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Non-enzymatic voltammetric glucose sensor made of ternary NiO/Fe_3O_4-SH/para-amino hippuric acid nanocomposite

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ABSTRACT

The present study has been focused on a facile strategy to fabricate an electrochemical sensor based on nickel oxide decorated Fe_3O_4 nanoparticles/poly(p-aminohippuric acid)-sodium dodecyl sulfate nanocomposite immobilized on the modified glassy carbon electrode (NiO@ Fe_3O_4-SH/PPAH-SDS/GCE). PPAH-SDS film was prepared by electropolymerization and nickel oxide nanostructures were deposited on the Fe_3O_4-SH/PPAH-SDS by electrochemical method. The as-synthesized nanocomposite was characterized by scanning electron microscopy (SEM) and energy dispersive spectroscopy (EDS) for morphological and elemental analysis, respectively and the electrochemical characteristics were studied by the cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS) methods. The differential pulse voltammetry (DPV) was used for the sensitive determination of glucose in alkaline solution in two linear concentration ranges, 0.1–10.0 μM and 10.0–300.0 μM, with a detection limit of 0.13 μM (S/N = 3) based on the first linear range. Meanwhile, the presented sensor showed a promising performance for the quantification of glucose in human serum as a real sample reflecting the practicability of fabricated glucose sensor.

1. Introduction

Diabetes is a group of metabolic diseases resulting from insulin deficiency reflected by glucose concentrations in blood and associated with long-term damage and failure of diverse organs specially the kidney, heart, eyes, and blood vessels [1]. Hence, the early and sensitive diagnosis of unusual glucose levels in the bloodstream is very significant for appropriate treatment to decrease the mentioned risks. Several methods have been applied to detect the glucose such as fluorescent, acoustic, optical, electronic, capacitive, transdermal and electrochemical methods [2–7] which among them some electrochemical sensing approaches have drawn so much attention due to their acceptable sensitivity, simple instrumentation, long stability, rapid response and flexibility [8–12]. On the other hand, considerable regard has been paid for developing and engineering the non-enzymatic electrochemical glucose sensors to overcome some disadvantages of the enzyme-modified electrodes such as instability, high cost of enzymes, complicated immobilization technique and critical operating procedure. Due to the absence of target enzyme in this kind of electrodes, an efficient sensitive electrocatalyst is necessary for the fabrication of prominent non-enzymatic biosensors, as it can greatly influence the processes of substrate direct oxidization with minimum resistance, electrochemical reactions occurring at electrolyte/electrode interfaces and electron conduction in electrodes [13,14].

In this context, extensive studies have been carried out for the synthesis of various nanostructured compounds to be used as an electrode material for the development of non-enzymatic glucose sensors. In earlier, numerous glucose sensors have been fabricated based on the noble metals (Pt, Pd and Au) but this kind metals suffer from the high cost and low abundance in the earth [15]. Recently, many glucose sensors were designed using transition metals and their oxide nanostructures such as Co, Cu, Ni, Co_3O_4, NiO, WO_3, TiO_2, CdO/CuO, MnO_2, Fe_2O_3, Mn_3O_4 and ZnO, etc. [16]. These catalysts can be employed for electrochemical detection of glucose due to their low cost, ease of synthesis and long-term stability [17,18]. It is noteworthy that among them, Ni-based nanomaterials exhibited the biocompatibility and also remarkable catalytic oxidation activity towards glucose as a result of the electrocatalytic effect originating from the generation of Ni(II)/Ni(III) redox couple on the electrode surface in alkaline medium [19,20]. Synthetic methodology to prepare nano-sized NiO involves electrochemical deposition [21], atomic layer deposition [22], magnetron sputtering deposition [23], hydrothermal synthesis [24,25], etc. which

