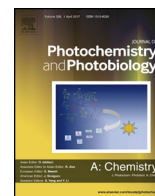




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Graphene-wrapped iron carbide nanoparticles as Pt-free counter electrode towards dye-sensitized solar cells via magnetic field induced self-assembly

Hongxia Xu^{a,1}, Chuanjian Zhang^{a,1}, Jianhua Yao^a, Shuping Pang^a, Xinhong Zhou^{b,*},
Guanglei Cui^{a,*}

^a The Qingdao Key Lab of Solar Energy Utilization and Energy Storage Technology, Qingdao Institute of Bioenergy and Bioprocess Technology, Chinese Academy of Sciences, Qingdao, 26610, People's Republic of China

^b College of Chemistry and Molecular Engineering, Qingdao University of Science and Technology, No. 53 Zhengzhou Road, Qingdao, 266042, People's Republic of China

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ABSTRACT

Graphene-wrapped iron carbide nanoparticles ($\text{Fe}_3\text{C@G}$) are employed as an alternative counter electrode to Pt in dye-sensitized solar cell via a convenient magnetic field induced assembly method. Benefit from the novel binder-free electrode fabrication route, the $\text{Fe}_3\text{C@G}$ nanosheets orderly are deposited on the FTO glass to form an effective catalyst layer with both favorable catalytic ability and electron migration rate. Comparable photovoltaic conversion efficiency with Pt endows the $\text{Fe}_3\text{C@G}$ a promising counter electrode for low cost but high performance dye-sensitized solar cells. Moreover, the magnetic field induced assembly method also shows potential application for other magnetic materials towards counter electrode.

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1. Introduction

Since the first innovative prototype proposed by O'Regan and Grätzel in 1991, dye-sensitized solar cells (DSSCs) have attracted extensive attention due to high power conversion efficiency and low cost production [1–3]. As a key component in DSSCs, the counter electrode (CE) plays the role of collecting and transferring the electrons from the external circuit and catalyzing the reduction of triiodide ions to iodide ions. Consequently, an ideal CE should possess a high electrical conductivity and excellent catalytic activity. Platinum (Pt) could be regarded one of the best CE materials because it satisfies all the requirements mentioned above. However, as one of the world's scarcest materials, platinum is the most expensive component in DSSCs which limit mass production of DSSCs. To solve this problem, several of Pt-alloy materials have been developed which showed favorable performances [4–6]. Moreover, there are a variety of effective Pt-free counter electrode materials being investigated worldwide including carbonaceous materials [7–9], transition metal compounds

[10–12] and conductive polymers [13,14]. Among these, transition metal compounds are considered as attractive catalytic materials for their low cost, high catalytic activity and good thermal stability under harsh conditions [15,16]. As an excellent CE material, not only the superior electrocatalytic ability, excellent electrical conductivity is highly required. However, Generally, abundant grain boundaries and defects of small transition metal compounds particles depress electron transport efficiency. One effective route is to introduce highly conductive materials to improve the electrical conductivity of CE materials. For instance, graphene (G) known for its excellent conductive two-dimensional network is considered as a superior media to incorporate with the catalyst for CEs [17]. The other promising approach to enhance the electrodes' electron diffusion rate is to construct binder-free counter electrodes. Because the inert binders in the electrodes can lower the catalytic and conductive capabilities of the CEs by reducing the active area of the active materials and blocking the corresponding electron transport. The most successful strategies for binder-free CE fabrication are electro-deposition [18,19] and hydrothermal method [11,20]. Radio frequency (RF) magnetron sputtering [21] and scalable spray pyrolysis technique [22] were also employed for highly conductive CE preparation. However, these routes are all highly energy-consuming. It is still a big challenge to fabricate high

* Corresponding authors.

E-mail addresses: zxhxx2008@163.com (X. Zhou), cuiql@qibebt.ac.cn (G. Cui).

¹ Authors contributed equally to this work.

performance CE using a cheap catalyst material via a low-cost but effective route.

Iron carbide (Fe_3C), also known as cementite with very low cost, is always considered as a very important component in metallurgy. Recently, several groups have reported that Fe_3C is an excellent Pt-free catalyst for oxygen reduction [23–28]. At the same time, both our and Fu's group [29] have proved that a $\text{Fe}_3\text{C}/\text{C}$ nanocomposites exhibited a comparable catalytic ability for I_3^-/I^- redox couple to Pt [30]. However, all the Fe_3C CEs reported were fabricated by mixing the catalyst materials and binder to form slurry followed by coated on the FTO glasses via doctor blade method. The electron transport rate may be influenced due to the presence of catalytic inactive components. It is worth noting that Fe_3C exhibits

magnetism which drives us to explore the feasibility of magnetic field induced self-assembly CE [31]. Inspired by this, we reported herein a graphene-wrapped Fe_3C nanoparticles hybrid material ($\text{Fe}_3\text{C}@G$) for Pt-free and effective CE in DSSCs by magnetic field induced assembly method. A catalytic $\text{Fe}_3\text{C}@G$ layer was deposited on the conductive surface of FTO glass by magnetic field induced assembly and was employed as CE. It is demonstrated that the $\text{Fe}_3\text{C}@G$ CE not only exhibited its effective intrinsic catalytic activity for I_3^-/I^- redox couple but also possessed a favorable electrical conductivity due to the introduction of conductive graphene nanosheets. More interestingly, the $\text{Fe}_3\text{C}@G$ CE ($\text{Fe}_3\text{C}@G\text{-M}$) fabricated via magnetic field induced assembly method displayed a superior electrocatalytic activity compared with the

