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A novel Pt@Te-reduced graphene oxide/polyimide composite catalyst for hydrogen evolution

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ABSTRACT

A promising Pt@Te nanoparticles catalyst has been synthesized on the surface of the reduced graphene oxide/polyimide (rGO/PI) substrate. First of all, Te was deposited on the rGO/PI using electrochemical deposition method to obtain Te-rGO/PI that has various morphologies. Secondly, a monolayer Pb film was deposited on the Te-rGO/PI by the similar electrochemical method to get a transition state material of Pb@Te-rGO/PI. Finally, the target Pt@Te catalyst on the rGO/PI with large surface area was obtained by a chemical oxidation method, in which Pb was stripped off from the Pb@Te-rGO/PI by an oxygenation of H_2PtCl_6 , while the oxygenation process, Pt replaced Pb to be deposited on the surface of Te-rGO/PI to form Pt@Te-rGO/PI. Then the prepared Pt@Te-rGO/PI was used as a working electrode to study the hydrogen evolution behavior in 0.5 mol/l H_2SO_4 . By comparison the catalyst of sole Pt-rGO/PI nanoparticles, Pt@Te-rGO/PI catalyst considerably abated the demanding of Pt. And it shows excellent HER activities with a low overpotential of 0.1 V. The Tafel slope of 55 mV dec^{-1} reveals most likely the Volmer–Heyrovsky mechanism for hydrogen evolution reaction (HER). On the other hand, the catalyst displays satisfactory stability. This work may pave a potential pathway for synthesizing multi-morphology nano-Pt catalyst with high surface area for application in the study of HER.

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Introduction

Collection of hydrogen from aqueous solution by HER and use hydrogen as a new energy-generating matter is a promising means for sustainable energy applications [1–3]. For raising the efficiency of HER, catalyst is indispensable. Up to now, one of the most active catalyst toward HER is known as Pt [4,5]. However, the supply of Pt is limited because its storage in the earth is lower and its cost is very high [6]. To

overcome this problem, it's very important to prepare a Pt-based catalyst that can minimize the dosage of Pt [7,8]. So, the route exploration of dramatically decrease the usage amount of Pt in the Pt-based catalyst has been extensively conducted in recent years. For example, some works have focused on the study of core–shell or core–shell like (including dendrites, particles-on-particles, raspberries and flowers) nano-structures, in which an ultra-thin platinum shell is deposited on a non-noble/noble metal nano-particle core [9–13].

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As a result, the catalytic activity of HER catalyst is strongly dependent on its shape, size, distribution and structure [14]. In addition to the catalyst, a suitable catalyst support is also necessary that can control the morphology of the final structure because the newly formed catalyst atoms can be deposited on the surface of the support [15]. Therefore, research efforts have focused on nanoscale Pt domains with large surface area, fast charge transfer and high conductivity support. In this case, one of the most frequently used conductive support/substrates is reduced graphene (rGO) [16–19]. On the other hand, aromatic polyimide (PI) is well known high performance polymeric materials owing to their outstanding properties such as slim, light, incorrodible, high stability and flexible, which made it easily be processed and recycled as a satisfactory support. However, pure PI has limitations in more wide applications for its insulating nature [20–26]. Therefore, rGO based PI composite membrane has been developed to improve the electrical conductivity. The rGO/PI composite membrane has been successfully applied by our group for developing new sensors and photoelectrocatalysis materials [27,28].

As a p-type semiconductor, Te has been applied in electric/optical devices, thermoelectric devices, gas sensor and so on [29]. One of the most attractive properties is that Te crystals have numerous structures such as one-dimensional (1D) plane structure, 2D scroll-like structure and 3D superstructures. The different structure of the Te shows different surface morphology, surface area and catalytic activity. The mostly used method for preparation Te with various structures is electrochemical-deposition method [30–32].

