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Pt nanoparticles modified Au dendritic nanostructures: Facile synthesis and enhanced electrocatalytic performance for methanol oxidation

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ABSTRACT

A facile and simple method is presented for the synthesis of bimetallic composites, Pt nanoparticles modified dendritic Au nanostructures (PtNPs/DGNs), in which dendritic Au was deposited on a glassy carbon electrode via a potentiostatic method and sphere-like Pt nanoparticles were decorated on Au substrates through a chemical reduction reaction. The compositions, morphologies, and structures of the PtNPs/DGNs were characterized by X-ray photoelectron spectroscopy, field emission scanning electron microscopy, and energy dispersive X-ray spectroscopy. Results indicated that bimetallic composites were successfully synthesized and spherical Pt nanoparticles were dispersed evenly on dendritic Au substrates. The number of Pt nanoparticles on Au surface was regulated by controlling the chemical reduction deposition time, allowing the electrocatalytic properties of the composite towards methanol oxidation to be tuned. Electrochemical measurements, including cyclic voltammetry and chronoamperometry, were performed to investigate the electrochemical properties and electrocatalytic behaviors of the PtNPs/DGNs towards methanol oxidation. Pt nanoparticles partially covered dendritic Au exhibited dramatically enhanced electrocatalytic activity (3.947 mA cm^{-2}), which was 2.65 times that of commercial carbon-supported Pt nanoparticles (1.487 mA cm^{-2}), along with much improved

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poisoning tolerance (current decline: 70.85% vs 99.36%). These enhanced performances were likely caused by the large active electrochemical area of the bimetallic nanocomposites and the change in the electronic structure of Pt when the Au surface was modified with fewer Pt nanoparticles.

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Introduction

Direct methanol fuel cells (DMFCs) are promising new energy resources being widely investigated and developed because of their simple structures, ease of operation, and high energy densities [1–3]. Pt is currently used as a suitable electrocatalyst because of its excellent electrocatalytic properties towards methanol oxidation. However, several core limitations, including the poisoning effects of CO-like intermediates and the high costs of Pt, have not been satisfactorily resolved, restricting the commercial applications of DMFCs [2,4,5]. Therefore, the exploration of new electrocatalysts with promising catalytic performances and lower costs is needed.

The introduction of additional (second or third) metals such as Ru, Ag, Pd, or Ni can both drastically reduce the required Pt loading and modify the electronic structure of the Pt surface through interactions of the atomic orbitals of Pt and the added metal(s) [6–11]. Moreover, an added metal can improve the concentration of surface-active –OH through the effective adsorption of H₂O, increasing interactions between active –OH and CO-like species [9,12–16] and further enhancing a catalyst's durability and resistance to poisoning. Therefore, the study of new Pt-based bimetallic electrocatalysts is very important for the development of improved DMFCs. Until now, many Pt-based bimetallic materials, including PtRu [17], PtSn [18,19], PtPd [2,5], PtCu [15,20], PtNi [21,22], and PtFe [23,24], with synergistically enhanced performances have been studied. PtAu bimetallic materials have been only limitedly investigated. Recent studies have demonstrated that the addition of Au to a Pt catalyst can effectively prevent poisoning from CO-like intermediates through the tuning of Pt's electronic structure [8,16,17]. The electrocatalytic performance and utilization efficiency of a Pt catalyst are enhanced by the introduction of Au, because the added Au greatly increases the electrochemically active surface area. In addition, Au has a high conductivity and stability, which increase the stability of the catalyst [17,25,26]. Therefore, binary PtAu nanocomposites are potential new materials for electrocatalysts in fuel cell anodes, and it is especially important to determine which PtAu bimetallic structures have optimum synergistic effects for the electrocatalytic oxidation of methanol through further investigation.

In our previous study, Au dendritic nanostructures were directly deposited on glassy carbon (GC) electrodes via a potentiostatic method. These materials had good electrocatalytic activities for glucose oxidation because of the large number of active sites provided by dendritic Au nanostructures [27]. Based on this study, Pt nanoparticles modified dendritic Au nanostructures (PtNPs/DGNs) were further designed and successfully fabricated using a simple two-step

deposition method. The PtNPs/DGNs exhibited much enhanced electrocatalytic performance toward glucose oxidation compared with pure Au dendritic nanoparticles; however, the detailed mechanism for the catalytic oxidation of glucose remained unknown [28]. By combining the high surface areas of Au nanostructures with the excellent catalytic properties of Pt nanoparticles, the PtNPs/DGNs bimetallic nanocomposites might work also as a superior electrocatalyst toward methanol oxidation for fuel cell applications. Moreover, it is meaningful to take PtNPs/DGNs as a typical model of bimetallic nanoparticles for investigating the synergetic effects of bimetal on the catalytic oxidation of small molecules such as glucose and methanol.

In this work, PtNPs/DGNs bimetallic nanocomposites were prepared using a previously reported two-step method, in which dendritic Au was deposited on a GC electrode using a potentiostatic method, and Pt nanoparticles were decorated on dendritic Au substrates via a chemical reduction reaction using ascorbic acid (AA) as soft reductant [28]. The microstructures and compositions of PtNPs/DGNs prepared with different Pt deposition time from 20 min to 120 min were characterized using field-emission scanning electron microscopy (FE-SEM) and X-ray photoelectron spectroscopy (XPS). The catalytic properties and CO-poison resistance of PtNPs/DGNs for methanol oxidation were systematically investigated using electrochemical tests (cyclic voltammetry and chronoamperometry). In addition, the time for the chemical reduction-based deposition of Pt was optimized to obtain the best electrocatalytic performance for methanol oxidation. Possible sources of improved electrocatalytic properties are discussed based on the results of SEM, XPS and electrochemical measurements.

Experimental

Chemicals

Potassium chloroplatinite (II) (K₂PtCl₄), chloroauric acid tetrahydrate (AuCl₃·HCl·4H₂O), AA, concentrated sulfuric acid (H₂SO₄), N, N-Dimethylformamide (DMF), and sodium hydroxide (NaOH) were purchased from Sinopharm Chemical Regent Co., Ltd. Commercial Pt/C was purchased from Alfa Aesar. Rod-shaped GC electrodes (diameter = 3.0 mm) were obtained from Gaoss Union Instrument Company. All electrodes were polished to a mirror finish with 0.3 μm and 0.05 μm alumina slurries and rinsed sequentially with purified water, ethanol, and purified water. After being washed, the electrodes were dried with nitrogen gas at room temperature. Water was purified for all experiments with a Kertone ultrapure water system (p60-CY, Kertone Water Treatment Co., Ltd; resistivity > 18 MΩ cm).

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