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Hypothetical multi-detection ultra-sensitive sensors using ZnO thin films and metamaterials: A review

Yan Soerbakti

Department of Physics, Universitas Riau, Pekanbaru 28293, Indonesia

ABSTRACT

This study aims to observe the ability of a thin layer of zinc oxide (ZnO) which is applied to sensor technology. Currently, the low detection limit of the sensor is a problem in its use. Metamaterials offer resonant properties in increasing sensitivity, but their performance is still below the current high modern technology. The high engineering properties of metamaterials provide opportunities for realizing renewable metamaterials. ZnO thin layer semiconductor material as a transparent conductive oxide can provide a wide detection potential. The ability of ZnO thin films to be adapted to metamaterial sensors can be further investigated and improved for the future.

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* **Corresponding Author**
E-mail address: yansoerbakti2@gmail.com

1. INTRODUCTION

The development of the modern digital era makes the role of scientists increasingly needed in analyzing and creating advanced technology with high performance quality. The technology currently being developed is a metamaterial-based sensor device with achievements that already cover the fields of industry [1], health [2], agriculture [3], and marine [4]. Metamaterials are artificial materials with unique characteristics and high sensitivity. The use of metamaterials as a breakthrough in advanced technology has very high and broad potential because the materials and structures are renewable [5-7]. Pure metamaterial structures such as split ring resonators (SRR) have been reported by Saktioto et al., where the implementation of hexagonal SRR metamaterials with FR-4 substrates can improve the profile of low microstrip antennas with a performance frequency of 8 GHz [8]. SRR metamaterials in their current applications are still below the needs of modern technology with high quality. This is a problem that needs to be studied to improvise metamaterials into superior materials. The high engineering properties of metamaterials [9] can realize hybrid metamaterials from a combination of other materials such as semiconductor materials as transparent conductive oxide to expand the conductive area of the sensor to be more sensitive [10, 11]. Previous research reported by Tao et al., explained that the antenna sensor was successfully carried out with a metamaterial metal structure on a substrate consisting of a layer of carbon fiber, polyimide (PI), and a thin layer of zinc oxide (ZnO) with a power loss of -65 dB [12]. ZnO is an n-type semiconductor material whose characteristics depend on the formation of a crystalline structure [13, 14]. In general, a thin layer of ZnO can be formed using various vapor physics deposition methods with various techniques [15-17]. The deposition of a thin layer of ZnO requires an annealing process at a temperature greater than 400 °C to stabilize the crystalline structure formed on the substrate [18]. Corning glass is the best substrate used as a medium for growing ZnO thin layers because it has heat resistance up to 800 °C, dimensional stability, and high manufacturing [19]. Previous research by Hong et al., explained the growth of a thin layer of indium zinc tin oxide on a corning glass substrate as an antenna application resulting in 80% transmittance and 7.76% efficiency [20].

2. METAMATERIAL

Metamaterial is a combination of two words from meta (μετα in Greek) which means "outside" and material. So metamaterial can be interpreted as a material that is outside its true natural properties [21, 22]. This is due to changes in the optical physical properties of the permittivity and permeability materials which have negative values [23]. The study of metamaterials began with the hypothesis of Victor G. Veselago (1968) which stated that a negative refractive index of material occurs if there is a negative permittivity and permeability, so that the propagation properties of electromagnetic (EM) waves become the opposite of normal materials [24, 25].

The general concept used by researchers for the design of EM metamaterials is actually built by several natural materials, but by modifying their structure, they can produce new properties when interacting with EM waves and not from their constituent elements [5, 26]. Metamaterials are artificial or engineered materials that are periodically composed of several individual metals [27]. Metamaterials have structural characteristics with dimensions smaller than the given EM wavelength [28]. Currently, metamaterials are not only limited to engineering EM properties, but have expanded to all types of waves. Engineering the properties of EM and acoustic waves is the most widely developed by scientists [29, 30]. Therefore, metamaterials can be expanded by controlling the properties of waves through engineering the structure of the material, where the properties of the waves do not occur naturally [31].

3. ZnO THIN LAYER

ZnO is a metal and oxide component material from groups II and IV which is also a semiconductor material with characteristics in the form of a gap energy of 3.37 eV and an electron excitation binding energy of 60 MeV [32, 33]. There are 3 types of ZnO crystal structures, namely zinc blende, wurtzite, and rock salt as shown in Figure 1.

