



High-performance cholesterol sensor based on the solution-gated field effect transistor fabricated with ZnO nanorods

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ABSTRACT

A high-performance cholesterol sensor based on solution-gated field-effect-transistor (FET) was fabricated by using the vertically aligned ZnO nanorods (ZnO NRs) grown selectively on pre-patterned substrate in solution. The structural characterization showed that the as-grown ZnO NRs are vertically aligned, high purity single crystalline. The active layer of ZnO NRs between source and drain electrodes was immobilized with cholesterol oxidase (ChOx) enzyme. The performance of the fabricated FET sensor has been examined with the cholesterol solutions with and without electroactive species, the human serum (H4522), and the freshly drawn blood sample. The FET sensor provided a real-time response towards a wide range of cholesterol concentration (0.001–45 mM) with high sensitivity ($10 \mu\text{A cm}^{-2} \text{mM}^{-1}$) and selectivity.

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1. Introduction

Accurate and fast determination of cholesterol concentrations is of great importance for clinical diagnosis because cholesterol is an important biomarker for the management of many diseases, including hypertension, coronary heart disease, arteriosclerosis and dysfunctions in lipid metabolism. Thus, there is an increased interest in the development of new cholesterol biosensors (Arya et al., 2008; Singh et al., 2012). However, the majority of cholesterol biosensors are based on the amperometric detection with cholesterol oxidase (ChOx) enzyme electrodes (Umar et al., 2009a, 2009b, 2009c; Ahmad et al., 2012a, 2012b). In an amperometric cholesterol sensor, the current is a direct measure of the rate of the enzymatic regeneration reaction carried out by oxygen or artificial redox mediators, providing quantitative information on the cholesterol level. However, this method suffers from poor manufacturability and a narrow range of linear response restricted to low cholesterol concentrations, which falls below physiological concentrations of cholesterol in human blood serum. Consequently, sample dilution is generally necessary for these methods. Therefore, it is highly desirable to develop a low-cost, portable, wide linear range and user-friendly analytical

platform for analysis of cholesterol concentrations in wide linear range.

Field-effect transistors (FETs) have attracted an increasing interest as primary candidates for fabricating state-of-the-art sensor platforms because they are capable of achieving high current amplification and sustaining an enhanced signal-to-noise ratio (Janata and Josowicz, 2003; Forzani et al., 2004; Star et al., 2004; Freeman et al., 2007). The sensing performance of the FETs is greatly affected by the integration method of the enzyme. In order to extend the dynamic range or increase the selectivity, additional membranes such as Nafion and polyvinyl pyridine (Umar et al., 2009; Kokkinos and Economou, 2011) were covered on the enzyme matrices. Most of the current works are focused on carbon nanotube and silicon nanowire, only a limited number of studies focusing on the use of oxide semiconductors. However, the silicon nanowire surfaces can be easily oxidized and thus degrade the device reliability and sensitivity (Bunimovich et al., 2006), while the chirality of carbon nanotube remains an unsolved problem. Thus, it is challenging to investigate metal oxide material for biosensing application.

Zinc oxide (ZnO) is one of the most exciting materials for its versatile properties such as semiconducting (band gap of 3.37 eV), piezoelectric, bio-safe and biocompatible nature (Ahsanulhaq et al., 2009; Hahn, 2011; Umar et al., 2008). It has been demonstrated that both the size and shape or morphology has an influence on the properties of material (Dai et al., 2009; Yang et al., 2009; Wei et al., 2010; Ahmad et al., 2012; Tripathy et al., 2012; Umar et al., 2006; Umar et al., 2007). ZnO nanorods (NRs) have attracted considerable interest in the aspect of sensors due to many advantages, including

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large surface-to-volume ratio, excellent biological compatibility, high electron-transfer rates, non-toxicity and bio-safety. Notably, ZnO with a high isoelectric point (IEP $\sim$ 9.5) is suitable for the adsorption of low IEP proteins or enzyme (ChOx, IEP $\sim$ 4.9). Positively charged ZnO nanorods matrix not only provide a friendly microenvironment for the negatively charged proteins or enzyme to retain its activity but also promote the direct electron transfer between the enzyme and the electrode to a large extent (Zhang et al., 2004; Wang et al., 2012).

Herein, we present the successful demonstration of a liquid-ion gated FET sensor using ChOx enzyme immobilized ZnO NRs to detect cholesterol. The fabricated FET sensor showed a high sensitivity and a wide linear range with low detection limit. To the best of our knowledge, the vertically aligned ZnO NRs are used, for the first time, for the fabrication of FET based sensor for rapid detection of cholesterol. The applicability of the sensor was assessed by the detection of cholesterol in human serum and whole blood samples.

2. Experimental details

2.1. Reagents

Cholesterol oxidase (EC 1.1.3.6 from *Streptomyces* species, 20 U/mg), Nafion (5 wt% in lower aliphatic alcohol and water mixture), bovine serum albumin (BSA > 98%), cholesterol (water soluble), glucose (d-(+)-99.5%), ascorbic acid (AA), L-cysteine (L-Cyst), uric acid (UA), human blood serum (H4522), polymethyl methacrylate (PMMA), methylisobutylketone (MIBK), isopropanol (C_3H_8O), Zinc nitrate hexahydrate ($Zn(NO_3)_2 \cdot 6H_2O$,

99%), hexamethylenetetramine (HMTA, 99%) and phosphate buffered saline (PBS, pH=7.4) were purchased from Sigma-Aldrich and used without further purification.

