



# Enhancing the efficiency of quasi-solid-state dye-sensitized solar cells by adding bis(trifluoromethane)sulfonimide lithium salt and camphorsulfonic acid to gel-based electrolytes



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## ABSTRACT

In this study, gel-based electrolytes were produced by mixing a liquid-based electrolyte with poly(methyl methacrylate) (PMMA), which served as a gelator to enhance the stability of the electrolytes and dye-sensitized solar cells (DSSCs). Furthermore, bis(trifluoromethane)sulfonimide lithium salt (LiTFSI) and camphorsulfonic acid (C<sub>10</sub>H<sub>16</sub>O<sub>4</sub>S) were separately added to the gel-based electrolytes. The results indicate that adding LiTFSI and C<sub>10</sub>H<sub>16</sub>O<sub>4</sub>S to the gel-based electrolytes effectively enhanced the device efficiency. In particular, the device with the 1:1 + 0.1 M LiTFSI gel-based electrolyte exhibited the optimal efficiency (7.33%), which was higher than that observed in the device with the liquid-based electrolyte. The results also proved that the DSSC devices with the gel-based electrolytes exhibited a longer stability. Therefore, using optimized gel-based electrolytes can enhance the power conversion efficiency and stability of DSSC devices.

## 1. Introduction

Energy shortages and environmental pollution are two major challenges facing humanity in the twenty-first century. Therefore, in recent years, scholars in the field of science have paid increasingly more attention to research on renewable energy, which refers to types of energy that are constantly and sustainably replenished and do not generate pollutants. Among all types of renewable energy, solar energy is one of the most advantageous because of its high sustainability and zero negative effect on the environment [1–3]. In 1991, a Swiss research team led by Professor Grätzel developed dye-sensitized solar cells (DSSCs), which were shown to be high-efficiency, low-cost devices [4]. Since then, research on DSSCs has received a considerable amount of attention. DSSCs feature high efficiency, low cost, a simple structure, and are easy to fabricate [5–7]. Structurally, DSSC devices comprise a transparent conductive substrate, nanoporous titanium dioxide (TiO<sub>2</sub>) semiconductor thin films, dyes, electrolytes, and a platinum (Pt) counter electrode [8–20]. Featuring redox properties, electrolytes serve as a key component in DSSC devices; they provide dyes with electrons to enable the dyes to return from an excited state to the ground state, thereby completing dye regeneration. Conventional DSSC electrolytes are mostly liquid-based. However, solvent evaporation and leakage

tend to occur with liquid-based electrolyte, which shortens the stability of DSSC devices [21]. Therefore, in recent years, numerous research teams have studied and improved DSSC electrolytes to enhance device stability. According to a recent literature review, the main research directions are currently focused on replacing conventional liquid-based electrolytes with ionic liquid, gel, or solid-based electrolytes [22–24].

Up to the present, many kinds of polymers or co-polymers have been used as the gelator in quasi-solid-state DSSCs such as poly(acrylonitrile-co-vinyl acetate) (PAN-VA) [25], poly(methyl methacrylate) (PMMA) [26], poly(ethylene glycol) (PEG) [27], poly(methyl methacrylate-co-methacrylate acid)/poly(ethylene glycol) [P(MMA-co-MAA)/PEG] [28], and poly(vinylidene fluoride-co-hexafluoropropylene) (PVDFHFP) [29]. PMMA is a long-chain polymer compound with properties such as high material softness, high mechanical strength, low crystallization tendencies, and high material transmittance; thus, it is an excellent gelator for preparing gel-based electrolytes [30]. Adding PMMA to electrolytes causes electrolyte gelling, thereby reducing electrolyte evaporation and leakage. However, adding gelators to electrolytes decreases the conductivity of the electrolytes and increases the impedance of the electrolyte layer, thereby lowering the power conversion efficiency of DSSCs. This study focused on fabricating gel-based electrolytes by mixing PMMA with a liquid-based electrolyte to

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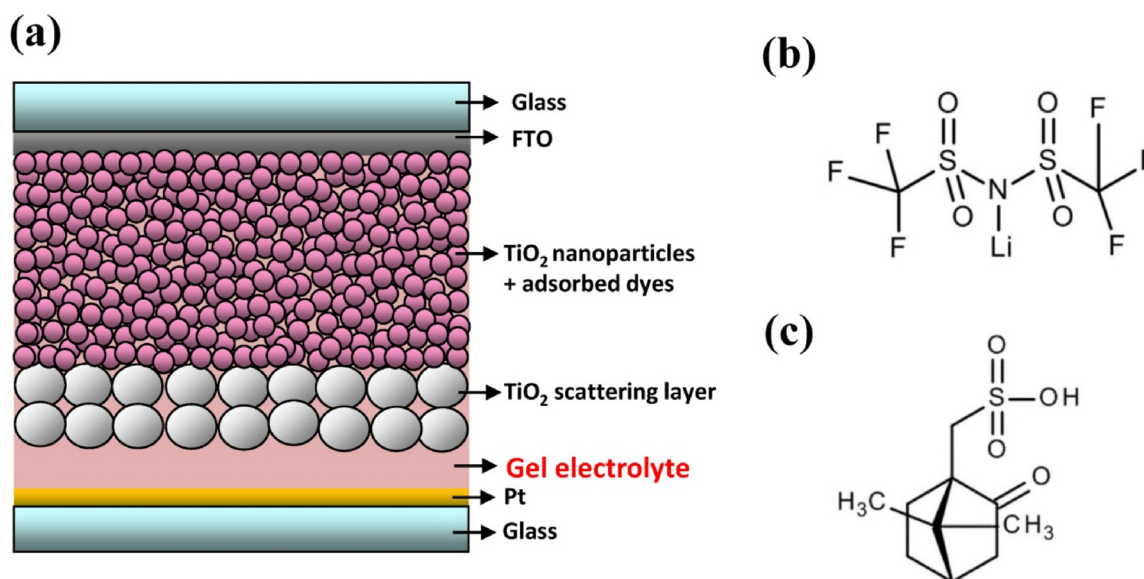