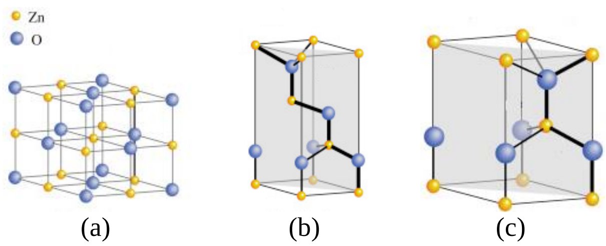

Figure 1. ZnO crystal structure: (a) rocksalt; (b) zinc blende; and (c) wurtzite [34].

According to its stability level, the wurtzite structure is better than the other structures. At room temperature, ZnO with a wurtzite structure has a hexagonal shape [35, 36]. ZnO material is a crystal that is widely used in various applications, including as a semiconductor, conductor and catalyst [37]. The characteristics of ZnO material are influenced by its size and growth method [13]. Thin ZnO layers are formed from the deposition process of ZnO material with nano-scale dimensions grown on substrates with several methods such as sol-gel [38], spray pyrolysis [39], and magnetron sputtering [40]. ZnO crystals also have excellent electronic and optical properties, and are widely applied as sensor materials, photocatalysts, and solar cells [41]. During the deposition process, a substrate that is resistant to high temperatures above 400°C is required, namely corning glass for the annealing process so that the formed ZnO thin layer becomes homogeneous [18]. Corning glass is an ideal type of glass for semiconductors and other electronic industries because of its high dimensional stability and pure clarity [42, 43]. Corning glass is very resistant to heat (> 800 °C), chemicals, and mechanical stress [19]. Its flexible properties can be fabricated as an antenna sensor [20]. The high melting point of corning glass makes it a good choice for semiconductor components. Corning glass is made from silicon with low production cost, very thin, and versatile [44].

4. ZNO-BASED METAMATERIAL-BASED SENSOR APPLICATIONS: A REVIEW

4.1. Acoustic Wave Surface Sensor

This sensor model explores a new concept of integrated sensing technology on carbon fiber through the integration of EM metamaterials and thin-film acoustic wave sensors, with the ability of noninvasive, in situ, and continuous monitoring of environmental parameters and biomolecules wirelessly. The sensor material uses a three-layer composite design. The carbon fiber is first coated with a PI layer, and then a ZnO film is deposited onto the PI/carbon fiber structure as shown in Figure 2 (a). Then fabricate the acoustic wave surface device and metamaterial on this composite material using conventional photolithography method and optimize the electrode design for integrated functions including liquid temperature control, UV sensing, and glucose monitoring as case studies for different applications as shown in Figure 2 (b).

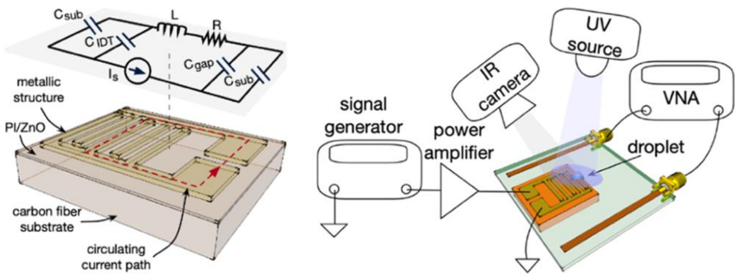

Figure 2. Illustration of (a) sensor structure design and (b) experimental [12].

ZnO thin films (5 µm thick) were deposited on PI-coated carbon fiber substrates using DC magnetron sputtering with a sputtering power of 400 W, an Ar/O_2 gas flow rate of 10/15 sccm, and a chamber pressure of 4×10^{-4} mbar. A zinc target with a purity of 99.99% was used, while the sample holder was rotated during deposition to achieve uniform film thickness. The interdigitation transducer was patterned using conventional photolithography and lift-off processes, in which Cr/Au films with a thickness of 10 nm/120 nm were selected as electrode materials and deposited using a thermal evaporator (EDWARDS AUTO306) [12].

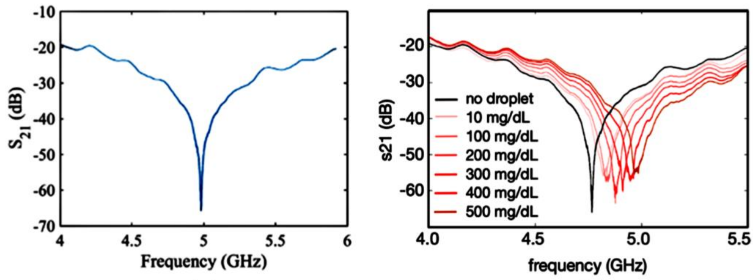

Figure 3. Performance parameters of (a) sensor transmittance and (b) glucose test transmittance [12].