2.2. Synthesis and characterization of ZnO nanorods

In a typical synthesis process, an equal molar of $Zn(NO_3)_2 \cdot 6H_2O$ (0.05 M) and HMTA (0.05 M) was dissolved in 50 mL of distilled water and transferred into a Pyrex glass bottle, followed by heating in a laboratory oven for 6 h at 80 °C after suspending the seeded substrates upside down. After the reaction, the electrodes were rinsed with DI water to remove impurities before characterization. Details of ZnO nanorods growth are available elsewhere (Ahsanulhaq et al., 2007).

The morphology and crystallinity of as-synthesized ZnO NRs was examined by field emission scanning electron microscopy (FESEM), transmission electron microscopy (TEM), UV/visible, Fourier transform infrared spectroscopy (FTIR), and X-ray diffractometer (XRD) measured with Cu-K α radiations ($\lambda=1.54178$ Å) in the range of 30–65° with 8°/min scanning speed.

2.3. Fabrication of FET based cholesterol sensors and measurements

A schematic diagram of the FET sensor is shown in Fig. 1(i). The sensing device consisted of a 200 nm thermally formed SiO_2 on Si substrates, where SiO_2 layer acted as the gate dielectric and Si as a back gate. ZnO NR arrays were fabricated directly on Si/ SiO_2 by the site-selective growth of nanorods by solution process. Fig. 1(ii) shows the fabrication process of FET-based cholesterol sensor. First, a thin layer of PMMA (400 nm thick) was spun on the SiO_2 and hard baked at 170 °C for 30 min.

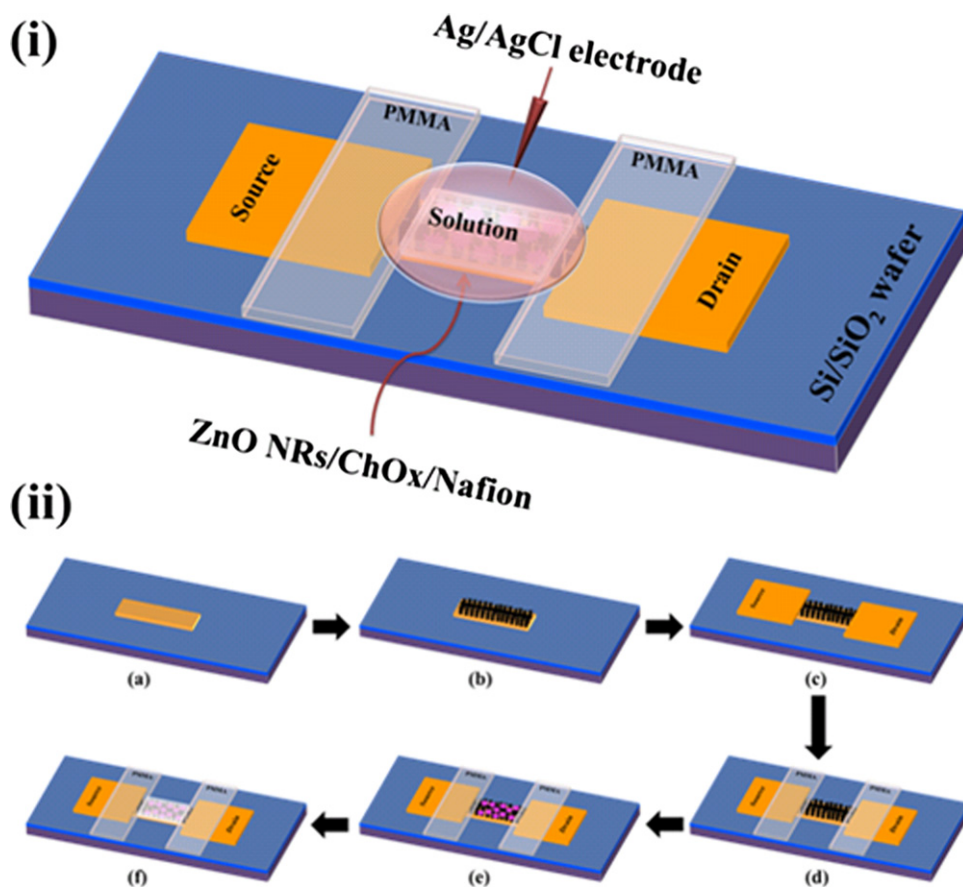


Fig. 1. (i) Schematic of FET sensor for cholesterol detection; (ii) the fabrication process of the sensor: (a) Patterning by electron-beam lithography and deposition of the ZnO seed layer; (b) growth of ZnO NRs; (c) Ag deposition by sputter; (d) PMMA coating; (e) ChOx immobilization; (f) covering by Nafion membrane.

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