#### Journal of Alloys and Compounds 712 (2017) 742-751

Contents lists available at ScienceDirect

# Journal of Alloys and Compounds

journal homepage: http://www.elsevier.com/locate/jalcom

# Multi-dimensional Ag/NiO/reduced graphene oxide nanostructures for a highly sensitive non-enzymatic glucose sensor



ALLOYS AND COMPOUNDS

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## A R T I C L E I N F O

Article history: Received 19 December 2016 Received in revised form 11 April 2017 Accepted 13 April 2017 Available online 15 April 2017

Keywords: Glucose sensor Graphene oxide Nickel oxide Silver Multi-dimensional structure

# ABSTRACT

A highly sensitive non-enzymatic glucose sensor was fabricated by hybridizing 0- dimensional (0D) Ag nanoparticles, 3D flower-like nickel oxide (NiO) nanostructures, and 2D reduced graphene oxide (rGO) as the sensing interface by hydrothermal synthesis. The resulting hybrid structures were characterized by Raman spectroscopy, X-ray photoelectron spectroscopy, field-emission scanning electron microscopy, energy dispersive X-ray spectroscopy, and high-resolution transmission electron microscopy. The Ag/ NiO/rGO fabricated in this study showed high electrochemical activity towards the oxidation of glucose in a 0.1 M NaOH solution. At an applied potential of +0.6 V, it exhibited a rapid response time (<4 s), a broad linear range of glucose concentrations up 25 mM with an extraordinarily high sensitivity of 1869.4  $\mu$ A mM<sup>-1</sup> cm<sup>-2</sup>. The detection limit was as low as 2.44  $\mu$ M. In addition, the response towards common interfering species, such as sucrose, lactose, fructose, ascorbic acid, dopamine, and uric acid were low enough to be avoidable.

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# 1. Introduction

Diabetes mellitus is one of the most common endocrine disorders of the carbohydrate metabolism, in which patients experience glucose concentration deviations from the normal range of 80–120 mg/dL (4.4–6.6 mM). According to the newly-released Diabetes Atlas, 7th edition, from the International Diabetes Federation (IDF), more than 415 million people worldwide suffered from diabetes mellitus in 2015, which is predicted to increase to 642 million people in 2040 [1]. Therefore, the development of fast, simple and reliable technology for monitoring the glucose concentration is one of the most important issues in many areas, such as the food industry, biotechnology, and clinical diagnostics.

Many studies have focused on the development of effective glucose sensor technologies since the invention of the first glucose enzyme electrodes by Clark and Lyons [2]. Enzymatic sensors based on immobilized glucose oxidase (GOx) are used widely for glucose detection owing to their excellent sensitivity and selectivity over interference materials, such as ascorbic acid and uric acid. On the other hand, there are many problems induced by biomaterials, such as poor stability, high cost of enzymes, and weakness to environmental conditions, including pH, temperature, humidity, and the

interferences caused by other substances [3,4]. To solve these problems, non-enzymatic electrodes have been explored widely, which allows glucose to be oxidized directly on the electrode surface without enzymes. Recent studies have shown that electrodes modified with noble metals (Ag, Pt, Au) [5-7], metal alloys (Pt, Au. Ir, Pd, Cu, and Co) [8–10], transition metals (Ni, Pd) [11,12] and metal oxides [13,14] can be used effectively to detect glucose in the absence of GOx. On the other hand, most non-enzymatic sensors developed so far are unsatisfactory due to the high cost of the rare metal precursors, low sensitivity, and poor selectivity, which are probably due to the adsorption and accumulation of intermediates, low electrocatalytic activity, and electrical conductivity, and/or surface poisoning during the electrochemical process [15,16]. Therefore, the design of effective electrocatalyst systems that can overcome these problems will be needed to fully replace enzymatic glucose sensors.

Among the various cost-effective metal oxides, nickel oxide (NiO) is used widely in many applications, including supercapacitors [17], gas sensors [18], biosensor [19], lithium batteries [20], and catalysts [21], owing to its unique optical, electrical, and electrocatalytic properties. Although NiO has attracted attention for non-enzymatic glucose sensors owing to its high electrocatalytic activity, electron transfer capability, and good biocompatibility [22], its low glucose sensitivity due to the low electrical conductivity and surface area should be solved. To overcome these drawbacks, many attempts have been made to enhance the glucose



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sensitivity by creating a range of nanostructured, which have shown promising characteristics for applications sensor in area of environmental monitoring [23,24], energy [25–27] and health [28]. Therefore, the design of a hybrid system composed of multicomponents with the multi-dimensional structures can be a promising strategy because the final properties can be improved dramatically by the synergistic effects of each component.

