



Role of ion conducting polymeric electrolyte in suppressing deterioration of cathode electrodes in dye-sensitized solar cells



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ABSTRACT

In this study, an ion conducting polymeric hydrogel (ICPH) is synthesized, characterized and used as a matrix for trapping the electrolyte solution to suppress the deterioration of cathode electrodes for dye-sensitized solar cells (DSSCs). Polymeric hydrogels are synthesized as free radical cross-linking copolymerization of acrylamide (AAM) and methylenebisacrylamide (BIS) as crosslink agent in slab forms with the same thickness. The polymer host is characterized by scanning electron microscopy (SEM). Impedance spectroscopy studies are carried out in the frequency range of 5 Hz–13 MHz at room temperature. It is observed that while the dielectric constant decreased, the loss tangents increased with increasing electrolyte content which is trapped in ICPH. The as-prepared ICPH/graphene electrodes, assembled with this material as cathode, are confirmed as substitution for platinum (Pt) counter electrode in DSSCs. The photovoltaic tests of the ICPH-DSSCs are carried out by the current–voltage (*I*–*V*) measurements and it is observed that ICPH-DSSC exhibited significantly enhanced electrical properties such as increased photovoltaic cell efficiency (PCE) which reached to 5.41% after using the ICPH. We predict that this work will open a way to fabricate the DSSC devices with a cathode electrode based on ion conductive polymeric hydrogels.

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1. Introduction

Polymer hydrogels are three-dimensional crosslinked polymer networks which can carry huge amount of water in it [1]. Hydrogels are electrical insulators in their dried state. When the gels swell in an appropriate solution, ions can move in the gel thus they become conductive. The conductivity of the hydrogels can be increased by doping with metal particles or salt molecules [2–4].

Hydrogels are soft materials and good candidate for filler since the hydrogels can keep their shapes with huge amount of water. Due to the conductivity properties, hydrogels became popular in the field of design in organic electronic devices. Synthesize of composite/hybrid hydrogels is a popular and promising field of research. The most applicable way of forming composite/hybrid materials by using hydrogel is the incorporation of different materials into a hydrogel matrix.

Hydrogels are sensitive to external stimuli like pH, temperature,

light intensity/magnetic/electric field, ionic strength and external stress [5–7]. Many researches focus on the ion conducting-polymeric hydrogels (ICPH) to develop new applications, in particular robotics, sensors, conducting materials, wave-absorbing materials and separation techniques and solar energy applications which is a new research field [8–10]. Because the amount of resources is decreasing day by day, renewable energy especially dye sensitized solar cell (DSSC) is a credible alternative to conventional solar cell [11].

The anode and the cathode electrodes of DSSCs are made of a specific type of glass that has a Transparent Conductive Oxide (TCO) coating on one side. The cathode is coated with a catalytic material such as platinum or carbon materials in order to catalyze the electron transfer process and for the circuit to be completed. However, a platinum metal is expensive and thus researchers have spent their efforts on finding a new material which is low cost and also catalyze the electron transfer efficiently.

An electrolyte solution, especially iodine solution, separates the electrodes and ensures charge transportation through a redox couple. Besides, electrolytes still suffer from solvent leakage because of their thermodynamic instability under high

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temperature.

In this study, for DSSCs we proposed a new cathode electrode designed with ICPH in which electrolyte was trapped, to overcome the problems, especially the electrolyte leakage and the catalyst flooding when the system is scaled up. To the best of our knowledge, the incorporation of electrolyte into ICPH for catalyze the electron transfer has not been studied yet.

Here, our attempt has been to carry out the dielectric characterization of ICPH by using the impedance spectrometer at room temperature. The structure and the dielectric properties of ICPH was investigated in detail. We also reported one promising cathode electrode material, which is both optically transparent and reasonably active for the I_3^-/I^- redox reaction. The synthesis and the photovoltaic properties of symmetric octakis (2-ethoxytetrahydro-2H-pyranthio) porphyrazine complexes were used as a sensitizer to fabricate dye-sensitized solar cells and were reported for the first time. The experiments were performed on metal-free and metalloporphyrazines such as Zn (II). In contrast to earlier reports [12–15], we obtained good solar conversion efficiency (5.68%) in DSSCs with an optically transparent ICPH/graphene cathode.

2. Materials and methods

2.1. Materials

The monomer, acrylamide (AAM); the initiator, ammonium persulphate (APS); and the crosslinkers, methylenebisacrylamide (BIS) were supplied by Merck (Darmstadt, Germany). All chemicals were used as received. Bidistilled water was used for synthesis.

All chemicals were reagent grade from Merck and Fluka. Solvents in reactions were distilled from appropriate drying agents prior to use and commercially available reagents were used without further purification unless otherwise stated. Flash column chromatography was carried out using silica gel 60 (0.04–0.063 mm) from Merck.