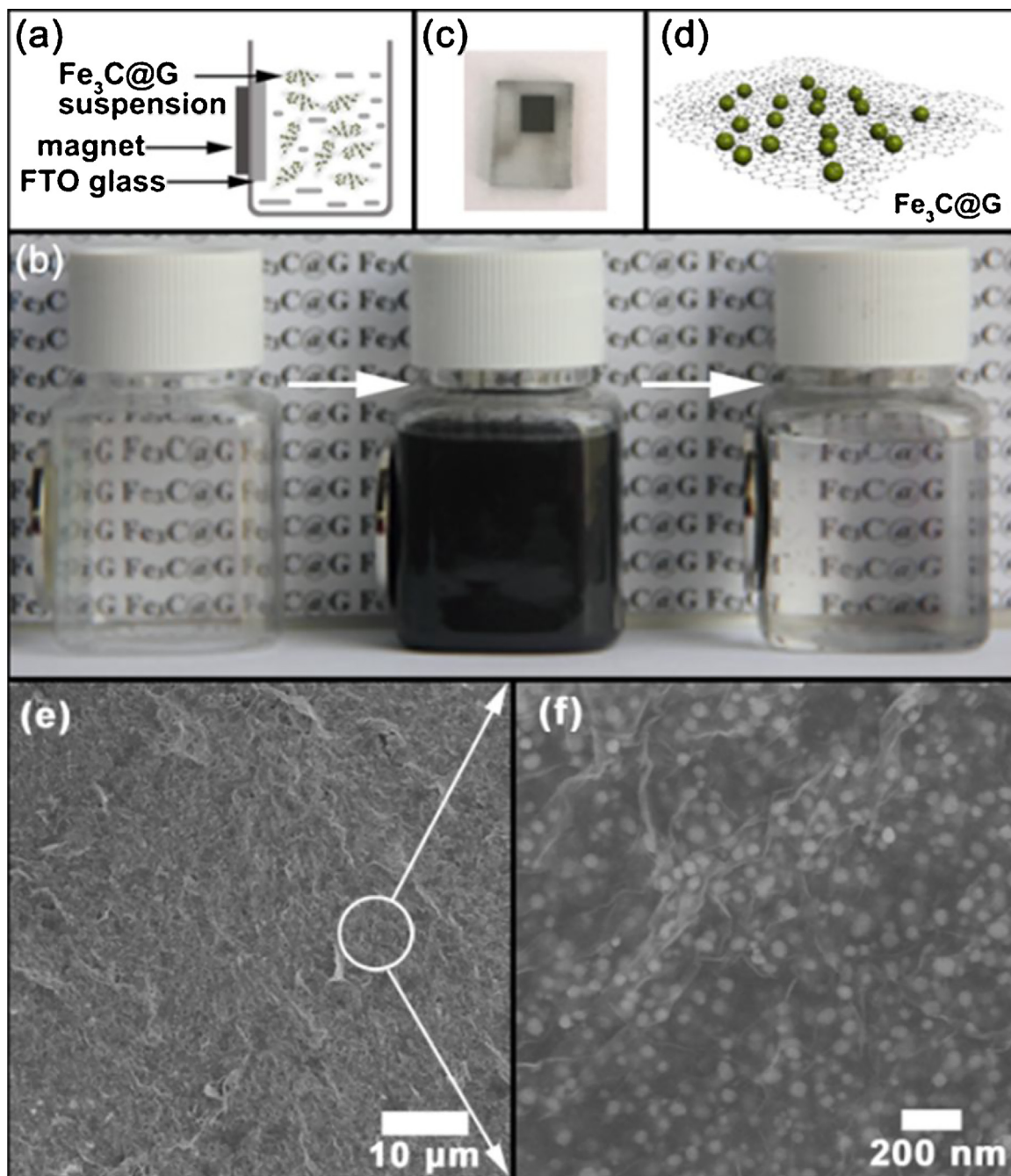


Fig. 1. (a) Schematic illustration and (b) the digital photographs of the preparation route of $\text{Fe}_3\text{C}@G\text{-M}$ counter electrode fabricated via magnetic assembly method, (c) Digital photographs of the as-synthesized $\text{Fe}_3\text{C}@G\text{-M}$ counter electrode, (d) Structure schematic of the $\text{Fe}_3\text{C}@G\text{-M}$ hybrid materials, Top-view (e) and corresponded enlarged (f) SEM images of the $\text{Fe}_3\text{C}@G\text{-M}$ counter electrode.

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