



Adding graphene nanosheets in liquid electrolytes to improve the efficiency of dye-sensitized solar cells

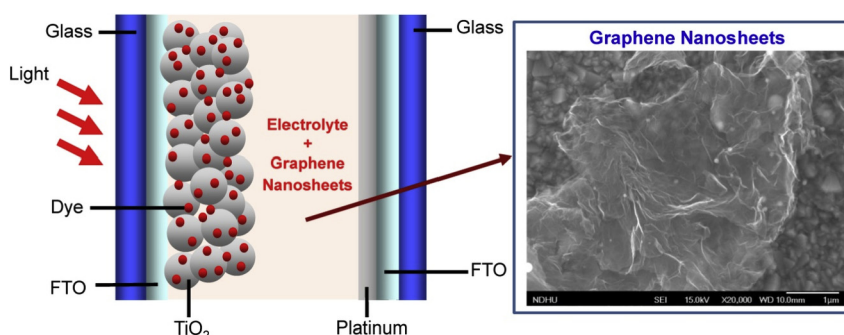
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HIGHLIGHTS

- Graphene nanosheets were added into the liquid electrolytes for DSSCs.
- The graphene nanosheets substantially reduced the impedance in the electrolyte.
- Adding graphene to the electrolyte effectively promoted the efficiency of the DSSCs.
- The DSSC with 20-mg graphene electrolyte attained the highest efficiency of 9.26%.

GRAPHICAL ABSTRACT



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ABSTRACT

In this study, graphene nanosheets (GNs) were added into liquid electrolytes to increase their electric conductivity through the exceptional electron mobility of graphene to improve the efficiency of dye-sensitized solar cells (DSSCs). The nanostructure of the GNs was analyzed through scanning electron microscopy, transmission electron microscopy, and atomic force microscopy. The properties of various electrolytes were examined through conductivity analysis, Raman spectroscopy, Fourier transform infrared spectroscopy, cyclic voltammetry, and the electrochemical impedance spectroscopy of simple symmetric devices. The results indicated that adding graphene to the liquid electrolyte improved the conductivity and electrochemical properties of the electrolyte. Furthermore, DSSCs with various electrolytes were characterized using current density-voltage, incident photon-to-current conversion efficiency, and electrochemical impedance spectroscopy measurements. The results revealed that adding graphene to the electrolyte solutions effectively promoted the efficiency of the DSSC devices; this was because the exceptional electron mobility in the GNs substantially reduced the impedance in the electrolyte solutions. In particular, the DSSC device with the 20-mg graphene electrolyte solution attained the highest efficiency (9.26%), which was 20% higher than that of the device with the non-graphene electrolyte solution.

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1. Introduction

Energy shortages and environmental pollution are two major crises facing humans in the twenty-first century. Renewable energy is obtained from inexhaustible natural resources and does not

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generate pollutants when used. In particular, solar energy is the most advantageous renewable energy, because it is infinite, is not restricted by geographical location, and does not incur environmental pollution. In 1991, Grätzel et al. developed a type of high-efficiency, low-cost, dye-sensitized solar cell (DSSC) [1]. Subsequently, DSSC research has garnered widespread attention. DSSCs exhibit several advantages, such as high efficiency, low fabrication costs, a simple structure, and an easy fabrication process [2–11]. The structures of DSSC devices are composed of transparent conductive substrates, nanoporous titanium dioxide (TiO_2) semiconductor thin films, dyes, electrolytes, and platinum (Pt) counter electrodes (CEs) [12].

Electrolytes are a vital part of DSSC devices. Redox couples in electrolytes provide electrons to oxidized dye molecules, thereby returning excited molecules to their ground states and completing dye renewal. Three types of electrolytes are applied in DSSCs, namely liquid, gel, and solid forms [13–16]. Currently, most electrolytes applied in DSSCs are liquid; solvent volatilization and leakage may occur in liquid electrolytes and thus shorten the lifespans of DSSC devices. Moreover, the poor charge transfer properties in liquid electrolytes reduce their charge transfer speed and thus inhibit the rate of dye reduction and efficiency of DSSC devices. Therefore, recent studies have focused on improving the electrolytes employed in DSSCs [17–23].

Graphene has gained considerable attention among scholars as a novel carbon material in recent years. In 2004, Geim and Novoselov from Manchester University successfully isolated graphene from graphite through exfoliation, confirming that graphene can exist independently; this breakthrough led to their winning the 2010 Nobel Prize in Physics [24]. Due to its unique material properties, such as its excellent mechanical strength (1100 GPa), thermal conductivity ($5000 \text{ W m}^{-1} \text{ K}^{-1}$), and carrier mobility ($200,000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$), graphene has become a focal topic in recent studies, in which it has been employed to fabricate DSSC devices [25–28]. Li et al. dispersed stacked graphene platelet nanofibers (SGNF) in the liquid electrolyte of DSSCs. The results showed that the SGNF can efficiently improve the charge transport and catalytic reactions of the Co(III)/(II) redox couple, and the quinoxaline dye AQ308-based DSSC containing SGNF exhibited a power conversion efficiency (PCE) of 9.81% [29]. Nikolakopoulou et al. suspended trace quantities of graphene by exfoliation of graphite in a variety of organic media, which were used either as electrolyte solvents or as deposition media in sensitized solar cells. The effect of added graphene was investigated in three types of solar cells: liquid electrolyte DSSCs, quasi-solid-state DSSCs, and solid-state quantum dot sensitized solar cells. The result showed that a very tiny quantity of pristine graphene properly suspended in electrolyte solvents or deposition media can enhance the efficiency of solar cells [30].

