

Investigating the role of graphene in the photovoltaic performance improvement of dye-sensitized solar cell



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ABSTRACT

This article compares and describes the photovoltaic performance of dye-sensitized solar cell (DSSC) consisting of platinum (Pt) and graphene-coupled-platinum based counter electrodes (CE). The power conversion efficiency of DSSC could be enhanced by 36% with multi-layer graphene flakes (MGF)/Pt CE as compared to Pt CE. Electrochemical impedance spectroscopy and cyclic voltammetry analysis show that DSSC with an MGF/Pt CE exhibits a higher electro-catalytic activity with lower series resistance. Two-diode model has been used to extract the recombination governing and the performance indicating parameters of a DSSC. An enhancement of $\approx 76\%$ in short-circuit current of MGF/CE based DSSC, as compared to Pt CE, could be attributed to optical and catalytic properties of graphene. This study provides an insight into electronic transport mechanism of DSSC, which changes under the influence of highly catalytic materials such as graphene.

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1. Introduction

Recently graphene has been widely studied for its applications in various fields, like nanoelectronics [1,2], sensors [3,4], composites [5,6], batteries [7], supercapacitors [8], hydrogen storage [9], solar cells [10–12] etc. Many researchers have used graphene and its composites for improving the performance and reducing the cost of dye-sensitized solar cells (DSSCs) [10–19]. A power conversion efficiency of 12.3% has been obtained from liquid electrolyte-based DSSC [19]. A recent study conducted by Burschka et al. [20] has shown a power conversion efficiency of 15% for the solid-state titanium dioxide (TiO₂) mesoscopic solar cells based on perovskite absorber. To uplift the technology of DSSC for commercialization, there is a need for low-cost alternatives to platinum (Pt) based counter electrode (CE) which meets the requirement of good electrocatalytic activity and rapid charge transfer ability toward triiodides (I₃⁻). The unique optical and electrical properties of graphene have invited a lot of attention as a potential candidate to reduce consumption of rare and expensive Pt in DSSC. Recently, Scheuermann et al. [21] reported an improved

catalytic activity of palladium (Pd) nanoparticles (NP) deposited on the graphene sheets for the Suzuki–Miyaura coupling reaction. Graphene has been used in combination with Pt as a counter electrode to improve the conductivity, surface area and catalytic activity by many researchers [22,23]. Most of these findings show significant increase in short-circuit current (≈ 1.3 -fold in some cases [24]), which cannot be explained only on the basis of catalytic activity of graphene. These studies are limited to chemical, physical and electro-catalytic behavior of graphene based CE, but a detailed theoretical framework to support experimental findings is lacking in the reported literature. To surmount such issues, an in-depth understanding of electrical and morphological characteristics of CE supported by theoretical framework is essential, which can offer an insight on the possible ways of improving the DSSC performance. Keeping in view above-mentioned shortfalls, we have undertaken a detailed study on: (a) two-diode model to explain non-linear *I*-*V* characteristics of DSSC, (b) validation of the developed model using actual performance parameters of DSSC fabricated in our laboratory, (c) effect of illumination on the performance of the DSSC and (d) the effect of recombination mechanism during electrolytic charge diffusion.

In this article, multi-layer graphene flakes (MGF) have been used in combination with Pt CE to study their electro-catalytic and optical behavior in performance improvement of the DSSC. Electrochemical impedance spectroscopy (EIS) and cyclic voltammetry

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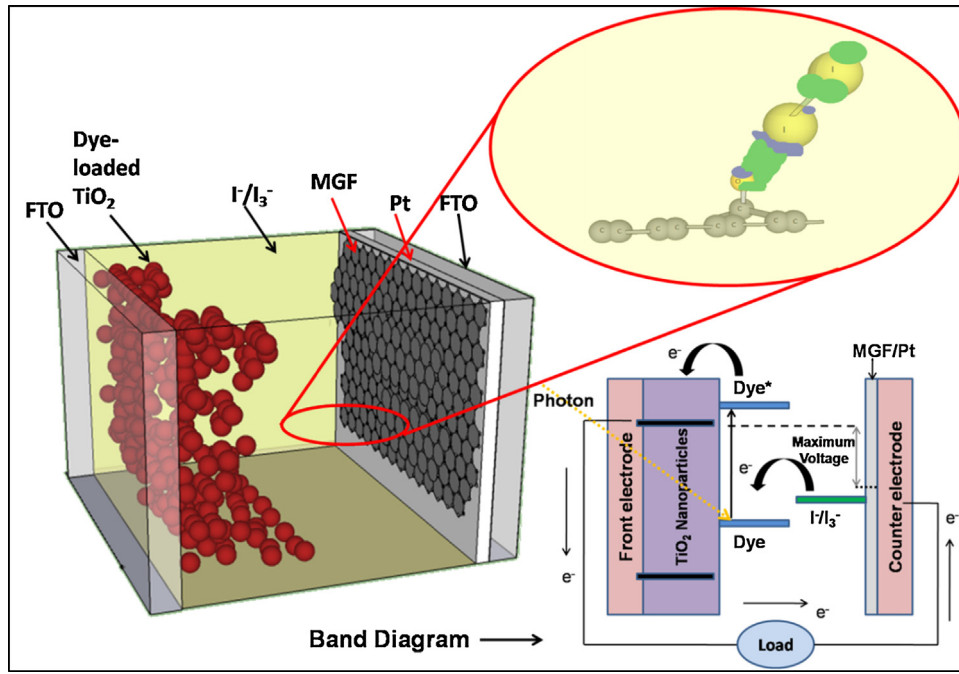