2.2. Synthesis

2.2.1. Synthesis of ion conductive-polymeric hydrogel (ICPH)

Ion Conductive-Polymeric hydrogels were synthesized as free

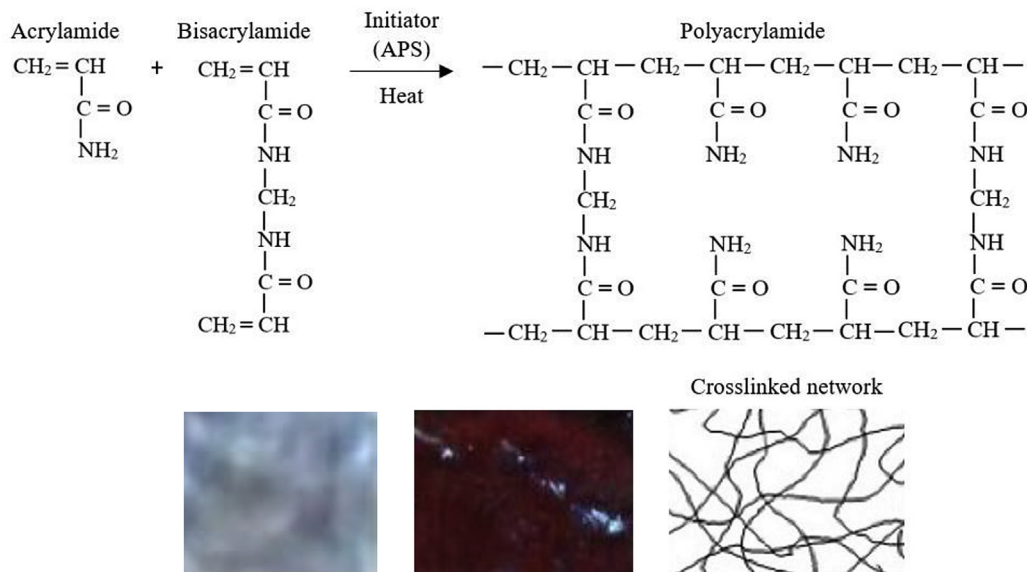
radical cross-linking co-polymerization of AAM and BIS as crosslink agent in slab forms with the same thickness, is shown in Scheme 1. The monomer, AAM, and the crosslinker, BIS, concentrations were kept fix as 1 M and 7.3 mM, respectively. After degassing the pregel solution by nitrogen lasted about 20 min. APS, as an initiator, was added into the pregel solution. These solution was kept in the heat bath at 60 °C for one hour to complete the gelation process.

2.2.2. Synthesis of octakis (2-ethoxytetrahydro-2H-pyranthio) porphyrazines and Zn(II) derivate

2.2.2.1. 2,3,7,8,12,13,17,18-octakis(2-ethoxytetrahydro-2H-pyranthio)-5,10,15,20-porphyrazine (H_2Pz). Pure magnesium porphyrazine (1.50 g) was dissolved in the minimum amount of CF₃COOH and carefully poured on the ice. The solution was then neutralized with concentrated ammonia. The dark product was then filtered. The crude product was chromatographed on silica gel using chloroform as the eluent. The oily product was chromatographed on silica gel using chloroform/n-hexane (1:1) mixture as the eluent. Yield: 1.35 g (51.0%). Calc. for C₁₈H₂₆N₂O₄S₂: C, 54.25; H, 6.58; N 7.03%; Found: C 54.32, H 6.26, N, 7.43%. FTIR (KBr), ν/cm^{-1} : 3285 (NH), 2922 and 2852 (aliphatic CH₂), 1278 and 1117 (ether group): ¹H NMR(CDCl₃): δ (ppm) 5.00 (t, 8 H, O-CH-O), 4.60 (t, 16 H, O-CH₂), 4.00–3.70 (m, 16 H, py-OCH₂), 3.50 (q, 16 H, S-CH₂), 1.80–1.20 (m, 48 H, py-CH₂), -1.1 (s, 2 H, -NH). MS (MALDI-TOF) m/z : Calc. 1621.87 [M⁺ Na]⁺.

2.2.2.2. 2,3,7,8,12,13,17,18-Octakis(2-ethoxytetrahydro-2H-pyranthio)-5,10,15,20 porfirazinato Zn(II) (ZnPz). 0.20 g (0.13 mmol) of the free-based porphyrazine was dissolved in DMF and added to the solution of 0.24 g (1.30 mmol) Zn (OAc)₂ in EtOH. The mixture was refluxed under argon for 1 h. After cooling to room temperature, insoluble excess metal salt was separated by filtering.

The reaction mixture was freed of the solvent under rotary evaporation and the crude product was purified by column chromatography on silica gel using a gradient technique. Yield: 0.15 g (72.0%). Calc. for C₇₂H₁₀₄N₈O₁₆S₈Zn: C, 52.11; H, 6.32; N, 6.75%. Found: C, 52.57; H, 6.75; N, 7.29%. FT-IR (KBr), ν/cm^{-1} : 2934 and 2867 (aliphatic CH₂), 1283 and 1134 (ether group): ¹H NMR(CDCl₃): δ (ppm) 5.00 (t, 8 H, O-CH-O), 4.60 (t, 16 H, O-CH₂), 4.00–3.70 (m, 16 H, py-OCH₂), 3.50 (q, 16 H, S-CH₂), 1.80–1.20 (m, 48 H, py-CH₂):



Scheme 1. Reaction scheme, photos (left-ICPH, middle-ICPH/electrolyte and right-crosslinked network) of the synthesis of the ICPH.

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