



COMMUNICATION

Dual integration system endowing two-dimensional titanium disulfide with enhanced triiodide reduction performance in dye-sensitized solar cells



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Abstract

State-of-the-art dye-sensitized solar cells (DSSCs) usually utilize noble and scarce Pt as counter electrodes to catalyze the reduction of triiodide in electrolyte, which largely hinders the practical applications of DSSCs. Accordingly, alternatives with low cost, excellent electrocatalytic activity, and superior electrochemical stability to Pt are highly sought after. Herein, we report novel two-dimensional titanium disulfide nanosheets assembled and decorated on graphene (TiS₂-G) through an integrated strategy of ball milling and high temperature annealing process. Benefiting from combined characteristics, when firstly applied as counter electrode, the TiS₂-G hybrids demonstrate superior electrocatalytic activity towards the triiodide reduction with a power conversion efficiency of 8.80%, outperforming the Pt reference (8.00%). The high catalytic activity of TiS₂-G hybrids is ascribed to synergetic effects derived from the dual integration system of highly electroactive TiS₂ species and G functioning as conductive matrix. Most importantly, the as-made TiS₂-G hybrids also deliver outstanding electrochemical stability. The present work provides an effective strategy to engineer the highly active and low cost replacement to noble Pt.

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Introduction

As the continuously increasing consumption of fossil energy, utilization of renewable solar energy has attracted enormous attention. State-of-the-art dye-sensitized solar cells (DSSCs), as

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outstanding representatives for next generation solar cells, have been widely investigated owing to their easy fabrication, low cost, and environmental friendliness [1-3]. In the case of DSSCs, counter electrodes (CEs) play vital parts in collecting electrons derived from conduction band of TiO_2 photoanodes, and catalyzing the reduction of triiodide (I_3^-) to iodide (I^-) [4]. However, the large-scale applications of DSSCs are still restricted because of employing noble metal Pt as CEs, which is costly, resource-limited, and electrochemically unstable [5,6]. Consequently, searching for inexpensive Pt alternatives with high electrical conductivities, superb electrocatalytic activity, and excellent electrochemical stability for DSSCs is highly desired. It is commonly believed that carbon materials would be promising candidates to Pt CEs due to its intrinsically economical feasibility, easy availability, large surface area, and robust stability in electrochemical environment [7-9]. Up to now, various carbonaceous alternatives, such as fullerene [10], carbon black [11-13], carbon nanoparticles [14,15], carbon nanotubes [16,17], and carbon nanofibers [18,19] have been explored to replace Pt. Nevertheless, these materials available now still suffer from poor electrocatalytic activity towards I_3^- reduction, thus leading to inferior power conversion efficiency (PCE) [20-22]. With this in mind, exploitation of carbon-based materials with electrocatalytic activity being comparable to or outperforming Pt reference is indispensable for high performance DSSCs, which is closely related to the structural design of CE materials [23,24].

Graphene (G), a new single-layer sp^2 carbon network with two-dimensional (2D) honeycomb carbon lattice, has been tremendously popular in both scientific and manufacturing fields due to its unique physical and chemical properties [25,26]. It is believed that G possesses two distinct regions that affect the bulk electrochemical activity, which are the basal planes and the edge planes [27,28]. Many reports suggested that the G edge planes could contribute to stronger electrocatalytic activity than that of the basal planes [29-31]. To enhance the electrocatalytic activity of bulk G, activation of G basal planes is one of efficient strategies, such as drafting active sites over G, doping heteroatoms into G backbone, and tailoring G to small-sized materials [32-37]. Chen et al. synthesized titanium nitride decorated nitrogen-doped graphene hybrids [38]. When applied as CEs for DSSCs, the hybrids showed a high PCE of 5.78%, outperforming Pt (5.03%). In our previous work, we reported the nitrogen-doped graphene nanoribbons with rich edge sites and fully exposed active basal planes [39], revealing excellent electrocatalytic activity and prominent electrochemical stability, as well as the unique roles of nitrogen species in the reduction of I_3^- . In this regard, increase of exposed G edge planes and activation of G basal planes are efficient strategies to configure high-performance G-based CEs for DSSCs.