This paper reports a non-enzymatic glucose sensor that shows the highest glucose sensitivity ever reported, as well as excellent selectivity and stability by hybridizing 0-dimensional (0D) silver (Ag) nanoparticles, 2D reduced graphene oxides (rGO), and 3D NiO nanoflower structures fabricated by a simple hydrothermal synthesis. Ag nanoparticles can increase the electrocatalytic activity by enhancing charge separation during the electro-oxidation/ reduction reactions [29] with less poisoning by the chemisorbed intermediates generated by the electrooxidation of glucose compared to other metals [30,31]. In addition, rGO can enhance the glucose sensitivity by increasing the surface area and electrical conductivity, enhancing glucose adsorption and charge transfer, and providing numerous anchoring and dispersion sites for metal and metal oxide nanostructures because it has abundant functional groups and restored electron pathways after reduction [16,32,33].

## 2. Experimental details

## 2.1. Materials

Nickel(II) nitrate hexahydrate (NiNO<sub>3</sub>.6H<sub>2</sub>O), ammonium hydroxide (NH<sub>4</sub>OH), silver nitrate (AgNO<sub>3</sub>), potassium permanganate (KMnO<sub>4</sub>), hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) (30 wt %), sulfuric acid 98% (H<sub>2</sub>SO<sub>4</sub>), sodium hydroxide (NaOH), ethanol (C<sub>2</sub>H<sub>5</sub>OH), polyvinylpyrrolidone (PVP, C<sub>6</sub>H<sub>9</sub>NO)<sub>n</sub>, nafion, isopropanol (IPA), D-(+)-glucose, D-(-)-fructose, lactose, sucrose, ascorbic acid (AA), uric acid (UA), dopamine (DA), horse serum (HS), and rabbit serum (RS)

were purchased from Sigma-Aldrich Co. (USA). Expandable graphite powder was obtained from Asbury Graphite Mills Inc. (USA). All chemicals were used as received. Deionized (DI) water was used to wash the samples in all experiments.

# 2.2. Preparation of Ag/NiO/rGOs

A GO suspension was prepared by the oxidation of expanded graphite powder by a modified Hummer's method, as described elsewhere [18]. To fabricate Ag/NiO/rGO, 0.1 mmol of nickel (II) chloride hexahydrate and a pre-calculated amount of silver nitrate was added to 10 mL of DI water and stirred at room temperature. 1 mL of ammonium hydroxide (NH<sub>4</sub>OH) and 5 mg of PVP was introduced to the mixture and stirred continuously for 1 h. The mixed solution was then added to 5 mL of an aqueous GO slurry with a 5 mg/mL solid content. Various amounts of AgNO<sub>3</sub> were tested as described in Fig. S1 and S2 (Supporting Information), and the optimal amount was found to be 5 mol% of AgNO<sub>3</sub> to NiO. The various volume ratios between Ag/NiO and GO were also tested, as shown in Fig. S3 (Supporting Information), and the optimal ratio was found to be 1:1. The samples were transferred to a Teflon-lined stainless steel autoclave, heated to 140 °C and maintained at that temperature for 12 h to build a network structure of GO. The optimal hydrothermal temperature was selected after testing over a wide range of temperatures, as shown in Fig. S4 (Supporting Information). After the autoclave was cooled slowly to room temperature, the obtained samples were submerged with deionized water (DI) to eliminate all the unreacted chemicals followed by freeze drying at  $-37\ ^\circ C$  for 2 days to remove the water without destroying the networks. Finally, the Ag/NiO/rGO was calcined at 400 °C for 3 h in flowing nitrogen to convert Ni(OH)<sub>2</sub> to NiO and obtain the multi-dimensional hybrid structure with enhanced electrical conductivity by the reduction of GO to rGO. The samples prepared in this study were labeled xAg/NiO/rGO and y:zAg/NiO/



Scheme 1. Schematic diagram of the fabrication steps of Ag/NiO/rGO.

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