The EM resonance of the device with a wavelength of 64 µm was also characterized, and the resulting S21 transmission spectrum is shown in Figure 3 (a). The EM resonance frequency was measured as 4.98 GHz. In this design, the wavelength of the interdigitation transducer does not change the resonance frequency because the effective capacitance parameter is dominated by the surface capacitance of the structure. Figure 3 (b) shows the S21 spectra recorded at different glucose concentrations. The resonance frequency of the metamaterial device increases with glucose concentration. This is expected because the permittivity of the glucose solution droplet decreases with increasing glucose concentration [45]. We observed a linear decrease in the resonance frequency in the measurement range with a sensitivity of 0.34 MHz/(mg/dL) [12].

4.2. Ultraviolet Detection Resonator Sensor

The use of planar microwave resonators in previous studies has been carried out to measure time-resolved microwave conductivity (TMRC) in TiO_2 nanotube arrays [46-48]. The TRMC measurement involves the placement of a ZnO membrane or film to be characterized directly deposited on a conducting copper (Cu) strip. The planar microwave ring resonator was designed and simulated in HFSS software. A thin ZnO semiconductor layer is electrodeposited that is aligned on the top microstrip line. The close contact of the ZnO layer with the copper microstrip line forms a Schottky contact with a space charge region at equilibrium, the width of which depends on the carrier concentration in ZnO, the relative permittivity, and the inherent potential. The simulated and experimental microstrip ring resonator structures are presented in Figure 4 (a). While Figure 4 (b) shows the resonator test setup with a ZnO thin film placed inside a dark box to isolate and suppress the effects of light at room temperature of 23°C with a relative humidity of 25% [49].

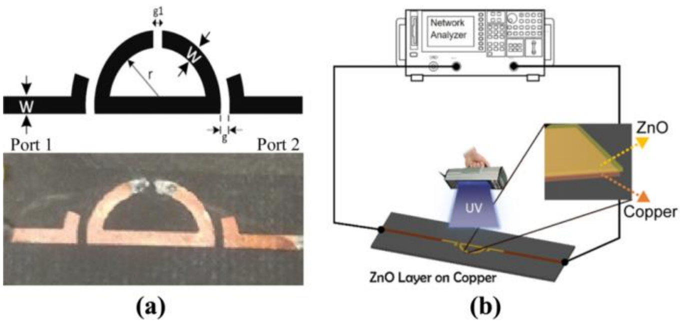

Figure 4. Design of (a) ring resonator and (b) experimental measurement [49].

Figure 5 presents the implemented structure and simulation results for the S21 resonance profile for varying electrical parameters (dielectric constant, loss tangent)of ZnO thin film. The dielectric constant of ZnO thin film is assumed to be 8.9 [50]. First, Figure 5 (a) shows that the resonant frequency and resonant amplitude are significantly affected by the presence of electrodeposited ZnO film. ZnO film creates an excess region of higher dielectric constant and confined conductivity around the resonator, which results in this change in the transmitted microwave energy. The experimental S21 parameters of the resonator, before and after electrodeposition of ZnO film, are presented in Figure 5 (b). It is observed that the resulting change in the effective permittivity of the microwave resonator causes a decrease in the resonant frequency and a decrease in the resonant amplitude [49].

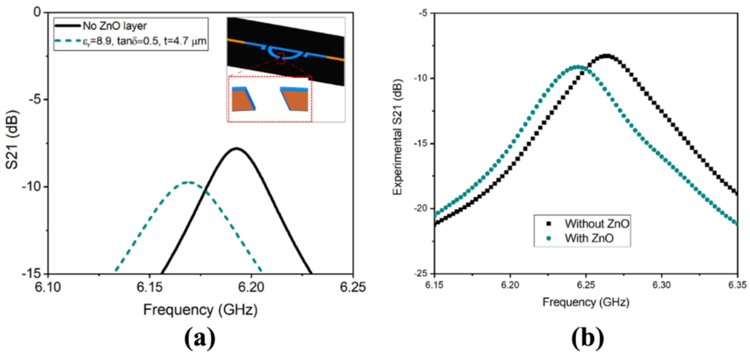

Figure 5. Transmission spectrum parameters of (a) simulation and (b) experimental results [49].

