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Modification of Surface Acoustic Wave (SAW) Sensor Using Zinc Oxide Nanowires

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ABSTRACT

The limitation and complexity of current available biosensor to detect nano-scale organism has introduce an idea to develop simple, high sensitivity and performance biosensor. Surface acoustic wave (SAW) device which widely used in telecommunication field had been remodeled as biosensor which able to detect the microorganism in aqueous state. Zinc oxide (ZnO) nanowires with high crystalline structure were prepared on the sensing area of SAW device through a low-cost sol-gel method coupled with a hydrothermal process. The ZnO nanowires were proposed as an effective strategy to enhance the shift frequency of the sensing device. Lift-off technique was used to develop the aluminum IDTs structure on the piezoelectric substrate. X-ray diffraction (XRD) and Scanning electron microscope (SEM) were used to study the crystal lattice orientation and physical properties of ZnO nanowires. The electrical test and frequency response were conducted to characterize the functionality of the fabricated device.

Keywords: Nanowires, Sol-Gel Method, Surface Acoustic Wave, Zinc Oxide.

I. INTRODUCTION

Surface Acoustic Wave (SAW) device has been use to develop as sensor for many applications. The evolution of SAW device is from communication field to biological field has leads to development of SAW biosensor. Compare to others biosensor, SAW biosensor provide better sensitivity, small in size, design flexible, high reliable and can operate in wide dynamic range [1, 2]. SAW biosensor enable the sensing for low electrochemical signal and convert it to electrical signal by voltage or current [3, 4]. A basic SAW device consists of a pair of Interdigital Transducers (IDT)s which fabricated on the surface of single crystal or piezoelectric substrate [5] as illustrated in Figure 1. The IDT function is to convert the signal from electrical to mechanical and vice versa. The mechanical wave is then traverse on the sensing area of the sensor. The change of the physical phenomenon on the sensing area will modify the mechanical signal which propagating along the sensing area [5, 6]. After that, the IDTs receive the deflected mechanical wave and transduce back to electrical signal as the output for the sensor. The difference between the input and output electrical signal indicate the physical experience on the sensing area of the sensor.

The difference in frequency, amplitude, delay time and phase characteristic between the input and output electrical signal are important characteristics to study performance device. In order to design a SAW device, impulse response (IR) model is used to calculated frequency response [7].

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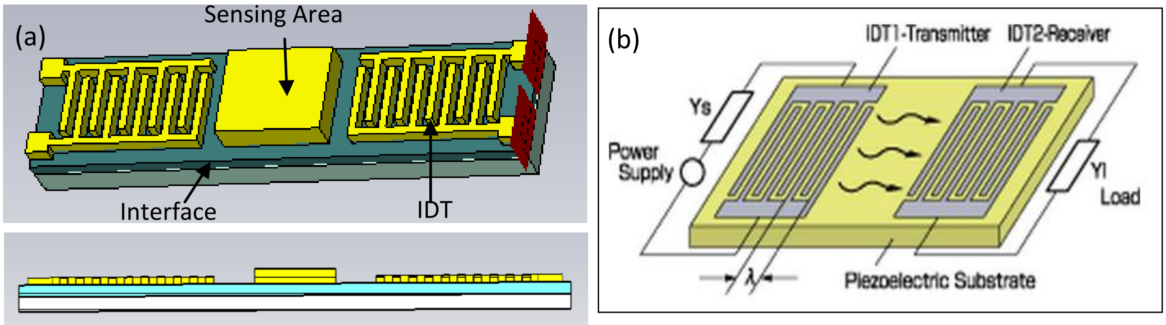

Figure 1. (a) A three-dimensional view and cross-section image of the fabricated SAW device which consist of piezoelectric substrates interface, a couple of IDT transducer and sensing area. (b) Schematic showing the principle of the SAW device as a sensor.

