

High-power non-enzymatic glucose biofuel cells based on three-dimensional platinum nanoclusters immobilized on multiwalled carbon nanotubes



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ABSTRACT

Non-enzymatic glucose biofuel cells (GBFCs) has been renewed interest because of good long-term stability and adequate power density. Here we demonstrate the application of a three-dimensional (3D) nanocomposites electrode for implantable GBFCs with simple fabrication protocol, good performance (a high power density 2.3 mW cm^{-2} and an open circuit potential 0.70 V in physiological environment) and excellent stability. 3D flowerlike platinum (Pt) nanoparticle clusters are electrodeposited onto multiwalled carbon nanotubes (MWCNTs) by using an all-electrochemical protocol, which involves a key, second step of a potential pulse sequence. The potential widths can change the size and distance of the nuclei and clusters. The resulting 3D Pt morphology of a new type is found to exhibit significantly higher electrocatalytic activity and better stability than the dispersive morphology for glucose oxidation reaction (GOR) and oxygen reduction reaction (ORR). We also investigate the application (polarization test, biofuel cell performance and degradation behavior) of this process for the fabrication of both anode and cathode in GBFCs. This new procedure might give credence for construction of a new generation of GBFCs operating at mild conditions or boost the power outputs and make them suitable for diverse applications.

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1. Introduction

The field of bioelectrocatalysis has flourished recently because of the growing interest in producing electrical energy from biologically renewable resources. Glucose biofuel cells (GBFCs) are particularly interesting nowadays due to continuous harvesting of oxygen and glucose from bioavailable substrates, activity inside the human body, and environmental benign [1], which generate electricity through oxidation of glucose on the anode and reduction of oxygen on the cathode [2]. Generally, two kinds of catalysts are used in GBFCs: biological and non-biological [3]. In the past few decades the biological GBFCs based enzymes have been intensively investigated [4–6].

Enzymatic catalysts for glucose/dioxygen (O_2) biofuel cells have excellent selectivity and reaction rates, however their infection potential and limited stability (due to the fragile nature of the enzymes and poor immobilization techniques) are critical disadvantages [7]. Furthermore, most of the GBFCs are reported at saturating conditions of glucose and O_2 [8–10]. Nevertheless, for *in vivo* implantation, the concentrations of glucose and O_2 in the physiological fluids are remarkably lower than those corresponding to the saturating conditions of glucose and O_2 in buffer [11].

Hence, there has been renewed interest in non-enzymatic GBFCs that use noble metals or/and carbon-support as catalysts [12]. Such biofuel cells have shown good time stability and have been successfully tested *in-vivo* [13]. The noble metals such as platinum (Pt) are promising candidates because of their long-term stability and biocompatibility [14]. However, slow reaction kinetics of glucose oxidation and catalyst poisoning are critical obstacles to the realization of noble metal based GBFCs [15]. Thus many efforts have been spent on both catalyst dispersion and carbon-support design in order to enhance the catalytic activity.

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As a new form of carbon, carbon nanotubes (CNTs) have been the subject of numerous theoretical and experimental studies [16]. Shortly after their discovery it was realized that the large accessible surface areas, unique electronic properties, antifouling ability, and high stability make them ideal candidates for electrodes and catalyst supports [17,18]. Metal/CNTs nanomaterials, possessing the advanced properties of each component with a synergistic effect, have attracted more interest in reaction promotion and process stabilization [9,15]. In recent years, high surface area Pt and CNTs nanocomposites have been successfully prepared via several routes [19–21]. However, the surface density of Pt nanoparticles is limited by the surface density of the broken CNTs bonding, which deteriorates the electrical conductivity and mechanical strength [22].

Considering the advantages of electrodeposition, such as high purity of deposits and mild procedure [23,24], we suggest a new approach based on a full-electrochemical three-step process to deposit three-dimensional (3D) flowerlike Pt nanoparticle clusters on multiwalled carbon nanotubes (MWCNTs). The key, second step is a potential pulse sequence, which can produce the unique 3D morphology, different from the dispersion morphology which is achieved when the second step adopts cyclic voltammetry instead of a potential-step method. The 3D Pt morphology of a new type is found to exhibit significantly higher electrocatalytic activity and better stability than the dispersive morphology for glucose oxidation reaction (GOR) and oxygen reduction reaction (ORR). The relationships between potential pulse width for the size of the nuclei/nanoclusters and performance of the resulting electrodes are analyzed to obtain an optimum electrodeposition conditions. Herein we also demonstrate the application of the 3D Pt/MWCNTs electrodes for implantable GBFCs. The power density, open circuit potential (OCP), short circuit current (SCC), electrode polarization, and degradation behavior are tested in half-cell configuration under various operation conditions relevant for GBFCs (phosphate buffered saline pH 5.0–7.5, 0–1 M glucose, 0.0%–28.0% oxygen saturation). This non-enzymatic electrode is beneficial due to shorter fabrication times, high surface density, enhanced electrocatalytic activity and good stability for implantable GBFCs.

