



Platinum nanoparticles decorated dendrite-like gold nanostructure on glassy carbon electrodes for enhancing electrocatalysis performance to glucose oxidation



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ABSTRACT

Platinum nanoparticles decorated dendrite-like gold nanostructure, bimetal composite materials on glassy carbon electrode (Pt/DGNs/GC) for enhancing electrocatalysis to glucose oxidation was designed and successfully fabricated by a facile two-step deposition method without any templates, surfactants, or stabilizers. Dendrite-like gold nanostructure was firstly deposited on the GC electrode via the potentiostatic method, and then platinum nanoparticles were decorated on the surface of gold substrate through chemical reduction deposition. X-ray diffraction (XRD), field-emission scanning electron microscope (FE-SEM), energy-dispersive X-ray spectroscopy (EDS) were applied to characterize the evolution of morphology and structure of the as-prepared Pt/DGNs/GC. Based on electrochemical measurements such as cyclic voltammetry, linear voltammetry and chronoamperometry, Pt/DGNs/GC exhibited significantly enhanced electrocatalytic performance to glucose oxidation compared those of pure dendrite-like Au nanoparticles in our previous report. Controlling chemical reduction deposition time, the amount of platinum nanoparticles on Au surface could be regulated, which further tuned electrocatalytic properties toward glucose oxidation. The dendrite-like gold surface partially covered by platinum nanoparticles dramatically enhanced the electrocatalytic performance for the oxidation of glucose because of excellent synergetic effects between gold and platinum species and the increased electrochemical active area from Pt nanoparticles loading. The non-enzymatic glucose biosensor based on Pt/DGNs/GC showed a rapid respond time (within 2 s), wide linear range (from 0.1 mM to 14 mM), low detection limit (0.01 mM), supernal sensitivity ($275.44 \mu\text{A cm}^{-2} \text{mM}^{-1}$, $R = 0.993$), satisfactory reproducibility and good stability for glucose sensing. It was demonstrated that Pt/DGNs/GC could work as promising candidate for factual non-enzymatic glucose detection.

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1. Introduction

For the past several decades, glucose sensing had attracted considerable attentions from many researchers because of the importance application in various fields, such as the clinical diagnosis, fuel cell, ecological and food monitoring [1]. Many efforts had been devoted for developing glucose sensor with good selectivity, better stability, wonderful sensitivity and fast response. According

to whether or not using glucose oxidase (GOD), electrochemical glucose sensors, one of effective approach for sensing glucose, can be divided into enzyme based glucose sensor and non-enzymatic glucose biosensor. The enzymatic glucose sensor initiated by Clark and Lyons' pioneer workers, had drawn considerable interests since then [2]. Although the enzymatic glucose sensor exhibited satisfactory selectivity and high sensitivity on glucose detection [3–6], some technical problems, such as the sensitivity to the environment, the instability and the denaturation of immobilized enzymes were the prominent difficulties to overcome in the process of form, storage and use of electrode [7]. In addition, the complicated immobilization course of GOD, still affected the reproducibility, stability and sensitivity of the enzyme-based glucose biosensor [8].

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Nonenzymatic glucose sensor based glucose direct oxidation on the surface of electrode modified with nanomaterials might open another effective way to solve the above mentioned problems for the advantages, including lower cost, desired stability, convenient store and high sensitivity. The used electrocatalysts on the electrode were considerably vital for direct non-enzymatic electrooxidation. To date, many noble metals and oxide materials such as Pt [9–11], Pd [12,13], Au [14–17], Cu_xO [18–22] and CeO_2 [23] have been applied in glucose electrocatalytic oxidation. Among these materials, Pt nanomaterials had been focused because of their prominent electrocatalytic ability towards glucose oxidation in neutral solution [24,25]. However, the most obvious shortcoming was that the surface of the Pt nanomaterials on electrode was easily covered by intermediate products, resulting in electrocatalysts passivating or poisoning and subsequently losing electro-catalytic activity during the experimental procedure.

Gold was another attractive material due to its comparatively strong anti-toxic capabilities from intermediates in the glucose electrooxidation, which resulted from its full filled 3d orbital [16,26]. In addition, it had a lower oxidation potential and satisfactory selectivity in neutral medium compared to the other metals. The limitations of high cost and weak electro-catalytic activity toward glucose molecules compared to Pt nanoparticles in neutral buffers hinder its application in biosensor [14,27,28]. Therefore it still faced challenges to further improve electrocatalytic performance of Au based nanomaterials. Combining Pt and Au to fabricate composite materials might integrate advantages of both, thus further enhancing the electrocatalytic and anti-poison ability [1,12,29,30]. However the studies about Pt and Au composite nanomaterials in glucose oxidation have been few reported.

In this study, platinum nanoparticles decorated dendrite-like gold nanostructure on glassy carbon electrode (Pt/DGNs/GC) was designed and fabricated by facile two steps deposition, electrochemical and chemical reduction deposition. The effect of Pt nanoparticles covering on electrocatalysis of glucose oxidation was systematically investigated and compared with pure Au dendrite nanostructures in our previous report through various electrochemical measurements. It was found that decorating appropriate amount of Pt nanoparticles on Au surface could significant enhance the electrocatalytic performance for glucose oxidation, which could be due to excellent synergic effect between Au and Pt and the increased electrochemical active area. The glucose sensor based Pt/DGNs/GC showed rapid response, good sensitivity and stability towards glucose sensing, which was desirable for the factual application.