From the IR model, the important design parameters such as number of IDT finger (*N*), finger width and spacing between them (*W*), and wavelength (λ), must be calculates [8]. Central frequency can be obtain using equation 1.

$$f_o = \frac{V_{saw}}{\lambda} \tag{1}$$

Where (f_o) is central frequency, V_{saw} an acoustic velocity of piezoelectric substrate and λ is the wavelength. Higher frequency will increase the sensitivity of SAW sensor. Important parameter in designing a high sensitive and low loss SAW device will be affected significantly by capacitance [9]. The For instance, the electromechanical coupling factor $k^2$of the SAW device will be affected by the total capacitance and the number of IDT fingers [10]:

$$f_o = \frac{k^2 2N}{C_T R_a \pi^2} \tag{2}$$

Where R_a is the radiation resistance, C_T is the total capacitance, N is IDT finger numbers and f_0 is the center frequency. From equation 2, the electromechanical coupling factor is proportional to the total capacitance and center frequency. k^2 is the electromechanical coupling factor which is an indicator of the effectiveness with which a piezoelectric material converts electrical energy into mechanical energy. SAW device with more IDT fingers in addition to high acoustic aperture yield high capacitance [10, 11]. Thus, this produce higher electromechanical coupling factor.

Conventional SAW sensor using aluminum, gold as a sensing area film. Several limitations have been found by using this technique [12, 13, 14]. For latest technology SAW device, a sensing thin film has been replaced by using metal oxide films such as (TiO_2), ZnO and SnO_3, TeO_2other layer metal oxide [12,13]. ZnO is n-type semiconductor materials and used for several sensor devices considered its sensitivity and easy doping higher electromechanical coupling factor method to improve sensing performance. It is characterized with its direct wide band gap of 3.37 eV at temperature room, high excitation binding energy of 60 meV [13]. There are several advanced techniques can be implemented to form a ZnO thin film on a variety of substrates such as laser ablation, E-beam, sputtering, sol-gel methods, thermal deposition, and metal organic chemical vapour deposition [15,16].

To improve the sensitivity of the devices the conventional thin film sensing area was replace by ZnO nanowires. Thus, we report herein the SAW sensor based on ZnO piezoelectric thin film using low cost sol-gel method as a substrate with ZnO nanowires as the performance-enhancement sensing layer. It is believed that, the SAW sensors with ZnO nanowires as sensing area element utilizes high sensitivity of frequency response compare to existing SAW devices [17, 18]. For this paper, a SAW device has been fabricated by using lift-off technique with compare aluminum thin film and ZnO nanowire as a sensing area. Then the frequency response result has been compared using capacitance and network analyzer technique.

2. EXPERIMENTAL

2.1 Samples Preparation

The 4-inch (100 mm in diameter) p-type silicon wafer was used as the starting material in this work. Prior to cleaning process, the wafers are then cut in the dimension of 3.0 cm x 1.5 cm. Standard cleaning procedure using RCA1, BOE, and RCA 2 was introduced to remove organic and inorganic contaminant on the sample's surface followed by washing in de-ionized water for the last steps. Then, the samples were dried on a conduction hot plate at 200°C for 5 minutes to remove residual water and cooled down in room temperature for 10 minutes. The cleaned samples were then deposited with a thin layer silicon oxide (SiO_2) with the thickness of 200 Å by dry oxidation process as illustrated in Figure 2 (step 1).The ZnO solution was prepared by mixing zinc acetate dihydrate [Zn $(CH_3COO)_2{\cdot}2H_2O$] and dehydrated 2-Methoxyethanol (2-ME). Monoethanolamine (MEA) serves as stabilizer which was added slowly to the mixture. To ensure the zinc powder was completely dissolved in the 2-ME solvent, the magnetic stirrer was used to stir the mixture at constant temperature about 60°C until the mixture become transparent. The mixture was then aging for 24 hours before use [19].

2.2 Device Fabrication

Piezo electric materials were used to improve the performance of the substrate rather than the silicon dioxide surface. Therefore, ZnO solution prepared by sol-gel method was introduced on the wafer to become piezoelectric substrate. For this purpose, the ZnO solution was dropped on the samples and coated by using spin coating technique which rotated at 2000 rpm for 30 seconds. The deposited films were then heated at 150˚C for 10 minutes on a hot-plate to evaporate the solvent and remove the organic residuals. Finally, the annealing process at 500˚C for 2 hour using Muffle furnace was employed to ensure that all organic species were expelled from the films for better surface uniformity as depicted in Figure 2 (step 2).