Fig. 1. Schematic diagram of DSSC with corresponding band structure (insert) to show electron transport and charge transfer between molecular iodine and MGF oxidized with functional groups.

(CV) analysis show that the DSSC with a MGF/Pt CE exhibits a higher electro-catalytic activity with a lower series resistance as compared to the one made of Pt CE. The reflection enhancement from the MGF loaded CE is explored as another reason for the performance improvement of DSSC. The theoretical modeling of DSSC based on MGF/Pt CE is validated with experimental data to explore the role of graphene in light trapping and catalytic action.

2. Theoretical model

The charge flow mechanism in a working DSSC is mainly governed by the relative kinetic rates of several electron transfer steps at various interfaces. Fig. 1 illustrates the schematic diagram of DSSC with a corresponding band diagram of TiO₂, dye, electrolyte and CE. The detailed working mechanism of DSSC is given in our previous work [25–27]. The sequence of electron transfer and charge-transport processes, which forms the basis of the DSSC's operation mode [28], is given in Supplementary Information (SI). The long-range intramolecular charge transfer has been investigated by time-dependent density functional theory (TDDFT) for various dye molecules, which is useful for finding the charge-transfer excitation energies and oscillator strengths of donor-bridge-acceptor systems [28,29].

For the case of highly active catalytic counter electrodes, like graphene, Eq. (A.4) plays a very crucial role toward performance improvement of DSSC. In order to quantify the effect of such catalytic electrodes Eq. (A.4) must be understood in terms of rate of reaction (r):

$$r = -\frac{d[I_3^-]}{dt} = -\frac{1}{2} \frac{d[e_{CB}^-]}{dt} = \frac{1}{3} \frac{d[I^-]}{dt} \quad (1)$$

The negative sign in Eq. (1) indicates the consumption/conversion of reactants to products. The second term (i.e., $-(1/2)(d[e_{CB}^-]/dt)$) defines current at the counter electrode (I_{CE}), which depends on total surface area of the catalyst. By increasing the surface area using graphene-flakes/metal-nanoparticles, the reaction rate may increase leading to higher photo-generated current collection from the DSSC. This regeneration mechanism is

influenced by the catalytic action of different types of counter electrodes [30]. The catalytic activity of graphene based CE increases as the number of oxygen functional groups increases thus resulting in higher power conversion efficiency [24].

Under a steady-state condition of illuminated DSSC, the electron injection from excited dye molecules, transport in the mesoporous semiconductor (TiO₂) thin film, and recombination with electrolyte at the TiO₂/electrolyte interface can be described by the electron diffusion differential equation [31–35] as given in SI. The general solution of Eq. (A.8) in terms of I - V relationship, with short-circuit current and open-circuit voltage boundary conditions, can be expressed as below:

$$I = I_{ph} - I_i \left\{ \exp\left(\frac{q(V + IR_S)}{nk_B T_C}\right) - 1 \right\} - I_r \left\{ \exp\left(\frac{q(V + IR_S)}{mk_B T_C}\right) - 1 \right\} - \frac{V + IR_S}{R_{sh}} \quad (2)$$

$$I_{ph} = [I_{SC} + K_1(T_C - T_{Ref})]\lambda \quad (3)$$

$$I_{SC} = \frac{\dot{C}C(r_{CE}, I_{CE})q\Phi_0 L_n \alpha [-L_n \alpha \cosh(d/L_n) + \sinh(d/L_n) + L_n \alpha \exp(-d\alpha)]}{A(1 - L_n^2 \alpha^2) \cosh(d/L_n)} \quad (4)$$

where I_{ph} is the photo-generated current, λ is the incident light in kW/m², I_i and I_r are the saturation current of the rectifying and recombination diodes, respectively, n and m represent diode ideality factor, q represents electron charge, k_B is the Boltzmann constant, $\dot{C}C(r_{CE}, I_{CE})$ is current enhancement coefficient that depends on the reflection from counter electrode (r_{CE}) and the enhanced current due to catalysis at counter electrode (I_{CE}), T_C is the cell temperature (≈ 298.14 K) and T_{Ref} is the reference temperature (≈ 298.14 K), L_n is the electron diffusion length equal to $\sqrt{D\tau}$; d is the mesoporous TiO₂ layer thickness; and A is the area of the DSSC. Based on the theoretical model described above, the DSSC is simulated using MATLAB/Simulink.

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