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Simultaneous formation of carbon nanosheet and loading of platinum nanoparticle to form platinum/carbon nanosheet composite and its electrocatalytic activity



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ABSTRACT

Platinum (Pt)/two-dimensional (2D) carbon nanocomposites with high catalytic performance and stability are mostly fabricated through two-step route, in which the 2D carbon supports are pre-prepared and then Pt nanoparticles are disposed on the surface of 2D carbon supports. Such method needs tedious processes to preprepare 2D carbon substrates and the binding between Pt and carbon might be weak. Herein, we report the synthesis of the Pt/carbon nanosheet (Pt/CNS) nanocomposites via a hydrothermal method, in which the formation of CNSs and the loading of Pt nanoparticles occurred almost simultaneously. We find that hydrogen bond can promote Pt nanoparticles anchored and dispersed uniformly on the surface of CNS. The good synergistic effect between Pt and CNS greatly improved the electrocatalytic performance for ethanol oxidation.

1. Introduction

Pt-based nanocomposites are most widely researched catalysts in direct alcohol fuel cell for the high catalytic performance and stability of Pt [1]. However, the Pt-based catalysts commonly have a high Pt content, which retards the extensive applications of them due to the high price of precious Pt [2]. In addition, it is easy to be poisoned by CO and CHx intermediate species because of strong adsorption. To reduce the amount of noble Pt while keeping its high catalytic performance, one feasible way is to decrease the size of Pt nanoparticles and anchor them on a substrate. A good substrate can not only bind Pt nanoparticles tightly to avoid their aggregation but also exert some synergic effects of different materials. Carbon materials are becoming the preferential supports due to their low cost, high chemical stability and good conductivity [3]. Among them, graphene is expected to be the most promising substrate for its high surface area, extraordinary conductivity capacity and excellent mechanical strength [4]. The common method for the preparation of Pt-graphene nanocomposites commonly involves two steps. Graphene is first ready and then Pt nanoparticles are deposited on its surface by chemical [5] or electrochemical deposition [6,7], or thermal decomposition of Pt precursors [8]. There are generally several disadvantages in the two-step preparation method. Due to the two-dimensional (2D) nature, graphene tends to stack together in the preparation of nanocomposite, which will lead to the decreasing of surface area, and then block the active sites of Pt nanoparticles and increase the resistance to mass transfer [9]. Though ultrasonic treatment is performed for a long time before the deposition of Pt nanoparticles, stacking can't be avoided absolutely. As a solution, three-dimensional (3D) architectures of graphene are developed [6,9]. However, the preparation of them is somewhat tedious and toilsome. Besides, to immobilize the Pt nanoparticles on the surface of graphene, surface modification is necessary to introduce active functional groups, which would bind with Pt and might play a part in the electro-catalysis.

In this paper, a hydrothermal method is reported to synthesize Pt/ carbon nanosheet (Pt/CNS) nanocomposites, in which the formation of carbon nanosheets (CNS) and loading of Pt nanoparticles occurred almost simultaneously. At very low amount of Pt loading (0.1 wt% Pt), the nanocomposite exhibited good activity for the electro-catalysis of ethanol (EtOH). The method was developed from our previous work [10] to prepare CNS and the obtained CNS possessed oxygen-containing group and graphitic characters, which might facilitate the immobilization of Pt nanoparticles and electro-catalytic performance

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Fig. 1. Characterization of CNS and 0.1% Pt/CNS: (a) SEM image of CNS without thermal treatment; (b) SEM image, (c) HRTEM image and (d) XRD pattern of 0.1% Pt/CNS after thermal treatment. The inset in c is an enlarged image from the marked part.



Fig. 2. CV curves of CNS, Pt nanoparticles and 0.1-5% Pt/CNS modified electrodes in 0.5 mol·L⁻¹ H₂SO₄ and 2 M EtOH solution.

[11,12].

2. Experimental

2.1. Synthesis of Pt/CNS nanocomposites

All the reagents were used as purchased without further treatment. The synthesis of 0.1% Pt/CNS (with 0.1 wt% Pt loading on the surface of CNS) was performed by using glucose ($C_6H_{12}O_6H_2O$) and platinum 2, 4-pentanedionate ($C_{10}H_{14}O_4Pt$) as precursors under a low-temperature hydrothermal condition. Typically, 0.50 g $C_{10}H_{14}O_4Pt$ was predissolved in 50 mL acetic acid to form a yellow clear solution and then 0.04 mL of it was mixed with 70 mL H₂O, 0.5 g glucose and 0.23 g dodecyl sodium sulfate (SDS) to form clear yellow solution. The mixed solution was transferred into a stainless steel autoclave with a Teflon liner of 100 mL capacity and heated at 180 °C for 6 h. After the autoclave was cooled down to room temperature, the floating product at the solution surface was collected and washed with EtOH and water. The obtained product was than dried in an oven at 80 °C for 12 h. The actual

Pt content in 0.1% Pt/CNS was measured by an inductively coupled plasma source mass spectrometer (Thermo Fisher Scientific, ICAP RQ) and the result was 0.46%. We guessed the deviation came from the incomplete carbonization of glucose and a measuring error. For convenient expression, the theory content of Pt (0.1%) was followed. So were 1% and 5% Pt/CNS.

To obtain 1% and 5% Pt/CNS nanocomposites, 0.4 and 2 mL preprepared acetic acid solution of Pt were added respectively while the other reaction conditions were same as those for synthesizing 0.1% Pt/CNS. To improve the conductivity of the products, the Pt/CNS nanocomposites were further carbonized at 600 °C for 2 h under the argon protection to remove the remnant CH₂ groups [10]. For comparison, CNS was synthesized under the same reaction conditions except the absence of $C_{10}H_{14}O_4Pt$. Pt nanoparticles were prepared by dropwise adding 2 mL sodium borohydride aqueous solution (1 mg·mL⁻¹) into 2 mL pre-prepared Pt acetic acid solution. The obtained Pt nanoparticles were than washed with EtOH and water, dried in the oven at 80 °C for 12 h.

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