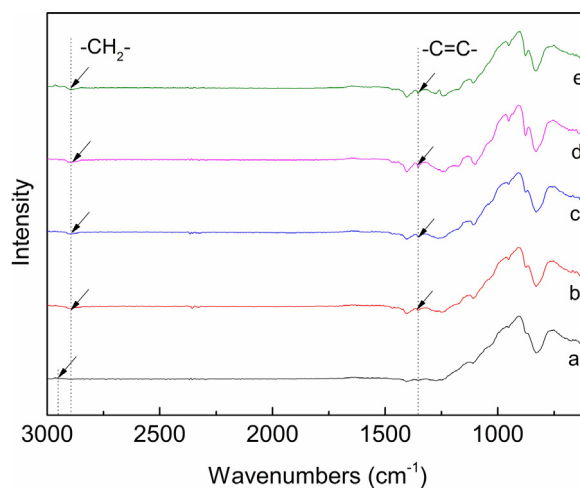


Fig. 2. ATR-FTIR spectra of CPE (a) modified with 0.10 g (b), 0.15 g (c), 0.20 g (d) and 0.25 g (e) MP.

of  $10^\circ\text{C}/\text{min}$  from  $-100$  to  $0^\circ\text{C}$  under  $N_2$  environment with the PerkinElmer Pyris Diamond (German).

The FT-IR spectra of the MP-modified CPE and original CPE are measured on a Bruker Tensor FT-IR spectrometer from  $3000$  to  $600 \text{ cm}^{-1}$  using the reflection mode.

### 2.4. Polymer electrolyte's ionic conductivity measurement

Agilent 4292 A precision impedance analyzer (USA) is applied to measure the impedance (Nyquist plot) of the polymer electrolyte over the frequency varying between 40 Hz and 1 MHz when the voltage amplitude is 10 mV. The sandwich-type samples just like Pt/polymer electrolyte film/Pt have been prepared before the measurement. The film thickness (ca.  $50 \mu\text{m}$ ) is controlled using an adhesive tape.

The ionic conductivity,  $\sigma$ , of the membrane is calculated from the following equation:  $\sigma = L/AR_b$  [6], where  $L$  is the thickness of the polymer electrolyte membrane, and  $A$  is the area of the electrode. The resistance ( $R_b$ ) is taken at the intercept of the Nyquist plot with the real axis.

### 2.5. Electrochemical impedance spectroscopy measurement

Electrochemical station (Zahner, Germany) is used to measure the electrochemical impedance spectroscopy of the polymer electrolytes. The frequency varies between 0.05 and  $10^6$  Hz when the voltage amplitude is 10 mV. The MP-modified DSSCs are kept in a dark environment, and a forward bias of  $-0.70 \text{ V}$  is applied to the samples.

### 2.6. Current–voltage characteristics measurements

A 1000 W xenon light source (Newport, USA) was used to give an irradiance of  $96.4 \text{ Mw cm}^{-2}$  (AM 1.5) on the DSSC cells with a mask of  $0.16 \text{ cm}^2$  aperture to measure the photo-to-current conversion efficiencies. The intensity of the incident light is calibrated by a Si-1787 photodiode. A Keithley 2400 digital source meter unit (USA) was used to measure the current–voltage curves under the light.

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