2. Experimental section

2.1. Materials

$\text{AuCl}_3 \cdot \text{HCl} \cdot 4\text{H}_2\text{O}$ ($\text{HAuCl}_4 \cdot 4\text{H}_2\text{O}$), ascorbic acid (AA), uric acid (UA), 4-acetaminophen (AP), absolute alcohol, $\text{Na}_2\text{HPO}_4 \cdot 12\text{H}_2\text{O}$, $\text{NaH}_2\text{PO}_4 \cdot 3\text{H}_2\text{O}$, glucose, concentrated sulfuric acid (H_2SO_4) and KCl were purchased from the Sinopharm Chemical Reagent Co. Ltd. K_2PtCl_4 was obtained from Aldrich Chem. Phosphate buffer solution (0.1 M PBS pH 7.4) was prepared from $\text{Na}_2\text{HPO}_4 \cdot 12\text{H}_2\text{O}$ and $\text{NaH}_2\text{PO}_4 \cdot 3\text{H}_2\text{O}$. Solutions of glucose, AA, UA, and AP were prepared by using PBS throughout the experiment. By using the slurries of 0.3 and 0.05 μm alumina, rod-shaped glassy carbon electrodes (GCE, 3 mm in diameter) purchased from Gaoss Union Instrument Company (Wuhan, China) were polished and sequentially washed with ethanol and pure water (p60-CY, Kertone Water Treatment Co. Ltd, resistivity > 18 $\text{M}\Omega \text{ cm}$). Then the electrodes were dried with nitrogen gas. All the chemicals were of analytical grade and used without further purification.

2.2. Apparatuses

Electrochemical measurements were accomplished with a 550 electrochemical work station (Gaoss Union Instrument Company, Wuhan, China) in a conventional three-electrode cell. The modified glassy carbon electrode was used as the working electrode, Pt wire acted as counter electrode, and Ag/AgCl electrode was applied as the reference electrode. Before each experiment, pure N_2 gas was bubbled through the solution to remove any dissolved oxygen. Structure and morphology were characterized by field-emission scanning electron microscopy (FE-SEM, ZEISS, Germany). EDS analysis of Pt/DGNs/GC was performed on X-Max (Oxford Instruments, UK) coupled with the field-emission scanning electron microscopy (FE-SEM, JEOL, JSM7100F, Japan). X-ray diffraction (XRD) characterization was carried out by a D8-advance diffractometer (Bruker, Germany) with a $\text{Cu K}\alpha$ radiation source ($\lambda = 0.15418 \text{ nm}$).

2.3. Synthesis of dendrite-like gold nanoparticles modified GC electrode

The preparation of dendrite-like gold nanoparticles modified glassy carbon electrode (DGNs/GC) was similar to the method reported in our previous study [14]. Firstly, the precursor solution containing 0.8 mL of 0.1 M $\text{HAuCl}_4 \cdot 4\text{H}_2\text{O}$ and 7.2 mL of 0.1 M KCl was prepared in an electrolysis bath. Then, electrodeposition was performed with a potentiostatic method (-0.3 V , 3600 s). Gold nanoparticles modified GC electrode was rinsed several times with deionized water and dried in room temperature.

2.4. Fabrication of Pt/DGNs/GC electrode

Briefly, the precursor solution of Pt was prepared by adding 1 mL of 20 mM K_2PtCl_4 solution and 1 mL of 100 mM ascorbic acid (AA) solution into 18 mL pure water. Then gold nanoparticles modified GC electrode was transferred into the precursor solution of Pt and left for different depositing time, 5, 10, 30 min, respectively. The prepared GCEs electrodes were rinsed with pure water several times and dried for further characterization. As control experiments, Pt nanoparticles modified GC (PtNPs/GC) was obtained by only immersing glassy carbon electrode into the Pt precursor solution for 10 min.

3. Results and discussion

3.1. Characterizations of structure and morphology of Pt/DGNs/GC, PtNPs/GC and DGNs/GC

As presented in the next electrochemical testing results, Pt/DGNs/GC fabricated under the condition that Pt nanoparticles were decorated on gold dendrite substrate for 10 min (Pt_(10min)/DGNs/GC) exhibits the best electrocatalytic performance toward glucose oxidation. Therefore, we mainly focused the following investigation to this Pt/DGNs/GC in comparison with PtNPs/GC and DGNs/GC in morphology and structure characterizations.

Fig. 1 showed the X-ray diffraction patterns of PtNPs/GC, DGNs/GC and Pt/DGNs/GC. The pattern of DGNs/GC showed the major peaks of Au at 38.2° (111), 44.4° (200), 64.6° (220), and 77.5° (311) which were in good agreement with the patterns of Au nanoparticles (DGNs/GC) as fcc Au (JCPDS 04-0784) [31,32]. After chemically depositing Pt nanoparticles, the main peaks corresponding Pt (fcc) crystal structure at 39.72° , 46.3° , 67.36° appeared in the pattern of Pt/DGNs/GC, which were accordance with those of PtNPs/GC. It indicated Pt nanoparticles were successfully modified on the surface of Au. The weak intensity and widening of the peaks could contribute to small amount and size of Pt nanoparticles.

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