In this work, variety morphologies of novel 3-D Te such as nano-sphere, nano-flower and nano-sheet with large surface area on the rGO/PI was prepared using electrochemical method under alkaline conditions. The aim of preparation Te-rGO/PI firstly is to induce obtained target Pt@Te catalyst on rGO/PI still keeping satisfactory morphology with large surface and minimize the dosage of Pt. Then, ultra-thin layer Pt was successfully prepared on Te-rGO/PI to form target Pt@Te catalyst on rGO/PI, as the final state of Pt@Te-rGO/PI, by using a chemical replacement method. In this case, a monolayer Pb was electrodeposited on the Te-rGO/PI to form transitional product of Pb@Te-rGO/PI using underpotential deposition (UPD) method. UPD is a surface-limited phenomenon, in which the electrodeposition of one metal atomic layer on a foreign metal substrate occurs at a potential that is more positive to the Nernst potential [33–35]. So we could get a morphology controlled core–shell structure for catalysis application via this progress [36,37]. Then the transitional product Pb@Te-rGO/PI was soaked into low concentration solutions of 0.2 mM H_2PtCl_6 to undergo oxidation reaction, in which, PtCl_6^{2-} ions in aqueous solution are reduced by Pb atoms on the surface of Pb@Te-rGO/PI electrode. That leads to spontaneous dissolution of the Pb from the Pb@Te-rGO/PI electrode surface and to Pt replacement Pb for obtaining the catalyst of Pt@Te-rGO/PI. There were a number of other materials to be synthesized by the similar chemical replacement progress for various application [38–46]. It was found that the prepared Pt@Te-rGO/PI catalyst in this work shows perfect catalytic activity in HER investigation. The purpose for decreasing the dosage of

noble Pt in HER catalyst preparation was accomplished successfully.

Experimental section

Apparatus and reagents

All the electrochemical experiments were performed on a CHI 832 electrochemical workstation (CH Instrument, USA). A conventional three-electrode system was used, consisting of a working electrode, a Pt wire as auxiliary electrode and a saturated calomel electrode (SCE) as reference electrode. The working electrode at the stage of electrodeposition Te is rGO/PI film, at the stage of electrodeposition Pb is Te-rGO/PI film and at the stage of HER study is Pt@Te-rGO/PI film. Before use, all the working electrodes were cleaned from ultrasonication in ethanol and water.

4,4'-oxydianiline (ODA), 4,4'-oxydiphthalic anhydride (ODPA) and N-methyl pyrrolidone (NMP) were purchased from Chinese academy of sciences, Chengdu Organic Chemistry Co., Ltd. (Chengdu, China). NMP was re-distilled to remove the water. ODA, ODPA and NMP were chemical pure and all the other chemicals were of analytical grade. Milli-Q water was used throughout. The rGO/PI membrane which acted as the working electrode was prepared according to our previous work [28].

10 mM H_2TeO_3 solution was prepared by adding 0.3990 g TeO_2 powder in 40 ml 2.3 M H_2SO_4 solution (5.0 ml concentrated sulfuric acid solution was diluted with double-distilled water to 40.0 ml) first; and then the mixture was heated to 120 °C stirring with glass rod until the solids were dissolved; Finally, the solutions was cooled to room temperature naturally and diluted with water to 250.00 ml.

For electrodeposition Te, 5.0 ml 10 mM H_2TeO_3 was added to the electrochemical cell and the acidity of the solution was adjusted to predetermined pH (for example, pH = 11) using ammonia solution. The final volume of the cell was diluted to 10.0 ml using water. For electrodeposition Pb, different concentration $\text{Pb}(\text{NO}_3)_2$ (for example, 5 mM) was prepared by added a certain volume of 10 mM $\text{Pb}(\text{NO}_3)_2$ stock solution to 5.0 ml 0.20 M HAC + 0.20 M NaAc buffer solution and then was diluted to 10.0 ml using water.

Preparation of the working electrode

For preparation of rGO/PI, a 1:1 M ratio of ODA and ODPA were added into anhydrous NMP solvent for 24 h in the magnetic stirrer to obtain polyamic acid (PAA). Then, appropriate amount of rGO and anhydrous PAA were mixed together and stirred with an ultrasonic until a homogenous suspension of rGO/PAA was obtained. Lastly, the rGO/PAA were paved onto glass, then the slide was evaporated for 3 h at 80 °C, following heated up to 300 °C in 220 min, the rGO/PI composite membrane was thus obtained. The electrolyzer for this pellet electrode designed in our laboratory was displayed in our former work [27,28]. Before use, the electrode was measured by CV in 0.5 M Na_2SO_4 electrolyte solutions to ensure the rGO/PI electrode was purity.

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