



# A supramolecular gel electrolyte formed from amide based co-gelator for quasi-solid-state dye-sensitized solar cell with boosted electron kinetic processes

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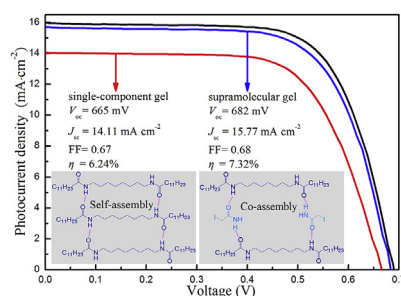
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## HIGHLIGHTS

- A supramolecular gel electrolyte was prepared by two-component amide based co-gelator.
- The supramolecular gel electrolyte provides a fast electron kinetic process.
- The QS-DSSC with supramolecular gel electrolyte exhibits boosted performance.

## GRAPHICAL ABSTRACT



## ARTICLE INFO

### Article history:

Received 20 March 2017

Received in revised form

24 April 2017

Accepted 26 April 2017

### Keywords:

Gel electrolyte

Co-gelator

Electron kinetic process

Quasi-solid-state

Dye-sensitized solar cell

## ABSTRACT

A supramolecular gel electrolyte ( $T_{\text{gel}} > 100\text{ }^{\circ}\text{C}$ ) is formed from *N,N'*-1,8-octanediybis-dodecanamide and iodoacetamide as two-component co-gelator, and introduced into the quasi-solid-state dye-sensitized solar cells (QS-DSSCs). The different morphologies of microscopic network between two-component and single-component gel electrolytes have influence on the diffusion of redox couple in gel electrolytes and further affect the electron kinetic processes in QS-DSSCs. Compared with the single-component gel electrolyte, the two-component gel electrolyte has less compact gel network and weaker steric hindrance effect, which provides more effective charge transport channel for the diffusion of  $\text{I}_3^-/\text{I}^-$  redox couple. Meanwhile, the  $-\text{NH}_2$  groups of iodoacetamide molecules interact with  $\text{Li}^+$  and  $\text{I}_3^-$ , which also accelerate the transport of  $\text{I}_3^-/\text{I}^-$  and decrease in the  $\text{I}_3^-$  concentration in the  $\text{TiO}_2/\text{electrolyte}$  interface. As a result, nearly a 12% improvement in short-circuit photocurrent density ( $J_{\text{sc}}$ ) and much higher open circuit potential ( $V_{\text{oc}}$ ) are found in the two-component gel electrolyte based QS-DSSC. Consequently, the QS-DSSC based on the supramolecular gel electrolyte obtains a 17% enhancement in the photoelectric

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<http://dx.doi.org/10.1016/j.jpowsour.2017.04.099>

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conversion efficiency (7.32%) in comparison with the QS-DSSC based on the single-component gel electrolyte (6.24%). Furthermore, the degradations of these QS-DSSCs are negligible after one sun light soaking with UV cutoff filter at 50 °C for 1000 h.

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## 1. Introduction

Dye-sensitized solar cells (DSSCs) are considered as the next-generation solar cells, owing to their low cost, simple manufacturing and low energy consumption [1]. There have been many attempts to increase the overall conversion efficiency of DSSCs, mostly using liquid electrolytes, and an impressive photoelectric conversion efficiency (14.3%) of DSSCs have been achieved [2]. However, a critical barrier to overcome for the liquid electrolyte based DSSCs is the liquid electrolyte itself, which lead to leakage and evaporation, dye degradation [3]. Quasi-solid-state electrolyte turns out to be a good candidate for substituting the liquid electrolyte to alleviate these problems above. Low molecular weight organic gelators (LMOGs) have the unique characteristics of both good solubility upon heating and inducement of smooth gelation of organic liquids at low concentrations [4,5]. Moreover, LMOGs can trap liquid electrolytes in cages formed in a three-dimensional network by self-assembly driven by non-covalent interactions, such as hydrogen bonding,  $\pi$ - $\pi$  interactions and so on [6]. Nevertheless, the photovoltaic performances of quasi-solid-state DSSCs (QS-DSSCs) based on LMOGs are usually inferior to that of corresponding liquid electrolyte based DSSCs due to the inhibited charge transportation caused by the steric-hindrance effect of the compact gel network in the gel electrolytes [7–11]. Thus the development of high-performance gel electrolyte materials with excellent microscopic morphologies for improving the performance of QS-DSSCs is of great important. Supramolecular gels comprised of multi-component LMOGs, by inserting an additional level of hierarchical control into the self-assembly process, offer highly tunable and controllable forms of materials behavior, confer multiple stimuli-responsive to behavior, including thermo-, chemo-, and mechanical-responses, and be widely used in different fields [12–15].