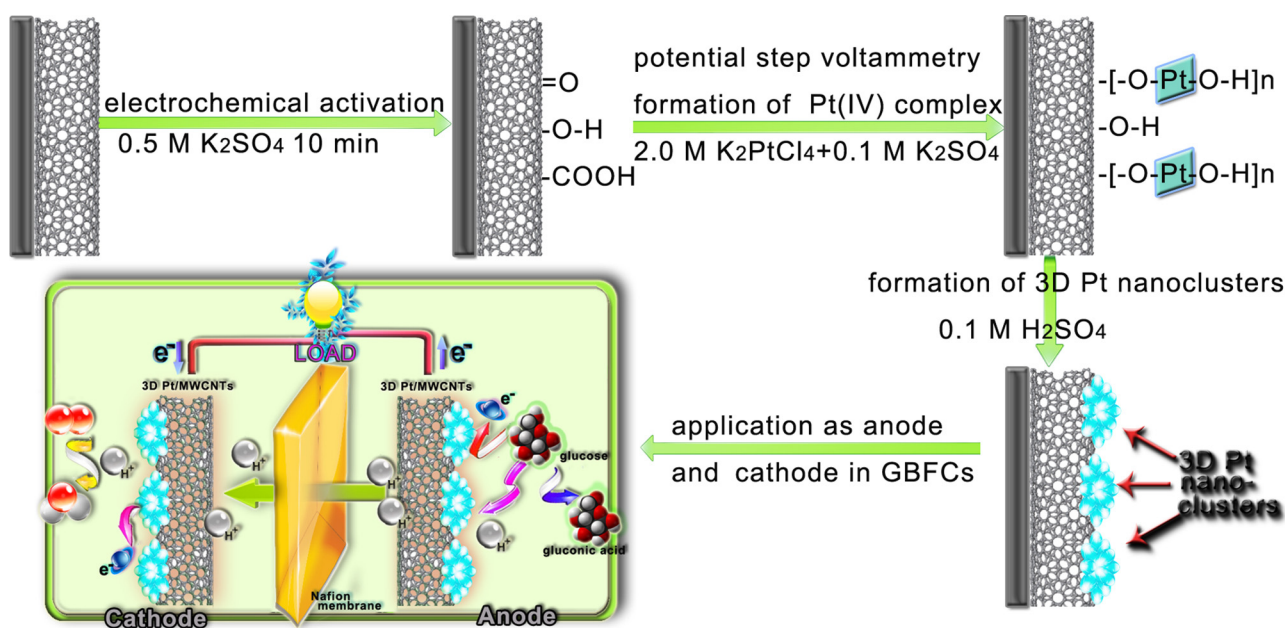
2. Experimental

2.1. Materials

MWCNTs (~90% purity on carbon basis, size 7–15 nm, 0.5–10 μm) were purchased from Shenzhen Nanotech. Port. Co., Ltd (Shenzhen, China) and further purified prior to use by stirring in concentrated nitric acid (HNO_3) for twelve hours. $\alpha\text{-D}$ (+)-glucose monohydrate was purchased from Sigma-Aldrich. Nafion 115 membrane (127 μm , 0.90 mequiv./g, DuPont Co., USA) was purchased from Shanghai River's electric Co., LTD (Shanghai, China) and boiled in 3% hydrogen peroxide and then in alternating baths of boiling water and 1 M sulfuric acid (H_2SO_4). Toluene ($\text{C}_6\text{H}_5\text{CH}_3$) (Park Co. Dublin, Ireland) was dried with sodium (Na) and refluxed for six hours before distillation and then stored in the presence of Na. Deionized water (H_2O) ($20 \pm 1^\circ\text{C}$, pH=7, $\rho=18.3 \text{ M}\Omega \text{ cm}$) was purified by ultra pure water system (Barnstead Co., USA). Other reagents include potassium sulfate (K_2SO_4) (AR, Nacalai Tesque, Inc. Kyoto, Japan), potassium tetrachloroplatinate(II) (K_2PtCl_4) (99.9% Pt, Aldrich), H_2SO_4 (AR, BeiHua-Inc., Beijing, China), and phosphate buffer solution (PBS) (AR, Tianjin Chemical Reagent Third Factory, Tianjin, China).

2.2. Electrode Preparation

The glassy carbon (GC) electrode (3 mm diameter) was polished to a mirror finish with emery paper and alumina (Al_2O_3) slurry (1.0 μm , 0.3 μm), and ultrasonically cleaned in toluene for a few minutes, then dried with a high-purity nitrogen (N_2) stream. A suspension of 0.1 mg mL^{-1} was prepared by dispersing 1 mg of purified MWCNTs in 10 mL toluene by ultrasonication for 30 min. To prepare a MWCNTs film electrode, 15 μL of the suspension was directly cast on the GC electrode surface, followed by solvent evaporation at room temperature.



Scheme 1. Schematic illustration of three-step process for electrochemical synthesis of the flowerlike 3D Pt/MWCNTs catalysts, and the GBFCs equipped with 3D Pt/MWCNTs electrodes in the enlarged view.

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