Figure 2 (step 3) illustrates the general steps of conventional photolithography and lift off process which are implemented in this work in order to develop the complete pattern of Interdigital Transducer (IDT) on the samples. The process started with the sample coating using positive resist by spin coating technique at 3000 rpm, and pre-baked on the hot plate at 90º C for 90 seconds. The pattern was transferred onto the resist coated samples through UV exposure during photolithography (MIDAS MDA-400M Mask Aligner) process. The exposed samples were developed in the RD-6 developer, and then post baked on the hot plate at 90º C for 90 seconds. Visual inspection was conduct to ensure the pattern was appropriately developed on the photo resist layer. The developed resist acts as the hard mask for the subsequent process. The aluminum layer was deposited on the sample surface using physical vapor deposition (PVD) and followed by immersed in acetone to remove the unwanted pattern in this lift off process.

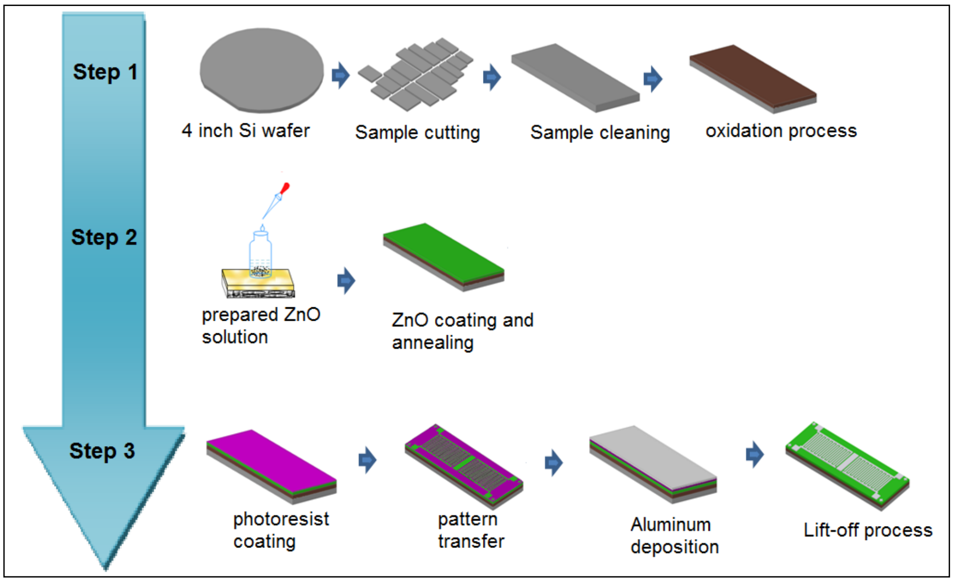

Figure 2. A schematic diagram of procedure used to develop SAW sensor integrated with ZnO layer.

2.3 Growth ZnO Nanowire as Sensing Area

Sensing area formation involves two main process steps which are preparation of ZnO solution and sample preparation. The ZnO solution was prepared by dissolve certain amount of hexamethylenetetramine (HMTA) powder into zinc nitride hexahydrate ($Zn(NO_3)2$) solution and DI water. The mixture was stirred in ambient temperature for 20 minutes by using magnetic stirrer. Next, for sample preparation, a hard mask using positive photo resist was introduced followed by photolithography steps to create an opening region on the sensing area as depicted in Figure 3 (a). A layer of ZnO solution was then coated onto the sample using spin coating process. After lift-off process, the unwanted photo resist layer was removed and ZnO seed layer are remained at the sensing area of SAW device as shown in Figure 3 (b). Then the sample was immersed into the prepared chemical solution. The top surface of the sample was facing down in the solution. The solution heated to 93°C for 5 hours. The ZnO nanowires were start to growth at this stage.