4.3. Nano-Textured Surface Antigen Sensor

The formation of nano-textured surfaces was reported by Assaifan et al. (2016) on flexographic printed ZnO thin films that provide an excellent platform for low-cost and highly sensitive biosensor applications. Zinc oxide thin films were formed by printing zinc acetate precursor ink solution and annealing at 300°C. An intricate nanotexturing of the film surface was achieved through a 150°C drying process. These surface nanostructures were found to be in the range of 100 to 700 nm in length with a width of 58 ± 18 nm and a height between 20 and 60 nm. The design of the sensor structure is shown in Figure 6 [51]. The structure as shown in Figure 6 (a) significantly increases the surface area to volume ratio of biosensing materials which is important for high-sensitivity disease detection. Illustrations of the biosensor functionalization steps are shown in Figure 6 (b-d). Silane groups of (3-Aminopropyl) triethoxysilane APTES are covalently bonded to the native hydroxyl groups of the ZnO layer as shown in Figure 6 (b). Next, the aldehyde group of glutaraldehyde is covalently bonded to the exposed amine group of the APTES layer as shown in Figure 6 (c). The amine group of the antibody then bonds to the exposed aldehyde group of glutaraldehyde as shown in Figure 6 (d).

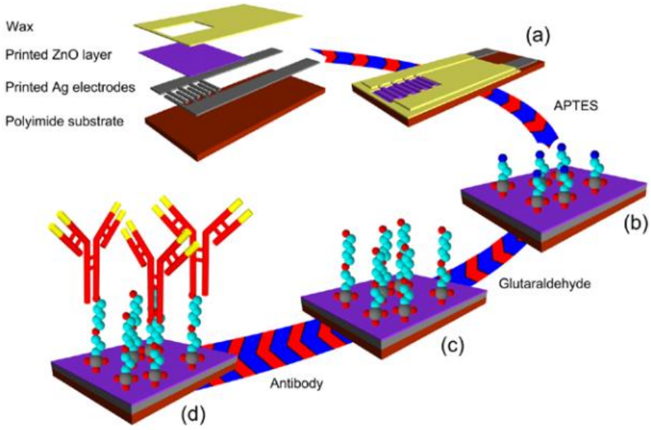

Figure 6. Schematic of human antigen sensor and test design [51].

Non-faradaic electrochemical impedance spectroscopy measurements were performed to detect the human cytomegalovirus pp65 antigen using a printed device, which has a low detection limit of 5 pg/ml. The response of the biosensor at 50 mHz to increasing concentrations of pp65 antigen was performed in triplicate for both positive and negative samples as shown in Figure 7. The sensor exhibits signals within the standard deviation. An example plot for the sensor tested in a positive sample, over the full test frequency range, is shown in Figure 7 (a). This plot shows that the impedance is stable while the phase shifts as the device is incubated in increasing concentrations of pp65 antigen. Figure 7 (b) in the Information shows that the capacitance of the device decreases with increasing concentrations of pp65-antigen [51].

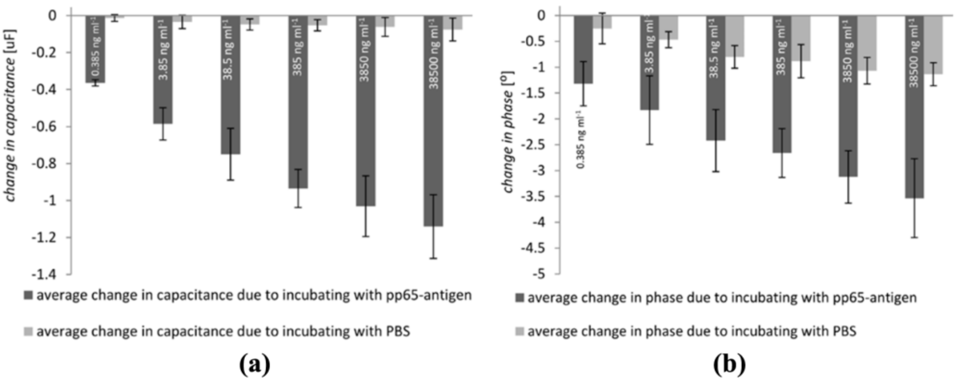

Figure 7. Biosensor response (a) capacitance change and (b) phase change [51].

5. CONCLUSION

A study on the application of ZnO thin films in metamaterial sensor applications has been reported. ZnO thin films have been proven to be versatile in enhancing the performance of metamaterial structure-based sensors. The detection space of metamaterial sensors is further enlarged by the conductive properties of ZnO thin films. Further studies should be continued to explore the potential of ZnO thin films and for the improvement of metamaterial sensor technology.

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