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Highly efficient and stable dye-sensitized solar cells based on nanographite/polypyrrole counter electrode

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ABSTRACT

Nanographite/polypyrrole (NG/PPy) composite film was successfully prepared via in situ polymerization on rigid fluorine-doped tin oxide substrate and served as counter electrode (CE) for dye-sensitized solar cells (DSSCs). The surface morphology and composition of the composite film were characterized by scanning electron microscopy (SEM), X-ray diffraction (XRD), Raman spectra and Fourier transform infrared spectroscopy (FTIR). The electrochemical performance of the NG/PPy electrode was evaluated by cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS). The results of CV and EIS revealed that the NG/PPy electrode possessed excellent electrocatalytic activity for the reduction reaction of triiodide to iodide and low charge transfer resistance at the interface between electrolyte and CE, respectively. The DSSC assembled with the novel NG/PPy CE exhibited an enhanced power conversion efficiency of 7.40% under full sunlight illumination as comparing to that of the DSSC based on sputtered-Pt electrode. Thus, the NG/PPy CE could be premeditated as a promising alternative CE for low-cost and high- efficient DSSCs.

1. Introduction

Dye-sensitized solar cell (DSSC) as a novel and powerful photovoltaic device, has achieved several advancements after two decades of research [1–3]. Typically, a DSSC mainly consists of three primary components: a photoanode (a dye sensitized oxide semiconductor), an electrolyte containing a redox couple, and a counter electrode (CE). As a crucial component, the material of an ideal CE should be of excellent electrocatalytic activity and high conductivity for the redox reaction, low overpotential, and long-term stability in the electrolyte. Currently, the most widely used CEs in DSSC are fluorine-doped tin oxide (FTO) or indium-doped tin oxide (ITO) glass substrates coated with platinum (Pt). However, the cost and the problem of Pt dissolution in corrosive electrolyte restricted the large-scale application of DSSCs [4]. In order to resolve the issues, several catalytic materials have been studied, such as carbon materials [5,6], conducting polymers [7,8], and composite materials [9,10]. Among of them, nanographite particles (NG), as a common member of the carbonaceous family, possesses much similar structure to graphene but low-cost, and has attracted extensive attention during recent years in the fields of microelectronic and optoelectronic devices, energy storage and conversion materials due to its unique features of the strongest mechanical strength, high electrical and thermal conductivity, large surface area, and high optical transmittance [11,12]. However, it may not to be an appropriate CE material for DSSC in the view of the limited active sites for I_3^- reduction reaction [13]. Conducting polymers, with high conductivity, low-cost, large electrochemical surface area, and good electrocatalytic activity for I_3^- reduction [11,12], has became promising candidates as CE materials in DSSC. For example, polypyrrole (PPy), as a typical conductive polymer, has been intensively studied for its advantages of intrinsic electrical conductivity, catalytic performance, easy to synthesis and low price [14]. For instance, Wu, et al., [15] have obtained PPy nanoparticles as CE catalyst and got good performance in DSSCs. In our previous reported [7], a composite CE composed of poly (3, 4-ethylenedioxythiophene):polystyrenesulfonate and PPy prepared by electrochemical polymerization showed a good catalytic performance in I^-/I_3^-electrolyte and an improved photovoltaic performance for DSSC. Yue, et al., [16] further demonstrated an enhanced performance of DSSC based on a molybdenum disulfide/graphene composite catalytic film prepared by hydrothermal route. In these works, the electrocatalytic activity of polymers and

