



# Nonenzymatic electrochemical detection of glucose using well-distributed nickel nanoparticles on straight multi-walled carbon nanotubes

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## ABSTRACT

A nonenzymatic electrochemical sensor device was fabricated for glucose detection based on nickel nanoparticles (NiNPs)/straight multi-walled carbon nanotubes (SMWNTs) nanohybrids, which were synthesized through in situ precipitation procedure. SMWNTs can be easily dispersed in solution after mild sonication pretreatment, which facilitates the precursor of NiNPs binding to their surface and results in the homogeneous distribution of NiNPs on the surface of SMWNTs. The morphology and component of the nanohybrids were characterized by scanning electron microscopy (SEM) and X-ray powder diffraction (XRD), respectively. Cyclic voltammetry (CV) and amperometry were used to evaluate the catalytic activity of the NiNPs/SMWNTs nanohybrids modified electrode towards glucose. It was found that the nanohybrids modified electrode showed remarkably enhanced electrocatalytic activity towards the oxidation of glucose in alkaline solution compared to that of the bare glass carbon electrode (GCE), the NiNPs and the SMWNTs modified electrode, attributing to the synergistic effect of SMWNTs and  $\text{Ni}^{2+}/\text{Ni}^{3+}$  redox couple. Under the optimal detection conditions, the as-prepared sensors exhibited linear behavior in the concentration range from 1  $\mu\text{M}$  to 1 mM for the quantification of glucose with a limit of detection of 500 nM ( $3\sigma$ ). Moreover, the NiNPs/SMWNTs modified electrode was also relatively insensitive to commonly interfering species such as ascorbic acid (AA), uric acid (UA), dopamine (DA), galactose (GA), and xylose (XY). The robust selectivities, sensitivities, and stabilities determined experimentally indicated the great potential of NiNPs/SMWNTs nanohybrids for construction of a variety of electrochemical sensors.

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## 1. Introduction

Glucose sensing is of significantly importance in diagnosis and treatment of diabetes mellitus as well as monitoring and control of food preparation processes (Jin et al., 1997; Mizutani et al., 1998). Until now, few spectrophotometric strategies have been developed for measuring the glucose levels, which is attributed to the lack of chromophoric or fluorophoric ligands for glucose. Electrochemistry holds great potential as the next-generation detection strategy for glucose because of its high sensitivity, reliability and selectivity, simple instrumentation, low cost and excellent compatibility with miniaturization. Glucose oxidases (GOx) were widely used for the fabrication of the electrochemical sensor devices for glucose determination (Zayats et al., 2002; Liu et al., 2010; Wang, 2008; Zeng et al., 2011; Liu et al., 2007). Although these enzyme-based sensors show high selectivity and excellent sensitivity, inevitable drawbacks such as chemical and thermal deformation originating from the intrinsic nature of the enzyme

as well as the severe interferences from other oxidable species in blood samples may limit their applications (Kohma et al., 2007). The nonenzymatic glucose sensors based on direct electrocatalytic oxidation of glucose at an electrode surface would exhibit conveniences and advantage to avoid the enzyme electrode drawbacks.

Recently, a major concern in a practical nonenzymatic glucose sensor has been focused on the efforts to develop highly electrocatalytic activity of the electrode materials, which is the key factor that affects both the sensitivity and selectivity of glucose detection. Various nanostructured metals (Pt, Au, Pd, Ni, Cu) (Park et al., 2003; Jena and Raj, 2006; Bai et al., 2010; Lu et al., 2009), alloys (Pt–Pb, Pt–Ir, Pt–Au) (Wang et al., 2008; Hindle et al., 2008; Ryu et al., 2010) and metal oxides (CuO,  $\text{Co}_3\text{O}_4$ , NiO) (Zhang et al., 2008; Ding et al., 2010; Safavi et al., 2009) have been explored as the electrode substrates for directly electro-catalyzing glucose. Among these, Ni-based nanomaterials exhibit remarkably catalyzed capability for glucose as result of the catalytic effect originating from the redox couple of  $\text{Ni}(\text{OH})_2/\text{NiOOH}$  on the electrode surface in alkaline medium. Most of Ni-based glucose sensors were fabricated by modifying traditional electrodes with Ni-based nanomaterials such as dispersing NiNPs in disordered graphite-like carbon (You et al., 2003), doping carbon paste electrode with nano-NiO powder

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(Mu et al., 2011), and mixing powdered nanoscale nickel hydroxide with graphite powder and ionic liquid (Safavi et al., 2009). However, one severe problem with these electrodes is the instability of the catalytic film for extended period of time under applied potential. To solve this problem, Ni was loaded on the surface of various nanomaterials such as carbon nanofiber (Liu et al., 2009), Ti/TiO<sub>2</sub> nanotube array (Wang et al., 2010). However, these support materials were not favorable for facilitating electron transfer during electrochemical process. Therefore, it is still highly desirable for Ni-based glucose sensor to develop novel electrode materials with good stability, excellent conductivity and high electrocatalytic activity.

