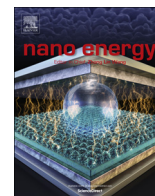




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Controlled synthesis of Se-supported Au/Pd nanoparticles with photo-assisted electrocatalytic activity and their application in self-powered sensing systems



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ABSTRACT

Bimetallic nanoparticles (NPs) have received increasing attention for their outstanding catalytic activity which the corresponding monometallic NPs can hardly achieve. In this paper, we developed a facile approach to prepare Se-supported Au/Pd NPs with high and photo-assisted electrocatalytic activity. Cyclic voltammetry (CV) measurements showed that the electrocatalytic activity of the Se-supported Au/Pd NPs was dependent on the Pd content. This result indicates that the interface between Pd and Au plays a vital factor in the electrocatalytic activity, while the individual metal components in the Se/Au/Pd system are of minor relevance for such activity. The Se-supported Au/Pd NPs exhibited mass activities of 4.25 (A/mg_{Pd}) and 1.21 (A/mg_{Au+Pd}) toward ethanol oxidation, which were higher than that of the commercial Pd/C catalyst (0.36 A/mg_{Pd}). The electrocatalytic activity of Se-supported Au/Pd NPs was further enhanced by 2.4 times under solar light irradiation due to the photoelectrochemical effect of the Se NPs. It was further demonstrated in this paper that the presence of Se-supported Au/Pd NPs is crucial in self-powered electrochemical sensing systems as the overall sensitivity is significantly improved.

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1. Introduction

Nanoparticles (NPs) with core/shell or alloy structures have received wide attention owing to their enhanced catalytic activities relative to those of their corresponding monometallic NPs [1]. A representative example is Au/Pd NPs which provided higher catalytic activity toward hydrogen peroxide synthesis than individual Pd NPs [2]. Besides chemical composition, the size and shape of the NPs are also critical factors in determining their catalytic activities [3]. For example, Pd concave cubes provided higher catalytic activity than Pd octahedra in Suzuki coupling reactions [4]. Several approaches have been demonstrated for the synthesis of bimetallic Au/Pd NPs with well-controlled shapes, sizes, chemical compositions and structures [5]. A two-step seeding growth method has been widely used to prepare bimetallic core/shell NPs with well-defined geometries, wherein a second metal layer is grown over the pre-synthesized seeds [6]. On the other hand, a galvanic replacement reaction [7] and co-reduction of two metal precursors with a suitable reducing agent [8] have been commonly employed to prepare bimetallic alloy NPs.

Various fuel cells, due to their advantage in producing smaller amounts of waste than the combustion of gas, have long been attractive as sustainable energy sources [9]. Among them, direct alcohol fuel cells (DAFCs) have been majorly applied as the power sources for portable electronic devices and fuel-cell vehicles [10]. Compared to gaseous fuels, liquid fuels have higher energy densities with lower prices and can be easily handled, stored and transported. Ethanol is one of the most advantageous liquid fuels which can exhibit lower toxicity and higher ability of large-scale production from agricultural products or biomass [11]. Owing to this fact, Pt-based NPs have been extensively investigated as the electrocatalysts for ethanol oxidation [12]. However, the development of DAFCs has been hindered by the barriers of high-priced and limited supply of Pt. To overcome the economic disadvantages of these electrocatalysts, many strategies have been proposed for the preparation of affordable alternative catalytic structures for DAFC anodes which can enable the oxidation of primary and secondary alcohols with fast kinetics and tolerable deactivations [13]. Pd is emerging as a promising replacement for Pt in DAFCs [14]. Pd NPs are able to provide a higher electrocatalytic activity and faster reaction kinetics than Pt toward ethanol oxidation in alkaline media [15]. Although a number of efforts have been devoted to the morphology control of Pd [16], Pd remains

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disadvantageous in its instability and cost-ineffectiveness as an anodic catalyst.

To further improve the durability and lower the cost of the anodic catalysts used in DAFCs, Pd-containing bimetallic NPs have been developed [17]. Among various bimetallic NPs used in DAFCs, Au/Pd NPs are the most fascinating due to their excellent catalytic activities and stabilities [18]. The incorporation of Au into Pd catalysts could enhance the catalytic activity and selectivity of Pd as well as the resistance to poisoning, mainly because CO-like intermediate species could be oxidized on the Au surface to produce CO₂ or other products, releasing the active sites of Pd [19]. Several approaches have been proposed for the preparation of various Au/Pd NP structures [20,21]. The one-pot synthesis of Au/Pd octapodal NPs was achieved by co-reduction of Au and Pd precursors with L-ascorbic acid in the presence of cetyltrimethylammonium chloride [22]. The corresponding mass activity of the octapodal NPs was 0.92 A/mg_{Pd}, which is approximately 2 and 4 times higher than that of the flower-like Pd NPs (0.49 A/mg_{Pd}) and the Pd/C catalyst (0.24 A/mg_{Pd}), respectively. Alternatively, a one-step electrodeposition route was applied to the synthesis of unsupported Au/Pd bimetallic heterostructure tubes with 50–100% Pd on an anodic aluminum oxide template [23]. When the atom percentage of Pd was up to 65%, the mass activity of as-prepared Au/Pd NPs increased to more than 2.0 A/mg_{Pd} and 1.0 A/mg_(Pd+Au), which was 2 times greater than the mass activity of the Pt/C catalysts.

