



Short communication

A novel non-enzymatic glucose sensor based on Pt₃Ru₁ alloy nanoparticles with high density of surface defectsJiangwei Yang^a, Xinyi Liang^{a,*}, Lan Cui^b, Haiyan Liu^a, Junbo Xie^a, Weixing Liu^a^a Tianjin Key Laboratory of Food and Biotechnology, Department of Applied Chemistry, College of Science, Tianjin University of Commerce, Tianjin 300134, PR China^b College of Materials Science and Engineering, Tianjin University, Tianjin 300072, PR China

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ABSTRACT

A novel non-enzymatic glucose sensor based on a glassy carbon electrode modified with Pt₃Ru₁ alloy nanoparticles (Pt₃Ru₁/GCE) was fabricated. Pt₃Ru₁ alloy nanoparticles were prepared by a reverse microemulsion method at room temperature. The X-ray diffraction (XRD) and energy dispersive X-ray spectroscopy (EDS) demonstrate that Pt₃Ru₁ nanoparticles are disordered alloy with face central cubic (fcc) structure and the atom ratio of Pt and Ru is 3:1. The high-resolution transmission electron microscopy (HRTEM) images show that Pt₃Ru₁ alloy nanoparticles were aggregated with a high density of surface defects. Furthermore, the sensor properties of Pt₃Ru₁/GCE were investigated by cyclic voltammetry (CV), electrochemical impedance spectroscopy (EIS) and chronoamperometry (CA) in 0.01 M PBS (pH 7.4). The results indicate that the proposed sensor exhibits a wide linear range of 5×10^{-7} M to 10^{-2} M ($R^2 = 0.9988$) with a low detection limit of 0.3 μ M for glucose. Moreover, the sensor demonstrates good sensitivity, stability, reproducibility, and better anti-interference performance toward ascorbic acid (AA), uric acid (UA), and fructose (Fru).

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1. Introduction

Accurate detection and quantitative analysis of glucose are of great importance in clinical diagnoses, biological medicine, food processing and other fields (Rakow and Suslick, 2000; Keay and McNeil, 1998; Fang et al., 2011, 2009). Since gold nanoparticles were first used in glucose detecting (Zhao et al., 1992), a variety of nanomaterials have been widely employed in the study of enzymatic and non-enzymatic glucose sensors. However, because enzymes are expensive and their activity is significantly affected by many factors (such as temperature, pH, ionic strength, etc.) (Clark and Lyons, 1962; Updike and Hicks, 1967; Luo et al., 2011), the non-enzymatic glucose sensor has become a hot research topic in recent years (Xu et al., 2008).

It is well-known that sensitivity, selectivity and anti-poisoning performance are key factors for non-enzymatic sensors. These factors also belong to the catalytic performance of material of modified electrodes. Generally, catalytic performance of nanocrystals (NCs) can be finely tuned either by their composition, which mediates electronic structure (Zhang et al., 2007; Stamenkovic et al., 2007), or by their shape, which determines the

surface atomic arrangement and coordination (Narayanan and El-Sayed, 2004; Vidal-Iglesias et al., 2004). However, due to the difficulty of synchronized reduction of noble metal ions and transition element ions, it is usually necessary to prepare alloy nanoparticles at high reaction temperatures or with high-temperature calcination (Venkataraman et al., 2003; Li et al., 2010; Chang et al., 2008). High-temperature calcination easily leads to primary particle sintering. As the particle size and morphology were difficult to control, the specific surface area of the particles and the catalytic activity could not be confirmed. In recent years, many methods for preparing alloy nanoparticles were investigated with room-temperature synthesis. For example, three-dimensional Pt–Pd nanoflakes (Pt–Pd NFs) were synthesized on a homemade screen-printed gold film electrode (SPGFE) substrate using electrochemical deposition without a template (Niu et al., 2012). Holt-Hindle et al. (2008) synthesized Pt–Ir nanostructures with different quantities of iridium, which were directly grown on Ti substrates using a one-step facile hydrothermal method. Mahshid et al. (2013) constructed a nanowire arrays system consisting of an ordered configuration of Pt, Ni and Co in a single-bath solution through pulse electrodeposition. Hu et al. (2015) fabricated Pt-replaced porous Cu frameworks by covering the porous Cu frameworks with a Pt monolayer via the galvanic replacement reaction. Alternatively, some carriers were induced for improving surface area and electronic conduction. Sheng et al. (2015)

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fabricated platinum–cobalt alloy nanoparticles on carbon using the chemical reduction method. Multiwalled carbon nanotubes (MWNTs) or TiO₂-hollow sphere supported PtRu nanoparticles were prepared by radiolytic deposition, ultrasonic-electrodeposition and chemical reduction methods (Li et al., 2008; Lee et al., 2013; Kwon et al., 2012; Xiao et al., 2009). However, these sensors cannot both have the satisfied detection limit and linear detection range for detecting glucose.

Herein, we present the development of a high density surface defect Pt₃Ru₁ alloy nanoparticles based non-enzymatic sensor for the detection of glucose without a carrier. Furthermore, the performance of the proposed sensor was studied.

2. Experiments

2.1. Preparation of Pt₃Ru₁ nanoparticles

Pt₃Ru₁ nanoparticles were prepared as follows: 250 μL of a 0.162 M aqueous solution of a Na₂PtCl₆/RuCl₃ mixture in a molar ratio of 3:1 was first added to 5 mL cyclohexane containing 0.6 g of dioctyl sulfosuccinate sodium salt. The solution was sonicated for 1 min, and then 50 μL of hydrazine hydrate was swiftly injected. After the aging step, the reaction mixture was centrifuged and washed 4 times with trichloromethane and ethanol to obtain Pt₃Ru₁ nanoparticles.

