



## Stable quasi-solid-state dye-sensitized solar cell using a diamide derivative as low molecular mass organogelator



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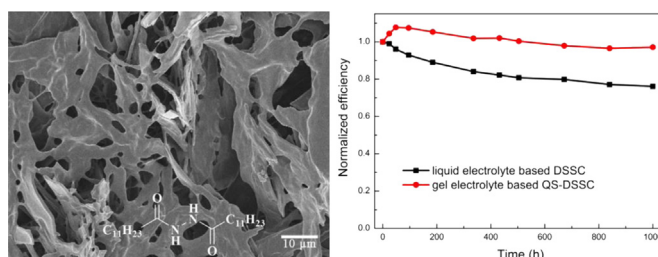
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### HIGHLIGHTS

- A diamide derivative is used as LMOG to gelate liquid electrolyte.
- A novel and intrinsic stable gel electrolyte with high  $T_{gel}$  is prepared.
- The kinetic processes of electron transport and recombination are investigated.
- The QS-DSSC exhibits excellent stability during the accelerated aging tests.

### GRAPHICAL ABSTRACT



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### ABSTRACT

High stability is a significant target for practical applications of dye-sensitized solar cells. 2-(1-oxododecyl)hydrazide, a diamide derivative, is synthesized and applied in quasi-solid-state dye-sensitized solar cells (QS-DSSCs) as a low molecular mass organogelator (LMOG). It is noteworthy that the transition temperature from gel state to liquid state ( $T_{gel}$ ) of this gel electrolyte is 125.2 °C, which ensures the gel state of the electrolyte at the DSSC operating temperature. The influences of the gel electrolyte on the kinetic processes of electron transport and recombination are investigated. The diffusion of redox species in the gel electrolyte is hindered by the crosslinked network, and the decreased electron recombination lifetime indicates an increased electron recombination in QS-DSSC. Significantly, the QS-DSSC exhibits excellent thermal and light-soaking stabilities during accelerated aging tests for 1000 h. Especially, there is almost no change in the short-circuit current density ( $J_{sc}$ ) in the QS-DSSC, while the  $J_{sc}$  of the liquid electrolyte based DSSC decreases to 79–90% of their initial values. These results are very important for the application and commercialization of DSSCs.

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## 1. Introduction

Since the original development of sensitized nanocrystalline solar cells (DSSCs) by Michael Grätzel and his coworkers [1], DSSCs are considered to be one of the emerging solar technologies that offers the potential to reduce the cost of photovoltaic electricity generation. It is clear that stability is a prerequisite for application of any photovoltaic technology and the extent of application is limited by the level of stability that can ultimately be achieved. The

electrolyte is one of the key components of DSSCs and its properties greatly affect the photovoltaic conversion efficiencies and stability of DSSCs [2,3]. And the organic solvent liquid electrolyte based DSSCs have been optimized to achieve the power conversion efficiency exceeding 12% [4], however, the volatile solvent in liquid electrolyte usually results in leakage and evaporation, which is considered as one of the critical factors limiting the long-term performance and practical use of DSSCs. Many alternatives have been made to overcome this problem by replacing the liquid electrolytes, including inorganic or organic hole conductors and polymer gel electrolytes. However, derivative problems such as contact between hole transporting materials and the nanoporous semiconductor films are remain to be solved. Therefore, the quasi-solid-state electrolyte which formed by low molecular mass organogelators (LMOGs) arouses wide concern. At proper temperature, the LMOGs can self-assemble through H-bonds, van der Waals Forces,  $\pi$ - $\pi$  bond and lead to a three-dimensional network which immobilizes the liquid solvent. The formed gel can be melted above the transition temperature from gel state to liquid state ( $T_{\text{gel}}$ ) and reformed gel below this temperature. This thermal reversible nature is beneficial to the fabrication of DSSCs. A lot of components, such as 12-hydroxystearic acid [5], cyclohexanecarboxylic acid-[4-(3-oxadecylureido)phenyl]amide [6], tetradodecylammonium bromide [7] and 1,3:2,4-di-O-benzylidene-d-sorbitol derivatives [8] have been developed as LMOGs and applied in the quasi-solid-state dye-sensitized solar cells (QS-DSSCs).

In this work, we synthesize 2-(1-oxododecyl)hydrazide and used it as LMOG to gelate 3-methoxypropionitrile (MePN) based liquid electrolyte to obtain a novel gel electrolyte. The kinetic processes of electron transport and recombination, the photovoltaic performance and both the thermal and light-soaking stabilities of the corresponding QS-DSSC were investigated in detail.