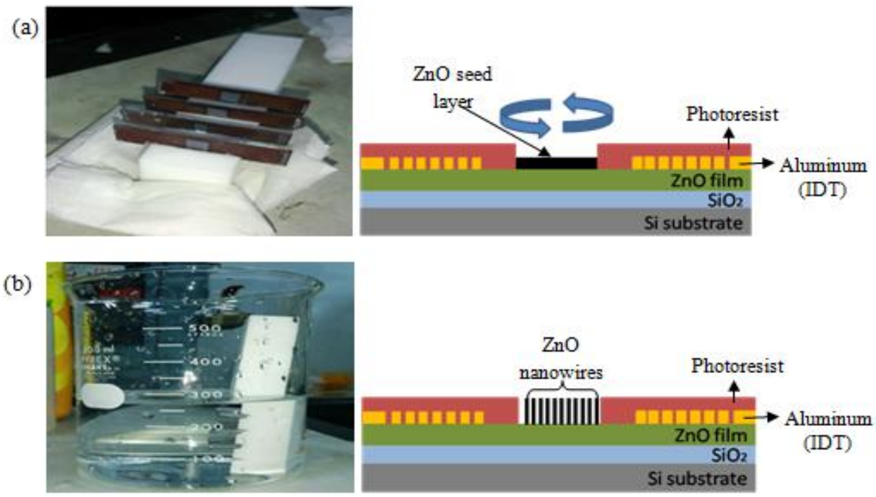

Figure 3. (a) Coated ZnO layer on the opening area of SAW sensing device. (b) the growth process of ZnO nanowires at the desired area.

2.4 Characterization

The ZnO nanowires was characterized by using PAN analytical X'Pert Pro X-ray diffractometer with Cu Kα radiation at λ = 1.5406. The x-ray diffraction (XRD) pattern was recorded in the range of 20° to 60° operating at 45 kV and 20 mA. Morphological studies were performed using scanning electron microscopy (SEM, JEOL, JSM6460LA) and Atomic Force Microscopy (AFM) SII Sciko Instrument INC SPI3800N Probe station. The electrical property of the developed sensor was characterized using the frequency response measurement technique. Agilent E5062A network analyzer was carried out to measure the frequency response of the devices. The magnitude and the maximum amplitude of the frequency can be obtained from the graph. The center frequency and insertion loss can be evaluated as well. For this work, the measurement for the fabricated SAW devices was used in the range of 300 KHz to 3 GHz.

3. RESULTS AND DISCUSSION

3.1 Characterization of Zinc Oxide (ZnO) Thin Film

The prepared ZnO solution by sol-gel method was coated on the samples using spin coating technique in order to develop the SAW sensor based on ZnO piezoelectric thin film as a substrate. The thickness of the ZnO film was measured using spectrophotometer and the corresponded results are tabulated in Table 1. The average thickness of ZnO film is 5161.6 Å with good uniformity of coated layer which is 96.06%.

Table 1 Thickness of ZnO layer

Measured point	Thickness (Å)
1	4976.5
2	5128.7
3	5143.0
4	5177.0
5	5383.0
Average thickness	**5161.6**
Non-uniformity (%)	**3.94**

The surface morphology of ZnO thin film before and after annealing process was studied using AFM as shown in Figure 4 (a). The results showed that the annealing process employed during the development of ZnO film was strongly influenced the properties of the materials. After annealing process at 500 °C for 2 hours, the topography of ZnO film changes from columnar to small spherical-like structure. The results are in agreement with the previous work reported [20], demonstrated that the annealing effect the structure of ZnO synthesized by sol-gel method to form nanostructure ZnO thin film. SEM analysis has been done to clarify this founding as depicted in Figure 4 (b). Besides, Cular. S and group had claimed that, 450 °C is the sufficient annealing temperature to improve the structural and properties of ZnO film [21]. However, due to the thick layer of ZnO produced by 25 layers coated on the samples that have been used in this work, 500 °C was found to be the optimum annealing temperature.

