



Iodine-free nanocomposite gel electrolytes for quasi-solid-state dye-sensitized solar cells



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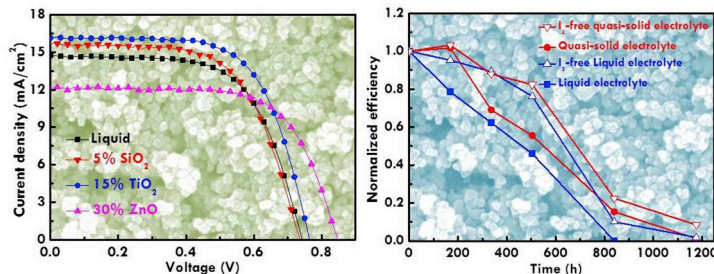
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HIGHLIGHTS

- I₂-free quasi-solid-state (QS) electrolyte is made for dye-sensitized solar cells.
- Inorganic particles play dual role of gelling agent and charge transport enhancer.
- The SiO₂, TiO₂, and ZnO are used for gelling dye-sensitized solar cells (DSSCs).
- The I₂-free QS DSSC gelled using 15% TiO₂ got an efficiency of 8.00%.
- The I₂-free QS DSSC gelled using 15% TiO₂ showed good long-term at-rest stability.

GRAPHICAL ABSTRACT



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ABSTRACT

Iodine-free quasi-solid-state electrolytes were developed for dye-sensitized solar cells (DSSCs) by removing I₂ from the conventional I⁻/I₃⁻ redox electrolyte and gelling the electrolyte with metal oxide nanoparticles. The inorganic nanoparticles played a dual role of gelling agent and charge transport enhancer. The removal of I₂ improved the long-term stability of DSSCs, but reduced the ionic diffusivity of electrolyte and thus the solar-to-electricity efficiency of DSSCs. By introducing a suitable nanoparticle gelator into the electrolyte, the ionic diffusivity and the interfacial charge transfer could be enhanced due to the formation of 3-D networks that facilitate Grotthuss-type ion exchange. Three different metal oxide nanoparticles (SiO₂, TiO₂, and ZnO) were evaluated, and the best performance was obtained by TiO₂ nanoparticles. The I₂-free quasi-solid-state DSSCs gelled by 15% TiO₂ achieved an efficiency of 8.00% vs. a 6.94% efficiency attained by the I₂-free liquid-state reference cells. The I₂-free quasi-solid-state DSSCs gelled by 15% TiO₂ also exhibited good long-term at-rest stability with an estimated life span of five to six years under ambient condition based on an accelerated stability test performed at 65°C with 65% relative humidity.

1. Introduction

The dye-sensitized solar cell (DSSC) is one of the promising solar

cells for practical applications because of its easy-to-fabricate and low-production-cost features [1–4]. The individual roles of electrochemical behaviors were noticed in previous literature [5–9]. A typical DSSC is

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composed of a dye-loaded photoanode, an electrolyte with a redox couple, and a counter electrode. The electrolyte is a crucial component needed for the charge transport between the photoanode and the counter electrode and continuously regenerates the dye during DSSC operation. The photovoltaic performance and the long-term stability of DSSC both depend greatly on the electrolyte. Traditional liquid electrolytes consisting of the iodide/triiodide (I^-/I_3^-) redox couple with additional additives in a volatile organic solvent like acetonitrile can produce highly efficient cells [10]. The traditional iodine-containing liquid electrolyte, however, has a few problems that limit the efficiency and long-term stability of DSSCs. Volatile liquid electrolyte is not practical for long-term operation due to its leakage issue. Furthermore, the electrolyte I_3^- ions, which are produced by the reaction between iodine (I_2) and I^- in the electrolyte, are corrosive and have prominent adsorption for visible light. The long-term stability and efficiency of DSSCs are thus compromised.

Several approaches have been proposed to overcome the volatile and leakage issues of the traditional organic solvent-based liquid electrolyte. Ionic liquid-based electrolytes are one possible approach [4,11,12]. Wang et al. reported a solvent-free electrolyte containing the ionic liquids 1-methyl-3-propylimidazolium iodide (PMII) and 1-ethyl-3-methylimidazolium dicyanamide (EMIDCN), which led to a 6.6% cell efficiency at 100 mW cm^{-2} light intensity [13]. Another extensively used approach is to introduce gelators like polymers or inorganic nanoparticles into the liquid electrolytes. These gelators establish physical or chemical cross-linking networks that trap the liquid and transform the electrolyte quasi-solid, thereby suppressing solvent evaporation, alleviating electrolyte leakage and improving the long-term stability of DSSCs [14–16]. Compared with polymer gelators, inorganic nanoparticles have the advantage of reducing the fluidity of electrolyte without sacrificing the cell efficiency. Silica nanoparticles were first applied as the gelator by Wang et al. Apart from having comparable solar-to-electricity conversion efficiency (η) as the liquid-state cells, the resultant quasi-solid state DSSC also showed superior stability, with 90% retention of cell efficiency after a 30-day stability test at 80°C [17]. Stathatos et al. used a sol-gel method to synthesize a silica-based nanocomposite electrolyte that contained ionic liquid and achieved 5.4% η for TiO_2 DSSCs [18]. Yanagida et al. tested several inorganic gelators, including TiO_2 , SiO_2 , SnO_2 , carbon black, and multi-walled carbon nanotubes, the quasi-solid state DSSCs derived from which yielded higher η than the liquid-state ones [19]. Of the inorganic gelators investigated by Yanagida et al., TiO_2 and SiO_2 nanoparticles produced the best results. Lee et al. employed TiO_2 and SiO_2 nanoparticles as the gelator for TiO_2 DSSCs and found the former yielded higher η . The TiO_2 -gelled quasi-solid state DSSCs also displayed high stability, with 80% retention of η after 1200 h of storage [20]. We previously tested three different metal oxide nanoparticles (SiO_2 , TiO_2 , and ZnO) as the electrolyte gelator for ZnO DSSCs and found all the resultant quasi-solid state DSSCs achieved higher η than the liquid-state controls. The highest η of 4.17% was recorded for the ZnO -gelled cells, which also displayed excellent at-rest stability with 95% retention of η after 150 days [16]. All these studies demonstrate that inorganic nanoparticles can increase the viscosity of the electrolyte without compromising the power conversion efficiency of DSSCs. It is thought that the 3-D network formed by the contiguous nanoparticles allows the alignment of ions in the electrolyte, so charge transfer can proceed by the Grotthuss-type ion exchange mechanism, where physical diffusion of ionic species is not necessary [17].

