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Fabrication of interdigitated high-performance zinc oxide nanowire modified electrodes for glucose sensing

R. Haarindraprasad [a], Uda Hashim [a,*], Subash C.B. Gopinath [a,b], Veeradasan Perumal [a], Wei-Wen Liu [a], S.R. Balakrishnan [a]

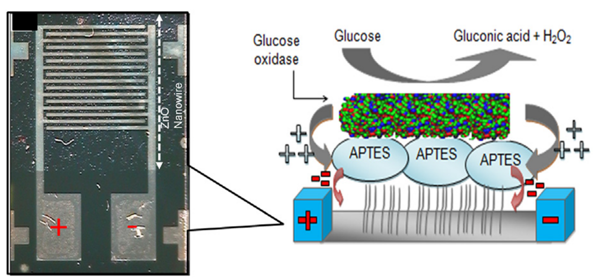
[a] Biomedical Nano Diagnostics Research Group, Institute of Nano Electronic Engineering (INEE), Universiti Malaysia Perlis (UniMAP), 01000 Kangar, Perlis, Malaysia

[b] School of Bioprocess Engineering, Universiti Malaysia Perlis (UniMAP), 02600 Arau, Perlis, Malaysia

HIGHLIGHTS

- ZnO nanowire (NW) modified Interdigitated electrode (IDE) to monitor the level of glucose is fabricated using silver.
- Amendment of ZnO-based thin film and NW were done.
- ZnO thin films that was annealed yielded a good quality crystallite sized, 50, 100 and 110 nm.
- A flower-model NW was obtained with the lowest diameter of 21 nm.
- Designed IDE was shown to have the detection limit as low as 0.03 mg/dL of glucose with a fast response time of 3 s.

GRAPHICAL ABSTRACT



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ABSTRACT

Diabetes is a metabolic disease with a prolonged elevated level of glucose in the blood leads to long-term complications and increases the chances for cardiovascular diseases. The present study describes the fabrication of a ZnO nanowire (NW)-modified interdigitated electrode (IDE) to monitor the level of blood glucose. A silver IDE was generated by wet etching-assisted conventional lithography, with a gap between adjacent electrodes of 98.80 μm. The ZnO-based thin films and NWs were amended by sol–gel and hydrothermal routes. High-quality crystalline and c-axis orientated ZnO thin films were observed by XRD analyses. The ZnO thin film was annealed for 1, 3 and 5 h, yielding a good-quality crystallite with sizes of 50, 100 and 110 nm, and the band gaps were measured as 3.26, 3.20 and 3.17 eV, respectively. Furthermore, a flower-modeled NW was obtained with the lowest diameter of 21 nm. Our designed ZnO NW-modified IDE was shown to have a detection limit as low as 0.03 mg/dL (correlation coefficient = 0.98952) of glucose with a low response time of 3 s, perform better than commercial glucose meter, suitable to instantly monitor the glucose level of diabetes patients. This study demonstrated the high performance of NW-mediated IDEs for glucose sensing as alternative to current glucose sensors.

* Corresponding author.
E-mail address: uda@unimap.edu.my (U. Hashim).

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1. Introduction

The World Health Organization (WHO) has reported that diabetes is a chronic disease, affecting 347 million people worldwide [1]. As a consequence, awareness of diabetes has become an important issue from a clinical viewpoint to prevent this metabolic disease and to have healthier lives. In addition to improved awareness, fast detection of the blood glucose level is another key factor for reducing complications with diabetes by taking medication earlier. Therefore, designing a portable and less expensive glucose biosensor has become a goal for 'bedside analysis.' Previously, the development of glucose biosensors has relied on electrochemistry [2], chemiluminescence [3], surface-enhanced Raman scattering [4], and electrochemical [5,6] and transistor sensors [7]. Enzyme-based electrochemical sensing was reported for straightforward, relatively low-cost, and more sensitive detection of glucose [8,9]. Recently, Luo et al. (2013) developed a surface acoustic wave (SAW) device for glucose sensing, which has extraordinary characteristics including higher sensitivity, higher selectivity, a fast response, and portability. However, the major drawback of the SAW device is that its fabrication is complicated, requiring the development of an alternate, simplified sensing method such as an interdigitated electrode (IDE). The IDEs generated in the past have been composed of multiple adjacent electrodes and can act as capacitive sensors [10]. Moreover, IDEs embedded in a suitable thin film facilitate better performance with surface functionalization for biosensing applications [11]. In this study, with a view to generate a novel sensing system, distinct from previous works, a zinc oxide (ZnO) thin film was embedded with an IDE and investigated for glucose sensing as a novel application.