Herein, we used Se NPs as the templates and reducing agents for the growth of Au and Pd atoms. For efficient utilization of Pd, Au NPs were grown first, which then acted as seeds for the growth of the Pd NPs. The Au and Pd contents on the as-prepared Se/Au/Pd NPs were simply controlled by varying the concentrations of the AuCl₄⁻ and [PdCl₄]²⁻ ions. Cyclic voltammetry (CV) results revealed that the electrocatalytic oxidation of ethanol using the as-prepared Se/Au/Pd NPs was dependent on the Pd content. The Se/Au/Pd NPs provided higher electrocatalytic activity than Se/Au NPs, Se/Pd NPs and a commercial Pd/C catalyst. Under solar light irradiation, we observed the photo-assisted electrocatalytic property of the as-prepared Se/Au/Pd NPs, mainly because of the noticeable photoconductivity (8×10^4 S/cm) provided by the Se NPs [24].

We also demonstrated that the as-prepared Se/Au/Pd NPs were critical for the development of self-powered sensing systems. Self-powered sensing systems which can function without external power have attracted increasing attention in recent years [25,26]. Some representative examples, including systems for the sensing of pH [27], light [28], temperature [29], humidity [30], catechin molecules [31] and mercury ions [32], have been successfully developed. The self-powered sensing system we designed mainly contains a triboelectric nanogenerator (TEENG) as the power source, a commercial capacitor to store the generated electricity, and an electrochemical cell for analyte detection. TEENGs, which were developed in 2012 [33], have become good candidates as the power sources for self-powered sensing systems due to its efficiency in converting mechanical energy into electricity [34–36]. Different types of TEENGs have been studied to harvest mechanical energy from various sources in the environment [37–39]. By integrating the as-prepared Se/Au/Pd NPs, the response of the self-powered sensing system toward glucose detection can be enhanced enormously and exhibit a remarkable sensitivity. To the best of our knowledge, this is the first study that uses bimetallic NPs in self-powered sensing systems and demonstrates their superiority in varied aspects.

2. Experimental section

2.1. Synthesis of Se NPs

We used a facile chemical reduction approach to synthesize size-controlled Se NPs at room temperature. The sizes of the Se NPs were controlled from 20 to 240 nm by merely adjusting the concentration of the reducing agent (sodium thiosulfate pentahydrate) from 5.2 to 52 mM. For instance, sodium thiosulfate pentahydrate (520 mM, 0.5 mL) was added to a solution of selenium dioxide (5.2 mM) containing 10 mM SDS (10 mL) at room temperature under constant magnetic stirring for the preparation of Se NPs with an average size of 130 nm. The solution color changed from colorless to orange after 6 h, indicating the formation of Se NPs. The as-prepared Se NPs were centrifuged (6000 g, 10 min) to remove excess chemicals and then the pellet was re-dispersed in a 10 mM SDS solution (9 mL) for the growth of the Au NPs.

2.2. Growth of Au NPs and Pd NPs onto the Se NPs

Through a redox reaction between Au³⁺ ions and the Se NPs, Au NPs were formed on the surfaces of the Se NPs, while SeO₃²⁻ ions were released into the bulk solution. In a typical synthesis, aliquots (0.2–0.8 mL) of aqueous sodium tetrachloroaurate solutions (final concentrations of 0.2, 0.4, 0.6 and 0.8 mM) were added to the Se NPs which had been re-dispersed in the 10 mM SDS solutions (final volume of 10 mL) for 10 min. The mixtures were maintained at ambient temperature (25 °C) for 30 min to allow the growth of the Au NPs on the Se NPs. The as-prepared NPs were represented as Se/Au NPs. We also synthesized Se/Pd NPs for comparison of the electrocatalytic analyses. Typically, aliquots (0.4 mL) of aqueous potassium tetrachloropalladate solution (final Pd²⁺ concentration of 0.4 mM) were added to the as-prepared Se NPs solutions. The mixtures were maintained at ambient temperature for 30 min to allow the growth of Pd NPs on the Se NPs. The as-prepared NPs were represented as Se/Pd NPs.

2.3. Preparation of Se/Au/Pd NPs

The as-prepared Se/Au NPs were centrifuged (6000 g, 10 min) to remove excess chemicals and then the pellet was re-dispersed in 10 mM SDS solution (9 mL). Through a redox reaction between Pd²⁺ ions and the Se NPs on the Se/Au NPs, Pd NPs were formed on the surfaces of the Au NPs, while SeO₃²⁻ ions were released into the bulk solution. In a typical synthesis, aliquots (0.2–0.8 mL) of the aqueous potassium tetrachloropalladate solution (final Pd²⁺ concentration of 0.2, 0.4 and 0.8 mM) were added to the as-prepared Se/Au NPs solutions. The mixtures were maintained at ambient temperature for 30 min to allow the growth of Pd NPs on the Se/Au NPs. The as-prepared NPs were represented as Se/Au/Pd NPs.

2.4. Fabrication of TEENG

First, polyethylene terephthalate (PET) was selected as the substrate material. Then, aluminum (thickness approximately 100 nm) was deposited on the two PET substrates using an e-beam evaporator. The dimensions of the PET substrate used in this paper were 5 cm × 5 cm. Two conducting wires were connected to the two aluminum electrodes as leads for subsequent electric measurements. The PTFE and gelatin/glycerol films were directly coated on the aluminum electrodes as triboelectric layers. Finally, the triboelectric layers were attached on a linear motor system for the system operation of TEENG.

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