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A Facile One-Step Electrochemical Synthesis of Nickel Nanoparticle/Graphene Composites for Non-Enzymatic Biosensor Applications

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Abstract

In recent years, graphene, two-dimensional closely packed honey-comb carbon lattice, has been attracting much attention in the field of electrochemistry due to its intrinsic properties and merits. Moreover, nickel has found extensive use in preparation of biosensors. In the present study, we applied a simple and straightforward approach for the development of nickel nanoparticle-graphene sheet (NiNP-GS) modified glassy carbon electrode (GCE) for nonenzymatic biosensor applications. The nanocomposites of NiNP-GS with various concentrations of nickel nanoparticles (NiNPs) synthesized through in situ chemical reduction procedure. In order to develop NiNP-GS modified GCE, 10μl suspension of NiNP-GS dispersed in deionized water was dripped on GCE surface and dried for 24 h at room temperature. The nanocomposites have been characterized using scanning electron microscope (SEM) and X-ray diffraction (XRD). Additionally, the sensing performance was evaluated by cyclic voltammetry. Results clearly demonstrated that the as-synthesized NiNPs with diameters less than 100 nm were well uniformly distributed on the surface of graphene nanosheets on graphene sheets. The characterization data also demonstrated that the nanocomposite film had a large surface area and enhanced electron-transfer rate compared with only Ni nanoparticles, due to an efficient electrical network through NiNPs direct anchoring on the surface of graphene. Furthermore, the sensivity and reproducibility of prepared electrode were better than other nickel electrodes reported. The good analytical performance, low cost and simple fabrication procedure make this novel electrode material promising for the development of effective non-enzymatic glucose sensor.

Keywords: Nano-Biosensor; Graphene; Nickel nanoparticles.

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1. Introduction

Diabetes is a world-wide public health problem. It is one of the leading causes of death and disability in the world. The diagnosis and management of diabetes mellitus requires a tight monitoring of blood glucose levels, Siangproh and Dungchai (2011), Rahman et al. (2010). In general, a glucose sensor is based on glucose oxidase due to high selectivity and sensitivity. However, the inevitable drawbacks such as the chemical and thermal instabilities originated from the intrinsic nature of enzymes as well as the tedious fabrication procedures may limit their analytical applications, Siqueira and Caseli (2010). To overcome the disadvantages of enzymes and mediator and develop a reagent less glucose sensor with low operating potential, alternative protocols have been developed via nanotechnology opening new perspectives in bioanalytical chemistry, Scognamiglio (2013).

Graphene is a new class of two-dimensional nanomaterial consisting of a single layer of sp^2 network of carbon atoms, Novoselov et al. (2004). Due to a host of intriguing properties such as high surface area, good biocompatibility, excellent electrical conductivity and ease of functionalization and production, wang et al. (2014), Liu et al. (2012), Ding et al. (2011), Baby et al. (2010), GNs provides an ideal platform to prepare functional nanomaterials for biosensors. Recently, GNs have been successfully combined with metal nanoparticles and applied to the fabrication of non-enzymatic glucose sensors, Yang et al. (2009). The combination of metal NPs and GNs not only prohibits the aggregation and oxidation of the metal NPs but also improve the performances of electrodes.

Ni-based nanomaterials exhibited remarkably catalytic oxidation activity over glucose as result of the catalytic effect originating from the formation of the redox couple of Ni (II)/Ni (III) on the electrode surface in alkaline medium, Nie et al. (2011). Considering the attractive properties of GNs, it is quite expected that GNs could provide an excellent support of the NiNPs for efficient electron transfer toward the oxidation of glucose. In addition, different morphological Ni nanostructures have been successfully synthesized via different methods in past decades. The high surface area- to-volume ratios of urchin-like nanostructures could increase the voltammetric signals for electroactive species that diffused from the bulk solution, Xu et al. (2008), Sarkar et al. (2011).

In this paper, a facile in situ synthetic route was used to prepare urchinlike NiNPs/GNs composites, and then the as-fabricated NiNPs/GNs modified glassy carbon electrode (GCE) was reported with high electrocatalytic activity toward the oxidation of glucose. Meanwhile, the interference was also investigated.

2. Experimental Procedure

NiNPs/GNs composites were prepared by in situ reducing reaction of GO and Ni-hydrazine complex using hydrazine hydrate. 100 mg of GO was dispersed into 50 mL of deionized (DI) water with ultrasonication for 1 h to form a stable GO suspension. 20 mL of an aqueous $NiCl_2$-$6H_2O$ solution (0.07, 0.14, 0.21 M) and 40 mL of $N_2H_4.H_2O$ were mixed with vigorous stirring to give a Ni–hydrazine complex suspension. The Ni–hydrazine complex suspension was added into GO solution and stirred for another 30 min. Subsequently, 20 mL of hydrazine hydrate and 40 mg of NaOH were added into the mixed solution, followed by ultrasonication for 15 min. After that, the above mixture was transferred into a 250 mL round-bottom flask and refluxed at 100 °C for 5 h. After cooled to room temperature naturally, the as-synthesized solid products were collected by vacuum filtration, washed with DI water and ethanol for three times to remove the excess chemicals. The final product was then dried in a vacuum oven at 60 8C for 24 h to obtain the dried NiNPs/ GNs nanocomposites.

The morphologies and microstructure of the samples were investigated by powder X-ray diffraction system (XRD Philips X'Pert with Cu Kα radiation), scanning electron microscope (SEM Seron AIS 2100). All electrochemical experiments including cyclic voltammetry (CV) were carried out using PARSTAT2273 electrochemical work Station. Electrochemical measurements were performed in 0.1 M NaOH supporting electrolyte at room temperature using a three-electrode system consisting of a KCl saturated calomel electrode (SCE) as the reference electrode, a platinum electrode as the counter electrode, and the NiNPs/GNs modified GCEs as the working electrode. The NiNPs/GNs modified GCE was fabricated by dripping 10 mL of the NiNPs/GNs dispersion onto the GCE surface and dried 12 h in air. The NiNPs/GNs dispersion was prepared by dispersed 10 mg GNs–NiNPs composites in 10 mL of deionized water with sonication for 10 min.