Carbon nanotubes (CNTs) have attracted growing interest in nonenzymatic sensors due to their combination of beneficial properties such as their relatively large surface area, excellent electric conductivity, good mechanical properties and outstanding ability to promote electron-transfer reactions. Recent studies of CNT-based enzyme-free sensors have focused on utilizing CNTs as the templates for supporting metal or metal oxides nanoparticles catalysts (such as Pt, Pd, Cu, MnO<sub>2</sub>) (Meng et al., 2009; Su et al., 2010; Yang et al., 2010; Chen et al., 2008), which cannot only improve the dispersion of catalysts but also enhance the load amounts of catalysts. Therefore, it is expected that the advantageous features of CNTs coupling with Ni-based nanomaterials in electrocatalytic oxidation of glucose can make Ni/CNTs nanohybrids very promising in the development of nonenzymatic glucose sensors with high sensitivity, selectivity, and stability. Although there are few reports about CNTs coupling with Ni-based nanomaterials for electrocatalytic oxidation of glucose (Shamsipur et al., 2010; Zheng et al., 2009), these strategies involve harshly acid-oxidized pretreatment of CNTs, which causes structural damage to the CNTs and lead to the loss of their electrical conductivity and mechanical properties. The loss of mechanical properties is not assisting catalysts stably distributed on the wall of CNTs, and usually results in the poor sensitivity and stability for glucose determination. Furthermore, these sensors exhibit poor reproducibility and great limitation in their applications in real samples. Therefore, it is indispensable to develop an efficient functionalized method without or with little damage to the CNTs for supporting catalysts in batch.

Herein, we utilized mildly pretreated straight multi-walled CNTs (SMWNTs) as the templates for supporting NiNPs catalysts, and developed a nonenzymatic glucose sensor based on NiNPs/SMWNTs nanohybrids, which were synthesized through in situ precipitation procedure. Comparing with the tangled CNTs widely applied in nonenzymatic sensors, SMWNTs can be more easily dispersed in solution after mild sonication pretreatment, which keep them intrinsic electrical conductivity and mechanical properties. These properties can facilitate the NiNP precursor binding to their surface, which make NiNPs stably and homogeneously distributed on the surface of SMWNTs. The well-distributed NiNPs can be easily and fully accessing to glucose, and amplifying the electrochemical signal for glucose determination. An efficient electrical network through NiNPs directly anchoring on the surface of SMWNTs can promote the electron transfer rate and improve the sensitivity. More importantly, the NiNPs/SMWNTs can be prepared on a large scale by precipitation procedure, which not only makes this sensor exhibit excellent reproducibility but also broadens their application in the real sample analysis. Moreover, the modification of glass carbon electrodes (GCE) with the NiNPs/SMWNTs nanohybrids not only increases their active area but also promotes the electron transfer for the glucose oxidation reaction via the SMWNTs. The proposed sensor demonstrated that NiNPs/SMWNTs could allow highly sensitive, fast, stable, and reproducible amperometric sensing of glucose.

## 2. Experiment

### 2.1. Reagents and materials

Straight multi-walled carbon nanotubes (SMWNTs) were prepared according to the previous report (Yang et al., 2008). D-(+)-Glucose, ascorbic acid (AA), uric acid (UA), dopamine (DA), galactose (GA), and xylose (XY) were obtained from Alfa Aesar. All of other reagents were of analytical grade and used without further purification. All aqueous solutions were prepared using ultrapure water, which was obtained through a Millipore Milli-Q water purification system (Billerica, MA) with an electric resistance >18.3 MΩ. The glucose stock solution was allowed to mutarotate for at least 24 h before use.

### 2.2. Apparatus and measurement

Scanning electron microscopy (SEM) images were conducted using (FEI Nova NanoSEM 200). X-ray diffraction (XRD) analysis was recorded on a (Smart APEX CCD, Bruker, Germany). The cyclic voltammetry (CV) and amperometry were executed on a CH Instruments model 760C electrochemical analyzer (Shanghai, China). All of 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 wire as the counter electrode, and the modified glass carbon electrodes (GCE) as the working electrode.

### 2.3. Synthesis of NiNPs/SMWNTs nanohybrids

Prior to the preparation of NiNPs/SMWNT nanohybrids, SMWNTs were pretreated by sonication in a mixture of sulfuric acid and nitric acid (3:1) for 30 min, and then washed with distilled water by centrifugation until the pH of the resulting SMWNTs solution become neutral.

NiNPs/SMWNTs nanohybrids were obtained from the reduction of NiO/SMWNTs complex, which were synthesized through in situ precipitation procedure. Briefly, 20 mg Ni(NO<sub>3</sub>)<sub>2</sub> was added into 20 mL of the treated SMWNTs solution and ultrasonic agitated for 1 h at room temperature. Subsequently, 60 mL of 2.2 mM NaOH solution was added dropwise to the above mixture under constant stirring at 80 °C, the Ni(OH)<sub>2</sub>/SMWNTs complex was formed and the reaction was carried out under magnetic stirring for 2 h at room temperature. Finally, the Ni(OH)<sub>2</sub>/SMWNTs complex was separated and rinsed with distilled water several times, followed by heat treatment at 650 °C for 2 h in the H<sub>2</sub>/N<sub>2</sub> mixture atmosphere (H<sub>2</sub>/N<sub>2</sub> = 3/1) to reduce Ni<sup>2+</sup> and get NiNPs/SMWNTs nanohybrids.

### 2.4. Fabrication of the NiNPs/SMWNTs-modified electrode

Prior to coating, GCE with a diameter of ~3 mm were polished on a microcloth (Buehler) with 0.3 and 0.05 μm alumina slurry for 5 min, followed by sonication in ultrapure water and ethanol for 5 min in each. Then, the polished electrodes were dried under mild nitrogen stream. 2 mg NiNPs/SMWNTs was dispersed in 10 mL ultrapure water with the aid of ultrasonic agitation to give 0.2 mg mL<sup>-1</sup> black solution. 10 μL of the black solution was dropped onto the GCE surface, and the electrode was dried slowly to obtain the NiNPs/SMWNTs modified electrode. A NiNPs-modified GCE and SMWNTs-modified GCE were prepared in the same way.

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