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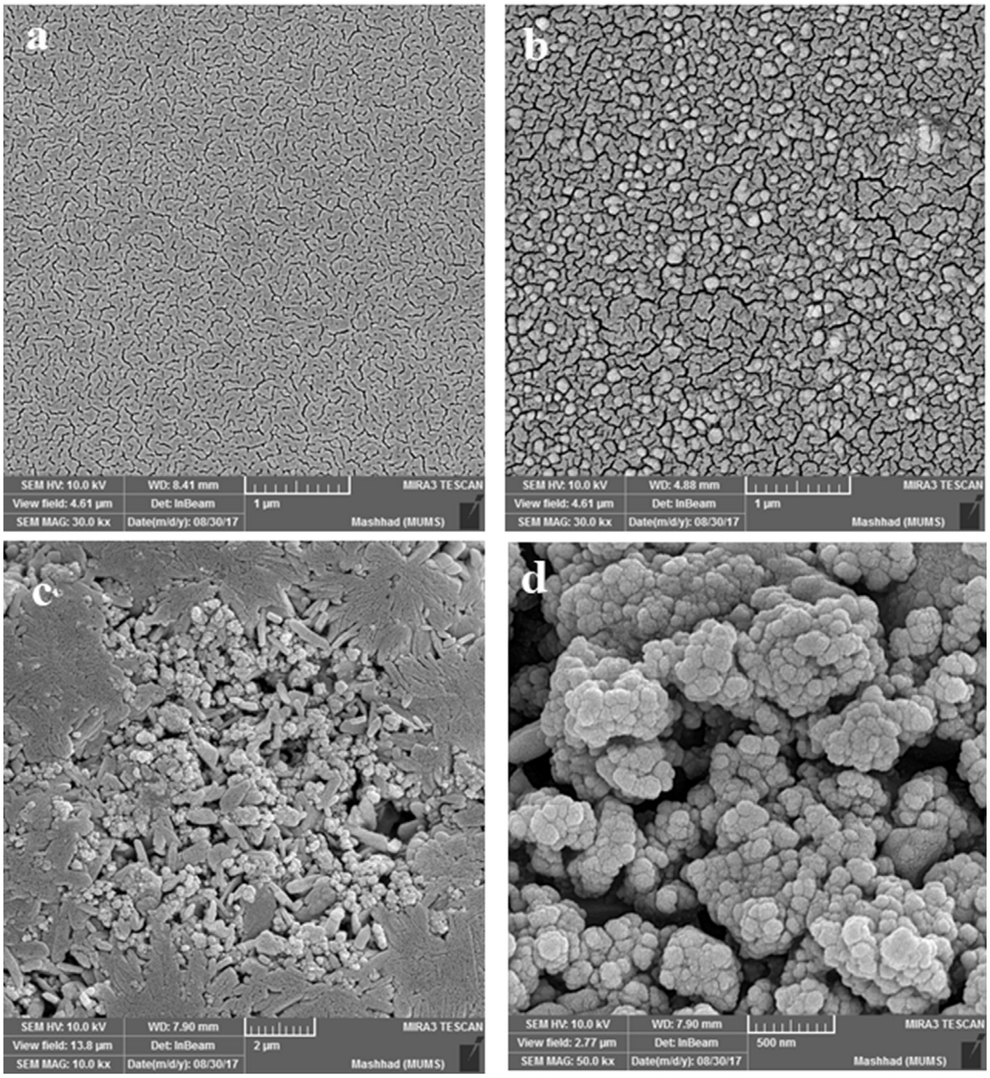

Fig. 1. SEM images of the PPAH-SDS/GCE (a), Fe_3O_4-SH/PPAH-SDS/GCE (b) and NiO@Fe_3O_4-SH/PPAH-SDS/GCE (c,d).

the electrodeposition has widely used due to its fast and facile synthetic procedure.

Additionally, magnetic iron oxide nanoparticles (Fe_3O_4 NPs) have also gained a great deal of regard due to their high biocompatibility, super-paramagnetic property, displaying no hemolytic activity or genotoxicity, better interaction with desired biomolecules [26,27] as well as their potential for providing the control of electrochemical processes [28,29]. In fact, the biological recognition elements can be attached to the surface of these NPs to develop the various catalytic and affinity sensors. Furthermore, they can be retained and removed with a magnet without affecting the transducer surface, thus creating possibilities for regeneration and reuse [30].

It is noteworthy that in biosensing platforms, the synergic effect of nanoparticles with other material, which have excellent conductivity and catalytic properties, can provide the necessary conduction pathways for electrons on the electrode surface to enhance the current signal [31]. In this regard, conducting polymer modified electrodes can be referred which are usually prepared by single-step electropolymerization. Conducting polymers as novel organic semiconducting materials have found applications in electrocatalysis and determination of biomolecules due to homogeneity of electrochemically deposited films, strong adherence to the electrode surface especially in contact with aggressive media such as strong acids or bases and their chemical and environmental stability [32]. Meanwhile, nanocomposites of metal- or metal oxide-conducting polymer are expected to be a significant category of materials in the nanotechnology [33] because they combine the advantages of inorganic materials (modulus, mechanical strength and thermal stability) and organic polymers (ductility, flexibility and process ability) [34].

Herein, we show that ternary NiO@Fe_3O_4-SH/PPAH-SDS nanocomposite can be used as sensory means for the electrocatalytic detection of glucose. The present work involves a description of synthesis procedure, electrode characterization and general analysis of its electrochemical performance for the detection of glucose. Additionally, the universal feasibility of the designed sensor is demonstrated by measuring the glucose concentration in blood serum as real sample.

2. Experimental

2.1. Materials

Glucose ($C_6H_{12}O_6$), sodium hydroxide (NaOH), nickel chloride hexahydrate ($NiCl_2.6H_2O$), Sodium dodecyl sulfate (SDS), Potassium chloride (KCl), para-amino hippuric acid (PAH), Ethanol 96% used in this work were obtained from Merck. Glucose (Sigma, 99%) was purchased from USA and used without further purification. The glucose stock solution was prepared by 0.1 M NaOH. All other chemicals were of analytical grade and were used as received without any purification and solutions were prepared with deionized water. The phosphate buffer solution (PBS) was prepared from phosphoric acid (H_3PO_4),