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Nicotinamide-assisted fabrication of high-stability gold-palladium nanoparticles on carbon fiber cloth for hydrogen peroxide electroreduction

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ABSTRACT

Bimetallic nanoparticles with excellent activity and stability have been widely studied as H_2O_2 electroreduction catalysts. Hereby, we have successfully found a green fabrication of high-stability Au-Pd nanoparticles directly supported on carbon fiber cloth (Au-Pd NPs/CFC) by using tobacco extract (nicotinamide) as a simple and ecofriendly reducing agent. Transmission electron microscopy, scanning electron microscopy, X-ray diffraction analyses and high angle annular dark field scanning TEM are performed to characterize the morphology, composition and structure of the as-prepared electrode. Results show that alloy nanoparticles prepared with different mole ratios of $HAuCl_4/PdCl_2$ mixtures in the wide range of 10–100 nm have a uniform distribution on every carbon fiber, presenting a regular circle shape. The 3D electrode is directly applied as the electrocatalyst for H_2O_2 reduction in acid solution and does not introduce into any polymer binders. The catalytic performance is investigated by chronoamperometry and voltammetry, exhibiting desired catalytic performance and improved utilization than other representative nanomaterials recently reported. Furthermore, Au-Pd NPs do not aggregate after the whole test due to the effect of in-situ immobilization, showing a great significance for the application in fuel cells. A direct peroxide-peroxide fuel cell (DPPFC) using Au-Pd NPs/CFC cathode with Ni/Ni-foam anode acquires a maximum power density of 22.8 mW cm^{-2}. Electrochemical stability test through long-term potential cycles (> 9 h) further confirms the high durability of the electrode.
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1. Introduction

Due to the ever depletion of fossil fuels and worsening environmental situation, fuel cells have attracted great scientific interest for their high efficiency and low pollution. Oxygen from ambient air is often applied as an oxidant in fuel cell devices, however, hydrogen peroxide (H_2O_2) have replaced oxygen as the oxidizer of some liquid-based fuel cells for using in some air-free environments (underwater or outer space), because of its higher energy density, higher cell potential, easier storage, faster reduction kinetics and manufacture. The performance of H_2O_2 reduction has great influence on the performance of a H_2O_2 based fuel cells. Generally, the catalysts for H_2O_2 reduction include noble metals, transition metal oxides and enzymes. Among them, Pd as the representative non-Pt noble metal is generally considered to be the promising catalyst because it possesses similar properties to Pt, while it has high abundance, leading to a cheap price [1]. Recently, bimetallic catalysts because of the unique magnetic, optical and catalytic properties often differ remarkably from the corresponding monometallic ones. Literature [1–4] have demonstrated that bimetallic catalysts, such as Au-Pt, Au-Pd and Pd-Pt, exhibit higher catalytic activity than any single one, resulting from the synergistic catalytic effect. Furthermore, the incorporation of Au can obviously improve the stability, and also modify the adsorption behavior of surface. Au-Pd has been studied intensively as one of the most promising catalysts because of its superior activity in a number of reactions, such as the direct synthesis of H_2O_2 from O_2 and H_2O [5], the complete oxidation of toluene/benzene [6–8], oxygen reduction reaction [9] and so on [10,11]. Although noble metals demonstrated a better overall performance owing to their high catalytic activity and superior stability, but the scarcity of noble

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metals make their large-scale applications commercially inviable. Increasing the efficiency of noble metals has attracted much attention in recent researches. Currently, the main method is to directly synthesis noble metals on a three-dimensional support material with proper structure to overcome the low catalyst utilization of traditional slurry coating method, and a wide variety of materials have been tested, such as Ni foam, carbon paper and carbon fiber cloth (CFC). Moreover, it can also avoid the increase of the internal resistance and the blockage of catalyst nano-pores. In terms of its large surface area and high mechanical variability, CFC has been extensively employed as an electrode substrate to fabricate electrochemical devices supports.

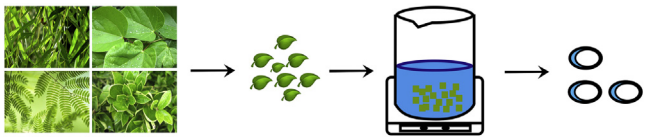
More importantly, besides reducing the noble metals loading, the conventional techniques are complex, cost-intensive and often involve multiple steps. The development of cost-effective methods to minimize the catalysts loading and simultaneously to improve the catalytic performance as well as stability under realistic conditions is an important aspect of fundamental research in this area. Recently, plant mediated biological synthesis of nanoparticles is gaining importance, which is not only an easy, flexible, low-cost and environmentally friendly synthesis method without using high pressure, energy or toxic chemicals, but also nanoparticles prepared with plant extract usually have a well-defined size and morphology than some of the physicochemical methods of production [7,12–16]. Generally, plants can serve both as stabilizing agents and reducing agents, because that its strong ability from different phytochemicals can reduce metal ions from their higher oxidation to zero oxidation state [17,18]. Plants like Ficus sycomorus [19], Solanum nigrum [20], Cinnamomum tamala [21], Cacumen Platycladi [22] have been applied to the fabrication of Ag, Au and Pd nanoparticles. To date, only a few attempts to fabricate biosynthesized catalysts on three-dimensional substrate have been documented.