As an important class of inorganic compounds, transitional metal dichalcogenides (TMDs) have drawn considerable attention due to their unique layered structure, distinct electronic and optical features [40-42]. In general, TMDs have a formula MX_2 , where M is transition metal element from group 4 to group 10 (Ti, Nb, Mo and so on) and X is a chalcogen (S, Se, Te). These versatile physicochemical properties of TMDs endow themselves with great potential in catalysis, energy storage, and sensing [43-45]. Lately, various TMDs have been reported, and exhibit excellent electrocatalytic activity, such as molybdenum disulfide (MoS_2) [46-48], tungsten disulfide (WS_2) [49-51],

molybdenum selenide (MoSe_2) [52,53], and tungsten selenide (WSe_2) [54,55], etc. However, the inefficient charge transfer rate of TMDs, originating from their band gap structure, largely impedes their further applications. It is believed that such issues can be improved or partially avoided by combining TMDs with conductive G matrix. Lin et al. prepared MoS_2 species dispersed on graphene nanosheets, showing Pt-like performance due to synergistic effects between active MoS_2 and conductive graphene, when employed as electrocatalysts for I_3^- reduction [56]. Wen and coworkers synthesized the MoSe_2 /graphene composites [57], demonstrating improved catalytic activity in other electrochemical systems in comparison to bare MoSe_2 species. Therefore, introducing these electrocatalytically active TMDs into the G matrix to make hybrid electrodes would be one of efficient strategies to increase the activity of buck G. On the other hand, there has little effect on the conductivity of the G, thus enhancing the catalytic activity and elevating the conversion efficiency of DSSCs.

Herein, we report a facile and efficient strategy to synthesize 2D titanium disulfides nanosheets decorated on G hybrids (TiS_2 -G) through ball milling coupled with high-temperature annealing method. By combining the catalytically active TiS_2 nanosheets with conductive G matrix, the as-made TiS_2 -G hybrids demonstrate synergistically enhanced electrochemical performance for reduction of I_3^- , leading to a superb PCE of 8.80%, outperforming Pt reference (8.00%). Furthermore, outstanding electrochemical stability of TiS_2 -G hybrids can also be achieved, indicative of great potential for replacing Pt reference in DSSCs.

Experimental section

Synthesis of graphene oxide (GO)

GO was synthesized through a modified Hummers method reported everywhere. For a typical run, graphite powder (5 g) and sodium nitrate (NaNO_3 , 2.5 g) were added into a beaker (1 L) containing 130 mL of concentrated sulfur acid (H_2SO_4 , 98%) in an ice bath. After stirring for 2 h, 15 g of potassium permanganate (KMnO_4) was carefully added, and another 2 h was needed to stir. Then, the beaker was transferred to a constant temperature water bath and kept for stirring at 35 °C for 1 h. Thereafter, 230 mL deionized water was added into the mixture by dropwise and stirred at 98 °C for 30 min. Additional 400 mL deionized water and 10 mL hydrogen peroxide (H_2O_2) were poured into the solution. The reaction mixture was centrifuged and washed by deionized water until the pH was neutral, then the GO dispersion was achieved.

Preparation of G, TiS_2 -G hybrids, and TiS_2 nanosheets

200 mg GO dried via lyophilization was grinded into powder, and placed into a stainless steel capsule (100 mL) containing 75 g of stainless steel balls (diameter, 5 mm), which was conducted in a glove box filling with argon. The capsule was then sealed and fixed in the planetary ball-mill machine, and rotated with 400 rpm for 12 h. The GO was transferred from the container to a quartz boat. Then, the G with abundant edge sites was achieved through annealing at 650 °C for 2 h under nitrogen.

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