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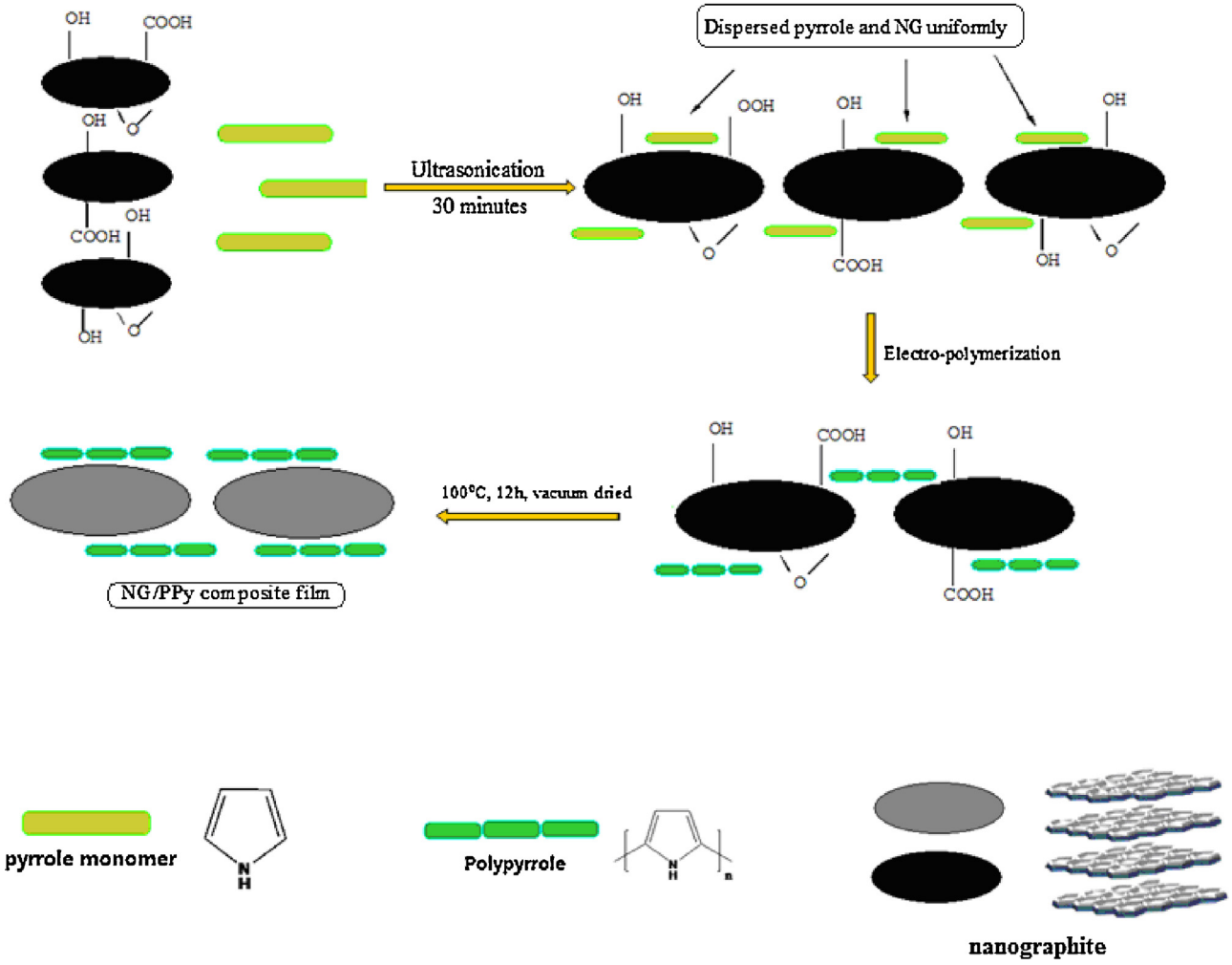

Scheme 1. The synthesis route of the NG/PPy composite film.

sulfide was remarkably promoted due to their synergistic catalytic effect.

Herein, we designed and prepared a novel composite film of nanographite particles modified polypyrrole as CE for DSSC by using electrochemical deposition method, by which the obtained film could possess excellent electrocatalytic activity and high conductivity. Cyclic voltammetry (CV), electrochemical impendence spectroscopy (EIS), and Tafel polarization measurement were employed to study the electrocatalytic activities of the nanographite/polypyrrole (NG/PPy) CE. The DSSC with NG/PPy CE exhibited an enhanced photovoltaic conversion efficiency of 7.40% at 1 sun illumination (AM 1.5G simulated solar light).

2. Experimental

2.1. Materials

The Lithium perchlorate ($LiClO_4$), oxalic acid ($C_2H_2O_4$), titanium tetrachloride ($TiCl_4$) and pyrrole were purchased from Shanghai Chemical Agent Ltd., China. The organometallic compound sensitized dye N-719 Cis-di(isothiocyanato)-bis-(2,2′-bipyridyl-4,4′-dicarboxylato) ruthenium(II) bis-tetrabutylammonium was obtained from Solaronix SA (Switzerland). Nanographite with purity of 99.9% was purchased from UNI-ONWARD Corp., Taiwan. Pyrrole monomer was distilled prior to use. All reagents were of analytical reagent grade.

The FTO glass substrates (8 $\Omega \cdot cm^{-2}$, Hartford Glass Co., USA) were cut into pieces with size of 1×2 cm^2 carefully and ultrasonically cleaned sequentially in detergent, acetone and distilled water for 10 min, respectively, which later stored in isopropyl alcohol.

2.2. Preparation of NG/PPy counter electrode

Briefly, the NG/PPy CE was prepared by using the electrodeposition method which outlined below. The electrodeposition was carried out with an electrochemical analyzer system (CHI660D, Shanghai Chenhua Device Company, China). All experiments were implemented in a three-electrode cell, including one Pt foil as CE, one Ag/AgCl electrode as reference electrode, and FTO glass with an exposed area of 1 cm^2 as working electrode. The base electrodeposition solution consisted of 0.1 M of pyrrole, 0.1 M of $LiClO_4$, 0.1 M of oxalic acid and nanogarphite in 50 ml deionized water and treated by ultrasonication for 30 min. A constant current density of 10 $mA \cdot cm^{-2}$ was served for electrodeposition (the synthesis route shown in Scheme 1). The FTO glass covered with NG/PPy composite film was put into an oven at 100 °C for 12 h, and then the NG/PPy CE was obtained.

2.3. Fabrication of DSSC

The TiO_2 anode was prepared as described previously [17,18]. The dye was loaded by immersing the TiO_2 anode in the 0.3 mM