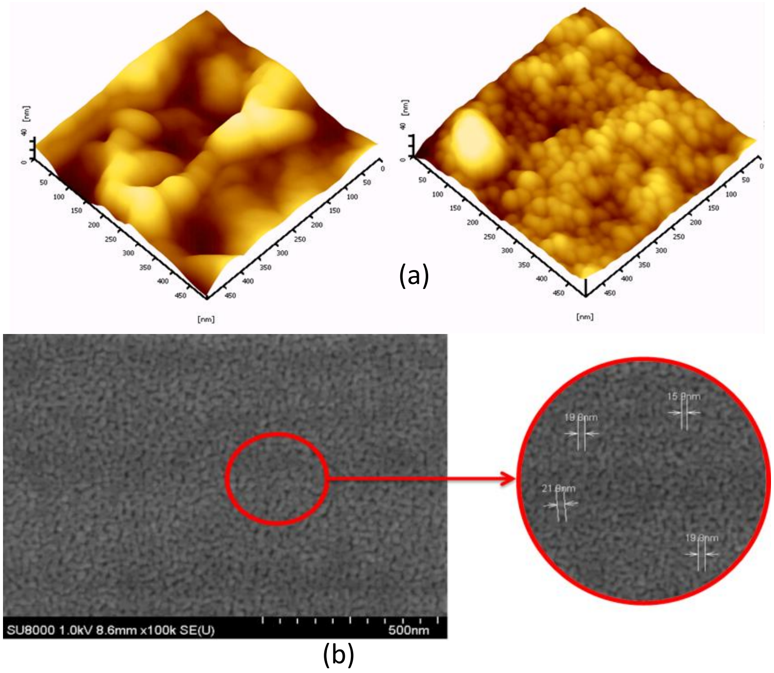

Figure 4. (a) Three-dimensional AFM images showing the differentiate between ZnO layer before and after annealing process. (b) SEM image of the thin film ZnO layer.

Morphological inspection gives a qualitative picture of the effect of annealing surface treatment, whereas mean roughness values, Rms, provide a quantitative parameter of surface structure. Based on the three-dimensional AFM images, it could be noticed that the surface roughness decreases after annealing process with the Rms value of 9.69 nm and 4.55 nm, respectively. In addition, the grain size of the film also decreases as the annealing process was employed on the sample surface. In reference [14], Chou and co-workers has claims that, smooth surface morphology, sharp interface and perfect c-axis texture of piezoelectric film can achieve good SAW performance. Therefore, the reduction in surface roughness after annealing process can enhance the performance of piezoelectric substrate.

3.2 Characterization of Zinc Oxide (ZnO) Nanowire

Vertically aligned ZnO nanowires are grown on the active sensor surface of SAW sensor via immersed the sample into a mixing solution of zinc nitrate hexahydrate and hexamethylenetetramine (HMT) in order to provide zinc ions and a stabilized release of OH^- ions during the growth process at 93 °C. The process as a whole involves the following reactions (1), (2) and (3):

$$C_6H_{12}N_4 + 6H_2O \rightarrow 6HCH) + 4NHH_3 \quad (1)$$

$$NH_3 + H_2O \leftrightarrow NH_4 + OH \quad (2)$$

$$Zn^{2+} + 2OH^- \leftrightarrow ZNO + H_2O \quad (3)$$

In addition to the standard reactions of growth ZnO nanowires, the oxidizing agents used of the solvents may affect the aluminum layer of electrode and release contamination of aluminum element in the solvent. Prior to the chemical solution process, we proposed the used of positive photo resist layer to cover the metal IDTs structures (as depicted in Figure 3) and the nanowires were grown directly on the sensor active surface. Figure 5 (a) shows the camera view of the final fabricated SAW sensor after removed the passivation layer (positive photo resist) using acetone. The input and output interdigital transducer (IDTs) consisted of 10 finger pairs with an electrode width and length of 0.3 mm and 4.5 mm, respectively and attached with the contact pad size of 2.0 mm x 2.00 mm. The IDTs had an acoustic aperture of 4.2 mm.

After the analysis of ZnO nanowires, found eye-detectable of new existence species on parts of SAW device sensing area with size of 5.00 mm x 3.00 mm. For further confirmation, SEM analysis has been done to verify the formation of nanowires. As depicted in Figure 5 (b), ZnO nanowires only grow on the desired area of the devices, which proves our method by coated positive photo resist as passivation layer potentially providing a convenient approach for integration of high-density nanostructures into pre-existing CMOS devices or any substrates, without introduced the use of any complex steps or chemicals. While, Figure 5(c) shows the top-view image of ZnO nanowires growth on the substrate. Figure 5(d) indicates the surface morphology of the nanowire structures, examined using AFM. Based on the three-dimensional images, the nanowires had a cone structures which means that the top diameter of the nanowire is getting smaller as compared to the bottom part of the structures. This preferential growth of ZnO nanowires is associated with the ZnO seed layers. The thickness of ZnO seed layer strongly affects the crystallinity of ZnO structure grown from aqueous solution. Moreover, Gronewold and co-worker had reported that thickness of the seed layer can dramatically [22] affects the dimension of ZnO nanostructures growth by using aqueous solution technique and rather than the growth time. Therefore, the formation of ZnO nanowires in this work is expected to have bigger diameter since there was coated with 25 ZnO seed layers. This observation was confirmed by a zoom-in image of the structures with magnification of 20000X as depicted in Figure 5(b). The ZnO nanowires grow almost perpendicularly onto the substrate and their average length is approximately 1.0 µm with diameter in the range of 100 nm to 500 nm.