## 2. Experimental

### 2.1. Electrolyte preparation

The 2-(1-oxododecyl)hydrazide compound was synthesized according the literature [9]. 1,2-Methyl-3-propylimidazoliumiodide (DMPII) was prepared as reported previously [10]. The liquid electrolyte for DSSCs was composed of 0.1 mol L<sup>-1</sup> iodine (I<sub>2</sub>: 99%, Aldrich), 0.1 mol L<sup>-1</sup> anhydrous lithium iodide (LiI: 99%, Aldrich), 0.5 mol L<sup>-1</sup> N-methylbenzimidazole (NMBI: 99%, Aldrich), and 1 mol L<sup>-1</sup> DMPII in 3-methoxypropionitrile (MePN: 99%, Fluka). Gel electrolytes with different concentrations of gelator were prepared by adding 2-(1-oxododecyl)hydrazide into liquid electrolyte and heated under stirring until the gelator melted. The gel electrolytes were formed after cooling to room temperature. The selection of the concentration of gelator for further study in this work is described in the [Supporting Information](#).

### 2.2. Fabrication of dye-sensitized solar cells

The nanocrystalline electrodes about 11.3  $\mu\text{m}$  thickness were obtained by screen-printing TiO<sub>2</sub> paste on FTO glass (TEC-8, LOF). After sintering at 450 °C for 30 min in air then cooling to 120 °C, the nanoporous TiO<sub>2</sub> photoelectrodes were immersed in an ethanol solution of 0.5 mmol L<sup>-1</sup> cis-dithiocyanate-*N,N*-bis-(4-carboxylate-4-tetrabutylammoniumcarboxylate-2,2'-bipyridine) ruthenium (II) (N719 dye) for 14 h. The platinized counter electrodes were obtained by spraying H<sub>2</sub>PtCl<sub>6</sub> solution to FTO glass followed by heating at 410 °C for 20 min. DSSCs were assembled by sealing the dyed nanoporous TiO<sub>2</sub> photoelectrode and the counter electrode with a thermal adhesive film (Surlyn 1702, Dupont, USA). The liquid electrolyte was injected into the internal

space between of two electrodes through the hole on the counter electrode, which was later sealed by a cover glass and thermal adhesive film. The gel electrolyte was heated to 135 °C under stirring until the gel transform to liquid completely. Then, the electrolyte (hot solution) was rapidly injected into the cell and the cell was sealed as the same as the liquid electrolyte based DSSC. After cooling to room temperature, a uniform motionless gel layer was formed in cell.

### 2.3. Differential scanning calorimetry (DSC)

The transition temperature from gel state to liquid state ( $T_{\text{gel}}$ ) of the gel electrolyte was determined by differential scanning calorimeter (DSC-Q2000, TA, USA). Approximately 5–7 mg of each sample was weighed and sealed in an aluminum pan and heated at a rate of 10 °C min<sup>-1</sup> under nitrogen flow from 25 to 145 °C for DSC measurement.

### 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 gel. And the xerogel sample was prepared by drying the gel (8 wt% gelator in MePN) at ambient temperature.

### 2.5. Linear sweep voltammetry measurements

Linear sweep voltammograms were recorded on a electrochemical workstation (Autolab 320, Metrohm, Switzerland) at 25 °C in two-electrode mode of DSSCs equipped with a 5.0  $\mu\text{m}$  platinum ultramicroelectrode (CHI107, CH Instruments Inc., USA) as working electrode, a 1 mm radius platinum disk electrode (CHI102, CH Instruments Inc., USA) as counter electrode and reference electrode [19,20]. The linear sweep voltammograms were obtained at scan rate of 5 mV s<sup>-1</sup>.

### 2.6. Electrochemical impedance spectroscopy (EIS) measurement

EIS measurement of DSSCs was recorded with an electrochemical analyzer (Autolab 320, Metrohm, Switzerland). To measure the impedance, a direct-current bias at 600 mV, and a perturbation amplitude of 10 mV within the frequency range from 10 MHz to 10 mHz was applied in dark. The obtained impedance spectra were fitted with Z-view software (v2.8b, Scribner Associates, USA) in terms of a transmission line equivalent circuit model to interpret the characteristics of DSSCs [11,12].

### 2.7. Controlled intensity modulated photocurrent/photovoltage spectroscopy (IMPS/IMVS) measurements

The experimental setup for IMPS and IMVS measurements have been described elsewhere [13–15]. Intensity-modulated measurements were carried out by an electrochemical workstation (IM6e, Zahner, Germany) with light emitting diodes (LED) ( $\lambda = 610 \text{ nm}$ ) driven by export (Zahner, Germany). The LED provided both the dc and ac components of the illumination. A small ac component is 10% or less than that of the dc component and the frequency range was 3 kHz–300 mHz.

### 2.8. Characterization of incidental photon-to-electron conversion efficiency (IPCE)

The photocurrent action spectra were recorded on a QE/IPCE measurement kit consisting of a 300 W xenon lamp (69911, Newport, USA), a 1/4 m monochromator (74125 Oriel Cornerstone 260,

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