2.2. Physicochemical characterization

The XRD analysis of the Pt₃Ru₁ nanoparticles was performed using a Rigaku D/max2500VB+/PC, at a scan speed of 8°/min and scan range of 20–90°. The HRTEM analysis of the Pt₃Ru₁ nanoparticles was performed with a Philips Tecnai G²F20. The chemical compositions of the Pt₃Ru₁ nanoparticles were measured by energy-dispersive X-ray spectroscopy (EDS) with the same HRTEM instrument.

2.3. Electrochemical measurements

The electrochemical measurements were performed using an electrochemical work station (CHI760D) in a conventional three-electrode electrochemical cell. The counter and reference electrodes included a platinum wire and a saturated calomel electrode (SCE), respectively. For the working electrode preparation, a 3-mm diameter glassy carbon electrode, was polished with 1.0, 0.3, 0.05 μm alumina slurries, washed with water, and subjected to ultrasonic agitation for 1 min in deionized water, and then a 10 μL (5 mg/mL) suspension of re-dispersed Pt₃Ru₁ nanoparticles in trichloromethane was pipetted on the glassy carbon electrode to build Pt₃Ru₁/GCE. All the electrochemical experiments were carried out at 25 ± 2 °C.

3. Results and discussion

3.1. Characterization of Pt₃Ru₁ nanoparticles

The XRD pattern of the Pt₃Ru₁ nanoparticles is shown in Fig. S1. As is shown, the 2θ angles of reflection peaks are 39.86°, 46.54°, 67.74°, 81.66°, and 86.12°, which contribute to the (111), (200), (220), (311) and (222) facets of the fcc structure, respectively. Compared with the XRD result of Pt, the diffraction peaks of Pt₃Ru₁ shift slightly towards a higher angle, which indicates the Ru atoms embedded into the crystalline lattice of Pt and the Pt₃Ru₁ alloy were formed (Viva et al., 2012).

Fig. 1 shows the HRTEM images of Pt₃Ru₁ nanoparticles. In Fig. 1A, the Pt₃Ru₁ nanoparticles were aggregated and the size of particles is 4 ± 1 nm. As is shown by the mark, the lattice spacing of (111) crystal plane is 0.2118 nm, which corresponds to the result of the XRD. Moreover, the high density of surface defects can be seen clearly, such as dislocation (a), stacking faults (b), interstitial defect (c) and twin boundary (d). The inset of e and f shows the fast Fourier transform (FFT) and inverse fast Fourier transform

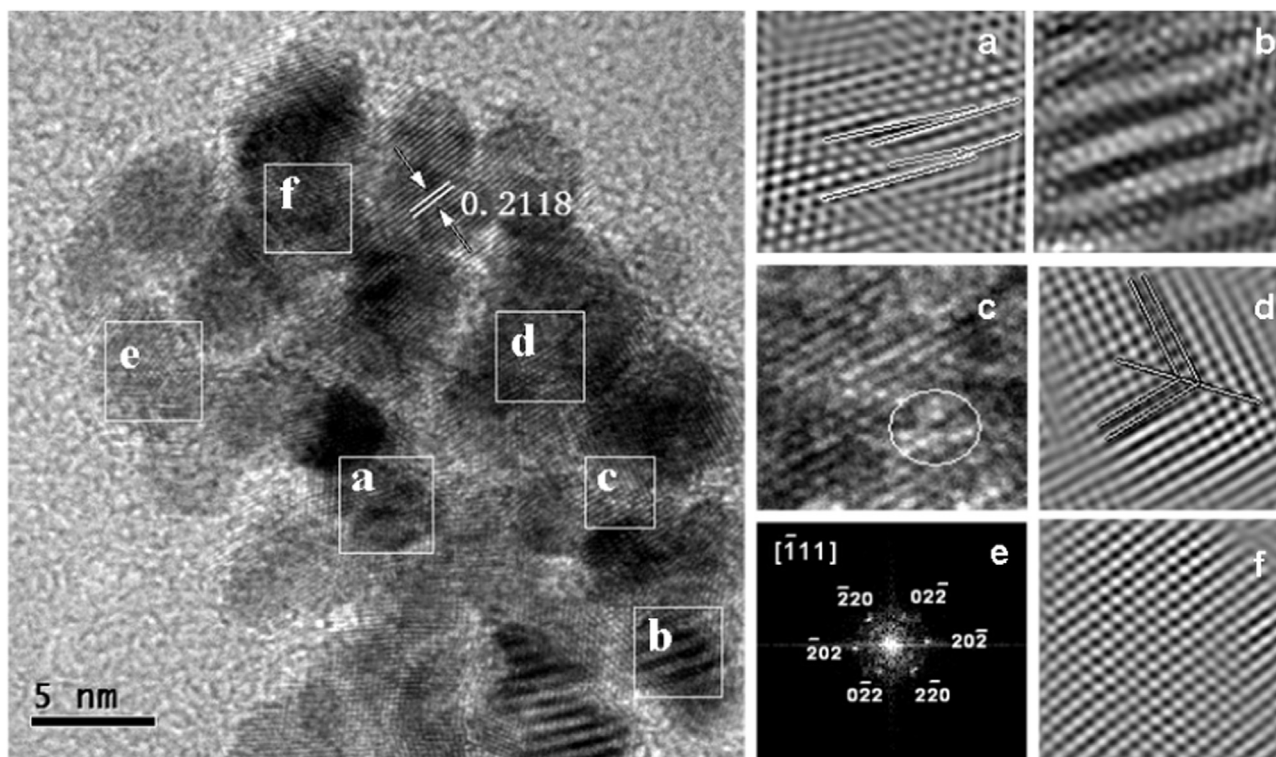


Fig. 1. TEM images of Pt₃Ru₁ nanoparticles.

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