ZnO has become a promising material among other oxide materials for its development in various applications due to its extraordinary electrical and optical properties [12–16]. Moreover, an easy and simplified route for synthesizing ZnO NWs has made it an attractive and essential material for the development of high-performance biosensors. ZnO NWs are widely considered to be a preferable option for use as transducers in biosensors due to their increased surface area to volume ratio, which is strongly associated with their elevated sensitivities [17–20]. ZnO NWs also facilitate the specific immobilization of negatively charged proteins, such as glucose oxidase (GOx), which has an isoelectric point of 4.2 (an acidic pH) [21,22]. With the appropriate material, the generation of sensors with promising sensitivity could be a hallmark for high-performance interaction analyses with a wide range of biomolecules, including small molecules [23]. Higher sensitivity with ZnO NWs has been shown through their significantly high electron mobility, which is close to 1000 cm^2/V [24]. Moreover, ZnO materials have a large binding energy (60 meV) and large band gap (3.37 eV) [25,26], which makes the materials more stable.

By applying the above favorable characteristics of ZnO, herein, we demonstrated the fabrication of a ZnO NW-modified IDE as an electrochemical glucose sensor, a novel type of sensing element. ZnO NW-modified IDE can be use in DC current (battery system) which is tailored for bedside analyses. To generate this sensor, a ZnO thin film was prepared using the sol–gel method; the film was coated on a silicon wafer, a straightforward and low-cost route of synthesis with the desired crystallite size range [13,27,28]. The construction and modification process taken to develop the glucose sensor is actually not time consuming compared to other developed glucose sensor. The newly developed sensor which involve Interdigitated electrodes (IDE) system is less complex because it only involve two working electrode system and only one mask compared to other sensor such as FET and ISFET system which involve complex fabrication steps. The fabricated ZnO NW-modified IDE electrochemical glucose biosensor is economical compared to conventional glucose sensor because conventional glucose sensor involve gold pad electrode which is very costly. The fabrication process of the ZnO NW-modified IDE glucose sensor is less complex and also reduces the production cost. Studies shows that zinc oxide material is considered economical globally. The proposed Interdigitated Zinc Oxide Nanowire modified electrode (IDE) sensor contains lot of advantages compared conventional glucose meter, such as the dimensional size of the sensor, simplicity, reliability and economical. We demonstrated the detection of glucose at lower levels, which is highly suitable for diabetes patients, can be feasible for and conducive to glucose monitoring, and has practical and commercial potential.

2. Experimental

2.1. Consumable materials

The ZnO seed solution was prepared using zinc acetate dehydrate [$Zn(CH3COO)_2.2H_2O$] (98%; Sigma–Aldrich, St. Louis, Missouri, USA), which was free of chlorine ions and used as a precursor. Solvents such as 2 methoxyethanol, 2-ME (99.8%; Sigma–Aldrich, J.T. Baker, Center Valley, Pennsylvania, USA) were used without further purification. Monoethanolamine (MEA; 99%; Merck, Kenilworth, New Jersey, USA) was used as a stabilizer. Buffered oxide etchant (BOE; 6:1; J.T. Baker, Center Valley, Pennsylvania, USA), negative photoresist (NR7-6000PY; Futurrex, Franklin, New Jersey, USA) and resist developer (RD6; Futurrex, Franklin, New Jersey, USA) were used for the photolithography. The growth solution was prepared by mixing equal concentrations (25 mM) of zinc nitrate hexahydrate (99%; Sigma Aldrich, St. Louis, Missouri, USA) and hexamethylenetetramine (99%; Merck, Kenilworth, New Jersey, USA) in DI water. (3-Aminopropyl) triethoxysilane, 99% (APTES) and glucose oxidase (GOx) G3660-1CAP were purchased from Sigma Aldrich Sdn. Bhd (Malaysia) and were used for immobilization process. The glucose sample, dextrose monohydrate (DEX) $C_6H_{12}O_6$, H_2O (100%, Malaysia), was purchased from a pharmacy.

2.2. Sample wafer preparation

Silicon p-type samples with an orientation of <100> were cleaned by immersing them in RCA 1 and RCA 2 solutions to eliminate unwanted substances. Next, samples were immersed in BOE solution to remove contaminants such as oxide layers and to improve the hydrophobic character of the samples. Removing the oxide layer is a necessary step in sample preparation to ensure that the ZnO thin film would be coated properly with minimal defects and in a uniform orientation.

2.3. Fabrication of interdigitated electrodes (IDE)

Silver IDE electrode was deposited on the silicon wafer sample with an orientation of <100>, using the traditional wet etching method. Positive photoresist (PR) was coated on the silicon wafer, followed by a soft bake for 90 s. Ultraviolet (UV) light exposure was conducted for 10 s to allow the pattern to be transferred from the IDE mask onto the sample. Next, the sample was developed for 15 s using the RD-6 developer. Then, sample was baked at 110 °C to remove unwanted moisture and to enhance the adhesion between the silver and SiO_2 layers. Finally, the unexposed area was removed using a silver etchant for 23 s and then cleaned with acetone. The time consume for fabricate the IDE sensor is approximately 180 s only. The fabrication process of the IDE electrode which involves photolithography process was conducted at room temperature (23 °C).