Fig. 1. (a) Device structure of a quasi-solid-state dye-sensitized solar cell. Molecular structures of (b) LiTFSI and (c)  $C_{10}H_{16}O_4S$ .

reduce electrolyte evaporation and leakage, thus extending the life of DSSC devices. By adding bis(trifluoromethane)sulfonimide lithium salt (LiTFSI) and camphorsulfonic acid ( $C_{10}H_{16}O_4S$ ) to gel-based electrolytes, the current study successfully enhanced the conductivity of gel-based electrolytes, further increasing the power conversion efficiency of gel-based DSSCs.

## 2. Experiments

### 2.1. Preparation of gel-based electrolytes

First, a gelator and a liquid-based electrolyte solution were prepared. The gelator was prepared by mixing PMMA and propylene carbonate at a weight ratio of 1 to 2 (i.e., 0.5 g of PMMA to 1 g of propylene carbonate), stirring the mixture for 3 h at 90 °C, and cooling the mixture at 4 °C in a refrigerator for 24 h to remove bubbles from the solution. The liquid-based electrolyte solution contained 0.6 mol/L (M) BMII, 0.05 M LiI, 0.03 M  $I_2$ , 0.5 M 4-*tert*-butyloxyridine, 0.1 M guanidine thiocyanate, and acetonitrile and valeronitrile at a volume ratio of 5:1. Fig. 1(a) shows the device structure of a quasi-solid-state dye-sensitized solar cell. This study examined the effect of mixing gelator and liquid-based electrolyte solution of varying volume ratios on the properties of fabricated devices. Gel-based electrolytes were obtained by mixing the gelator and liquid-based electrolyte solution at the volume ratios of 1:0.5, 1:1, and 1:1.5. The properties of the electrolytes and devices were then analyzed. In addition, the effect of adding LiTFSI (Fig. 1(b)) and  $C_{10}H_{16}O_4S$  (Fig. 1(c)) to the gel-based electrolytes on the device properties was examined. Specifically, LiTFSI (0.05 M, 0.1 M, and 0.15 M) and  $C_{10}H_{16}O_4S$  (0.05 M, 0.1 M, and 0.15 M) were separately added to the gel-based electrolytes to analyze the electrolyte and device properties.

### 2.2. Analyzing the properties of the electrolytes

In this study, a conductivity meter (CON 500) was used to analyze the electric conductivity of the electrolytes. A simple symmetric device was fabricated by encapsulating two identical Pt electrodes in a sealing foil through thermocompression method, with the electrolyte injected between the two electrodes. Through electrochemical impedance spectroscopy (EIS), an electrochemical analyzer was employed to measure the electrochemical impedance of the simple symmetric device. Specifically, the electrolyte–CE interface impedance as well as the

charge transfer impedance in the electrolyte was analyzed.

### 2.3. Fabrication of DSSC working electrodes

The DSSC working electrode was fabricated as follows. First, a piece of fluorine doped tin oxide (FTO) conductive glass was cleaned. Subsequently, strips of 3 M tape with 4-mm-diameter holes punched into them were fixed to the glass. The holes, which had an area of approximately 0.126 cm<sup>2</sup>, served as the coating area of the devices. A layer of 25-nm-sized anatase  $TiO_2$  nanoparticles was coated uniformly onto the FTO substrate by using the doctor blade coating method. The coated substrate was heated for 10 min at 150 °C and allowed to cool to room temperature. The  $TiO_2$  coating procedure was repeated, from which an approximately 12- $\mu$ m-thick working electrode was obtained. Another layer of  $TiO_2$  paste composed of approximately 200-nm nanoparticles was subsequently coated using the same method to serve as the scattering layer. The substrate was then placed inside a furnace and sintered at 500 °C for 30 min. After the  $TiO_2$  electrode was cooled to 80 °C, it was soaked in a prepared dye solution for 24 h and then removed from the dye solution. Regarding the dye solution preparation, 0.5 mM N719 dye was mixed with 0.5 mM chenodeoxycholic acid, with the latter material serving as a coabsorbent in this experiment. Moreover, the solvent was prepared by mixing a 1:1 vol ratio of acetonitrile and *tert*-Butyl alcohol.

### 2.4. Fabrication of DSSC devices

The method for fabricating the DSSC counter electrode is described as follows. A piece of FTO conductive glass with two predrilled holes (for pouring the electrolyte solution) was used as the substrate. The substrate was then washed with deionized water and alcohol for 3 min by using an ultrasonic cleaner. The substrate surface was then dried using nitrogen gas. A Pt nanoparticle paste was uniformly applied onto the FTO substrate by using the doctor-blade coating method. The substrate was placed inside a furnace and sintered at 450 °C for 30 min. The Pt counter electrode was fabricated when the substrate cooled naturally to the room temperature. The method for encapsulating the DSSC devices is presented as follows: A piece of 60- $\mu$ m-thick sealing foil was cut into a 2.5 × 2.5-cm square with a 0.8 × 0.8-cm area cut from the center. The sealing foil was used to assemble the working and counter electrodes. The assembled device was continually pressurized at 3 kg/cm<sup>2</sup> at 130 °C for 3 min to facilitate adhering the upper and lower

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