In this paper, we focus on two-component supramolecular gel electrolyte system formed by *N,N'*-1,8-octanediylbis-dodecanamide and iodoacetamide as co-gelator, which has significant changes in the morphology and electrochemistry between the two-component assemblies and the single-component one formed by *N,N'*-1,8-octanediylbis-dodecanamide. The changes in the microscopic structures of the gel networks were studied by field emission scanning electron microscopy (FE-SEM) and X-ray fiber diffraction (XRD) measurements. The influences of different morphologies between supramolecular and single-component gels on the electrochemical properties of the electrolytes, and the electron kinetic processes in the devices were investigated by linear voltammetric measurement and intensity-modulated photocurrent/photovoltage spectroscopy (IMPS/IMVS). It is remarkable that the photoelectric conversion efficiency of QS-DSSC was largely improved by using the supramolecular gel electrolyte, and these QS-DSSCs based on the gel electrolytes exhibit excellent long-term stability during the accelerated aging test.

## 2. Materials and methods

### 2.1. Electrolytes preparation

The liquid electrolytes contained 0.1 mol L<sup>-1</sup> LiI (99%, Aldrich),

0.6 M mol L<sup>-1</sup> I<sub>2</sub> (99%, Aldrich), 0.45 mol L<sup>-1</sup> *N*-methylbenzimidazole (NMBI, 99%, Aldrich), and 0.9 mol L<sup>-1</sup> DMPII in 3-methoxypropionitrile (MePN, 99%, Fluka). Two steps were required to prepare the gel electrolyte. Firstly, *N,N'*-1,8-octanediylbis-dodecanamide (LMOG) was dissolved (6 wt% vs. liquid electrolyte) in liquid electrolyte at 120 °C under stirring for 30 min. Secondly, cooling the mixture solvent to room temperature (25 °C), the single-component gel electrolyte (Gel A) was formed. Similarly, the two-component supramolecular gel electrolyte (Gel B) was prepared by adding iodoacetamide (98%, J&K) and *N,N'*-1,8-octanediylbis-dodecanamide according to the mole ratio of 1:2 (6 wt% vs. liquid electrolyte) into the liquid electrolyte, and heated to 120 °C under stirring until the gelators were dissolved, then cooling to room temperature (25 °C). 1,2-Methyl-3-propylimidazoliumiodide (DMPII) and *N,N'*-1,8-octanediylbis-dodecanamide were synthesized according to the Supplementary Materials as previous literature [16]. The detailed synthetic procedure, <sup>1</sup>H-NMR and elemental analysis data of the organogelator can be obtained in the Supplementary Materials.

### 2.2. Differential scanning calorimetry (DSC)

The gel to solution transition temperature ( $T_{gel}$ ) of the gel electrolyte was carried out with a differential scanning calorimeter (DSC-Q2000, TA, USA) [17]. Briefly, 5 mg of each sample was prepared on sealed aluminum pans. Nonisothermal experiments were performed in the temperature range 25–140 °C at a heating rate of 10 °C·min<sup>-1</sup> and under a nitrogen purge gas flow of 50 ml min.

### 2.3. X-ray fiber diffraction (XRD)

X-ray diffraction patterns of xerogel were measured on a X-ray powder diffraction analysis (XRD, TTR-III, Rigaku Corp., Japan) with Cu-K $\alpha$  irradiation ( $\lambda = 1.5418 \text{ \AA}$ ). Each sample was scanned over the  $2\theta$  range from 5° to 30°.

### 2.4. Field emission scanning electron microscopy (FE-SEM)

Field emission scanning electron microscopy (JSM-6330F, JEOL, Japan) was used for investigating the morphology of the gels. And the xerogel sample was prepared by drying the Gel A and Gel B at ambient temperature.

### 2.5. Linear sweep voltammetry measurements

Linear sweep voltammetry (LSV) measurements were conducted with an electrochemical work station (Autolab 320, Metrohm, Switzerland). The steady-state current–voltage curves for the different electrolytes contain liquid and quasi-solid-state electrolytes. For the LSV, the Pt ultramicroelectrode (CHI107, CH Instruments Inc., USA) is used as working electrode, a 1 mm radius platinum disk electrode (CHI102, CH Instruments Inc., USA) as counter electrode and reference electrode [18,19]. The potential was cycled from –0.8 to 0.6 V at a scan rate of 5 mV s<sup>-1</sup>.

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