Hereby, we describe a novel route for the fabrication of Au-Pd alloy nanoparticles (Au-Pd NPs) loaded on CFC by using the tobacco extract (nicotinamide) as a reducing agent. It is worth to note that the green biosynthetic method by natural tobacco extract. Briefly, CFC substrate can be first formed functionalized surfaces by the decoration of polymer molecule, and then metal precursors were self-assembled through electrostatic interaction, finally the metal precursors can be reduced to metal particles using plant extracts. Such obtained Au-Pd alloy nanopaticles exhibits a very high and stable electrocatalytic performance towards the H_2O_2 reduction in acidic solutions. This study provides a general method to fabricate metallic NPs supported on 3D carbon substrate, which can be useful for biosynthesized catalysts in a broad variety of applications.

2. Experimental

2.1. Reagents

Carbon fiber cloth (thickness: 0.3 mm) was bought from Shanghai Hesen electric Co., Ltd. Chloroauric acid ($HAuCl_4$), palladium chloride ($PdCl_2$), 1,6-hexanediamine ($C_6H_{16}N_2$, 99.0%), sulfuric acid (H_2SO_4), nitric acid (HNO_3), hydrochloric acid (HCl), potassium chloride (KCl), hydrogen peroxide (H_2O_2), sodium hydroxide (NaOH), potassium hydroxide (KOH) were purchased from Enterprise Group Chemicals Reagent Co., Ltd. China. All chemicals are analytical grade and were used as-received without further purification. Ultrapure water (Millipore, 18 MΩ cm) was used throughout the study. The natural tobacco named as Longjiang 237 (Product Grade: Qualified) was obtained from a local market in Harbin, China and thoroughly washed with distilled water before use. Nicotinamide was extracted from natural tobacco by the method of steam distillation (Scheme 1).



Scheme 1. The schematic diagram of preparation progress of tobacco extract.

2.2. Preparation of tobacco extract

The preparation progress of tobacco extract is as following: Firstly, 30 g of dried powder of tobacco was refluxed with 10% HCl for 30∼40 min. Secondly, the reaction mixture needed cooling in the beaker, and then added 40% NaOH solution into alkaline. Finally, the mixtures was extracted by boiling in double distilled water and the supernatant as the resulting extract can be acquired form the aqueous extract through centrifugation progress.

2.3. Preparation and characterization of Au-Pd NPs/CFC electrode

The electrode was prepared by the biosynthesis method, and the schematic diagram of preparation progress is shown in Fig. 1. The surface of CFC substrate became more active after pretreatment with concentrated H_2SO_4-HNO_3 (volume ratio = 3:1), benefitting for deposition of alloy particles. Subsequently, the substrate was rinsed with water repeatedly to neutrality and then immersed into the solution of protonated 1,6-hexanediamine (HDA) for 4 h to prepare adsorbed HDA monolayers on CFC. The resulting HDA modified CFC was immersed in an aqueous solution containing $HAuCl_4/K_2PdCl_4$ mixtures in molar ratios of 3:1, 1:1 and 1:3 as Au_3Pd_1, Au_1Pd_1 and Au_1Pd_3, respectively, for 4 h, in which the precursors were self-assembled through electrostatic interaction. After CFC rinsing with water several times, the bound $AuCl^{4-}$ and $PdCl_4^{2-}$ was reduced in the aqueous solution containing nicotinamide at ambient temperature for 8 h and the formation of alloy particles on CFC was achieved.

2.4. Materials Characterization

A scanning electron microscope (SEM, JEOL JSM-6480) and a transmission electron microscope (TEM, JEM-2010FEF, 200 kV) were used to characterize the morphology of alloy particles. High angle annular dark field scanning TEM (HAADF-STEM) imaging, elemental line-scanning and x-ray diffractometer (Rigaku TTR III) were examined to analyze the composition and structure. The Au and Pd loading were measured using an inductive coupled plasma emission spectrometer (ICP, Xseries II, Thermo Scientific). Au and Pd in the 1.0 cm^2 electrode were first dissolved in aqua regia solution and then diluted to 1 L solution for the ICP measurement.

2.5. Electrochemical measurements

Cyclic voltammetry (CV) and chronoamperometry (CA) experiments were carried out in a conventional three-electrode electrochemical cell using a computerized potentiostat (Autolab PGSTAT302, Eco Chemie) controlled by GPES software. CFC was applied as the working electrode, which was placed between two pieces of platinum foil (1.0 × 1.0 cm) in parallel as the counter electrodes. A saturated Ag/AgCl (KCl sat.) electrode was used as the reference electrode, and all potentials in this work were referred to this reference electrode. The electrocatalytic experiments were conducted on the as-prepared electrode in H_2SO_4 solution containing H_2O_2. All measurements were performed at ambient temperature under N_2 atmosphere.

Direct peroxide-peroxide fuel cell (DPPFC) was prepared using Au-Pd/CFC cathode and Ni/Ni-foam anode. Ni/Ni-foam anode was

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