In the present study, graphene nanosheets (GNs) were added to a liquid electrolyte to increase its electric conductivity through the exceptional electron mobility of the graphene, thereby improving the efficiency of DSSCs. In addition, this study examined how applying different amounts of graphene to a liquid electrolyte affects the electrolyte and DSSC device properties.

2. Experiments

2.1. Preparing the electrolytes

Fig. 1 shows the schematic device structure of a DSSC using the liquid electrolyte containing GNs. The GNs (graphene P-MF10) were purchased from a commercial source (Energe Inc.). To prepare the liquid electrolyte, 798 mg of 1-butyl-3-methylimidazolium iodide (BMII), 38 mg of I_2 , 0.36 mL of 4-tert-butylpyridine (4-TBP), 59 mg of guanidine thiocyanate (GuSCN), 4.25 mL of acetonitrile, and

0.75 mL of valeronitrile were mixed in a sample vial through ultrasonication for 20 min. A total of four bottles of electrolyte solutions were prepared. No GNs were added to one of the solutions, whereas 10, 20, and 30 mg of GNs were added to the other three solutions.

2.2. Analyzing the properties of the electrolytes

The nanostructure of the GNs was analyzed through scanning electron microscopy (SEM) and transmission electron microscopy (TEM). The surface morphology and roughness of the GNs were analyzed through atomic force microscopy (AFM). A conductivity meter was used to analyze the electric conductivity of each electrolyte solution. The material properties of the solutions were analyzed through Raman spectroscopy, and the chemical bonds in the solutions were observed through Fourier transform infrared spectroscopy (FTIR). In addition, a cyclic voltammetry (CV) analysis was conducted to examine the catalytic properties of the solutions via a three-electrode system; the three electrodes were a working electrode (Pt), a CE (Ag/Ag^+), and a reference electrode (Pt wire). The scanning speed was 10 mV/s, and the voltage ranged from -0.4 to 1.6 V. A simple symmetric device was fabricated by encapsulating two identical Pt electrodes in a sealing foil through thermocompression, with the electrolyte injected between the two electrodes. Through electrochemical impedance spectroscopy (EIS), an electrochemical analyzer was used to measure the electrochemical impedance of the simple symmetric device based on various electrolytes. Specifically, the electrolyte/CE interface impedance and the charge transfer impedance in the electrolyte solutions were analyzed.

2.3. Fabricating the DSSCs

To fabricate the DSSC working electrodes, an FTO glass substrate was first cleaned, and strips of 3M tape with 3-mm-diameter circular holes (an area of approximately 0.07 cm^2) were fixed to the substrate to indicate the coating region. The blade coating method was employed to evenly coat 25 nm of TiO_2 paste onto the FTO glass substrate, which was then heated to 150°C for 10 min and cooled to room temperature. The TiO_2 coating process was then repeated once to obtain a 12- μm -thick working electrode. To form a scattering layer, the blade coating method was again employed to coat a layer of TiO_2 paste containing particles approximately 200 nm in size. The substrate was then heated to 500°C in a high-temperature furnace for 30 min. After the TiO_2 electrode cooled to 80°C , the working electrode was soaked in a dye solution for 24 h. The dye solute was prepared by mixing 0.5 mM N719 dye with 0.5 mM chenodeoxycholic acid as the coabsorbent; the solvent was a 1:1 mixture of acetonitrile and tert-Butyl alcohol.

Another FTO conductive glass substrate was used to fabricate the CE. Two small holes of 1-mm diameter each were predrilled on an FTO substrate in advance of injecting the electrolytes. The substrate was then sonicated with deionized water for 3 min, sonicated again with alcohol for 3 min, and dried with nitrogen gas. The blade-coating method was again employed to evenly coat the paste of the chemical Pt precursor on the FTO substrate, which was sintered to 450°C in a high-temperature furnace for 30 min and left to cool naturally to room temperature, thus completing the fabrication of the Pt CE.

To encapsulate a DSSC device, 60- μm -thick sealing foil was cut into $2.5 \times 2.5 \text{ cm}$ squares, with a $0.8 \times 0.8 \text{ cm}$ section cut out of the center. The foil was used to encapsulate the working electrode and CE, which were pressurized for 3 min at a temperature of 130°C and a pressure of 3 kg/cm^2 . After the encapsulated device was left to cool, a micropipette was used to inject approximately 5 μL of

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