To alleviate the corrosion and light adsorption problems of electrolyte I_3^- ions, efforts have been made to develop iodine-free electrolytes [21]. Apart from using alternative redox couples [22], iodide-based electrolytes that contain no I_2 have also been developed [23]. In these iodine-free electrolytes, I_3^- is generated through the reaction of oxidized dye and I^- in the electrolytes when the cells are illuminated ($3 I^- + 2 \text{ dye}^* \rightarrow I_3^- + 2 \text{ dye}^*$). The self-formed I_3^- is then reduced to I^- by accepting electrons from the counter electrode. The problems

associated with I_3^- ions are thus minimized due to low concentrations of I_3^- ions in the electrolyte. Some of the I_2 -free iodide-based electrolytes were made quasi-solid through the incorporation of polymers. For example, Chen et al. employed polyethylene oxide (PEO) to gel an I_2 -free acetonitrile-based electrolyte that contained an ionic liquid (1, 2-dimethyl-3-propylimidazolium iodide, DMPII) and potassium iodide (KI) for charge transport. The η of resultant quasi-solid-state TiO_2 devices reached 5.87% [24]. More recently, the same group tuned the visible light absorption and rheological properties of the PEO-based quasi-solid electrolyte with polyethylene glycol (PEG) and applied it to Ti-foil-based DSSCs. The highest η attained for these back-illuminated cells was 6.44% [25]. Rong et al. developed I_2 -free polymer gel electrolyte by employing a PEO/PVDF-HFP (poly(vinylidene fluoride-co-hexafluoropropylene) composite for monolithic DSSCs. By tuning the concentrations of DMPII and other electrolyte additives, monolithic quasi-solid-state DSSCs with η approaching 7% were obtained [26].

In this study, we employed metal oxide nanoparticles instead of polymers to gel I_2 -free iodide-based electrolytes for TiO_2 DSSCs, in order to capitalize the unique advantages offered by nanoparticle-built 3-D networks, which have been shown to facilitate charge transfer through the Grotthuss-type ion exchange mechanism. Our I_2 -free electrolytes contained an ionic liquid (PMII) and an alkali salt (lithium iodide, LiI), which function as charge transfer mediators and provide I^- ions. Three different metal oxide nanoparticles (SiO_2 , TiO_2 , and ZnO) were evaluated as electrolyte gelators, and their effects on the photovoltaic performance and long-term stability of resultant DSSCs were investigated based on photocurrent-voltage (J - V), cyclic voltammetry (CV), and electrochemical impedance spectroscopy (EIS) analyses. For comparison, corresponding I_2 -containing and liquid-state reference DSSCs were also prepared.

2. Experimental

2.1. Preparation of electrolytes

The I_2 -containing liquid electrolyte was made by dissolving 0.6 M PMII (Sigma Aldrich), 0.05 M I_2 (99.99%, Sigma Aldrich), 0.1 M LiI (99%, Sigma Aldrich), and 0.5 M 4-tert-butylpyridine (TBP, 99%, Sigma Aldrich) in acetonitrile (ACN, 99.9%, ECHO). The I_2 -free liquid electrolyte had the same composition as the I_2 -containing one except for the omission of I_2 . To prepare quasi-solid-state electrolytes, adequate amounts of metal oxide nanoparticles were mixed into the liquid electrolytes as a gelator. The inorganic nanoparticles were desiccated in a vacuum oven for at least a day before use. Three different types of nanoparticles were utilized: SiO_2 (99.5%, 5–15 nm, Sigma-Aldrich), TiO_2 (P25, Anatase 70%, Rutile 30%, 21 nm, Degussa), and ZnO (> 97%, < 50 nm, Sigma-Aldrich). The weight percentage (wt%) of nanoparticles was abbreviated as percentage (%) afterwards.

2.2. Assembly of dye-sensitized solar cells

The fluorine-doped tin oxide (FTO) glass (Nippon Sheet Glass, 8–10 Ω/\square , 2.2 mm-thick) was cleaned sequentially with acetone, a neutral cleaner, and deionized water under sonication. The photoanode was fabricated by depositing TiO_2 films on the FTO glass via screen printing (active area = 0.16 cm^2). The TiO_2 photoanode had two layers: a dye-adsorption layer composed of anatase TiO_2 nanopowders (a- TiO_2) and a light scattering layer consisting of commercial TiO_2 powder (QF-Ti-1125F, 200–300 nm). The details of the preparation of a- TiO_2 and its screen-printing paste can be found in our previous work [27]. The screen-printing paste for the light scattering layer was prepared in a similar fashion as a previously reported procedure [28]. Briefly, to prepare the scattering layer paste, an absolute ethanol solution (40 mL) containing 1 g of TiO_2 powder (QF-Ti-1125F, 200–300 nm) was mixed thoroughly with another absolute ethanol solution (20 mL) containing 0.35 g ethyl cellulose (45 cp) and 0.45 g ethyl cellulose (10 cp). Then

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