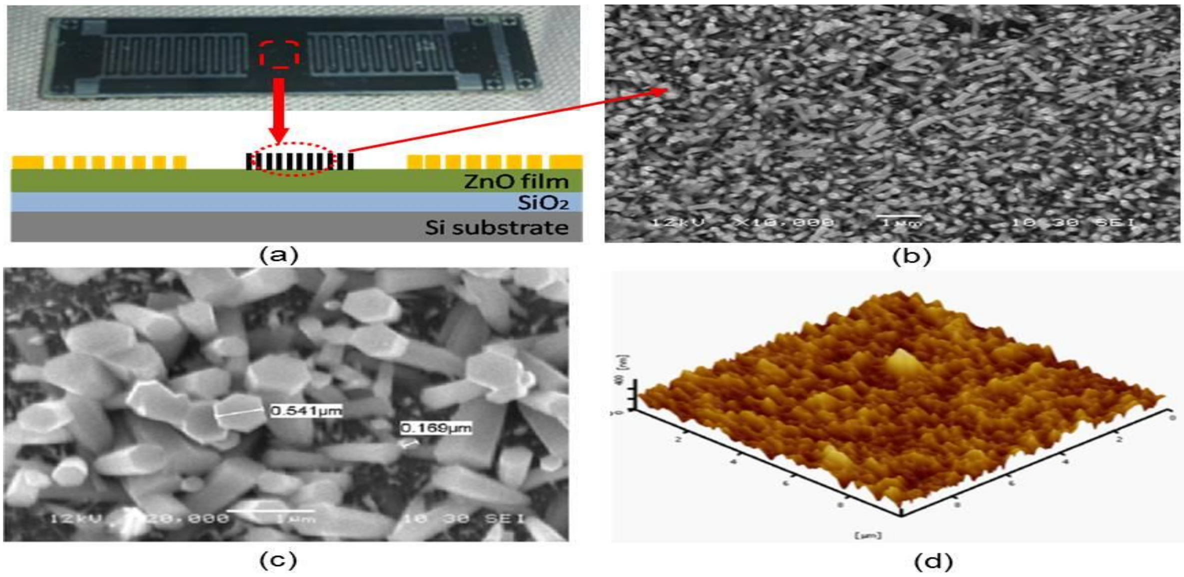

Figure 5. (a) A camera view of final fabricated devices with cross-section image presented a sensor layout (b) top view of synthesized ZnO nanowires observed by typical SEM images on SAW Sensing Area, (c) zoom-in image of vertically growth nanowires with width approximately in the range of 100nm to 500nm (d) 3-D view of AFM images ZnO Nanowire.

3.3 X-ray Diffraction (XRD) Characterization

The crystal structure of the synthesis ZnO thin film and ZnO nanowires were studied using XRD analysis. The blue line color indicates the XRD pattern for ZnO thin film, while the red line color represents the pattern for ZnO nanowires as shown in Figure 6. The peak intensities were measured in the range of 30° to 45° at 2θ. A narrow and sharp diffraction peak can be observed for the high crystalline structure. A strong diffraction peaks were obtained between 33.8° 35.5°indicates the prepared ZnO film and ZnO nanowires have preferential crystal growth at the

(002) plane [16]. The result was in agreement with the data base of the EVA program system based on hexagonal lattice of ZnO, [ZnO: PDF 00-036-1451]. The stronger intensity of the ZnO reflection peaks may reflect the ZnO nanowires having been indexed to a hexagonal structure with high purity phase. This finding showed that the ZnO materials consist of pure hexagonal wurtzite structures with high *c*-axis orientations. Other refraction peaks of ZnO such as (100), (101), (102) and (110) were observed in the XRD result when the measurements 2θ vary from angle 30° to 70° as depicted in Figure 6 (a) and Figure 6 (b). The results showed that no other impurities diffraction peak found in the samples.

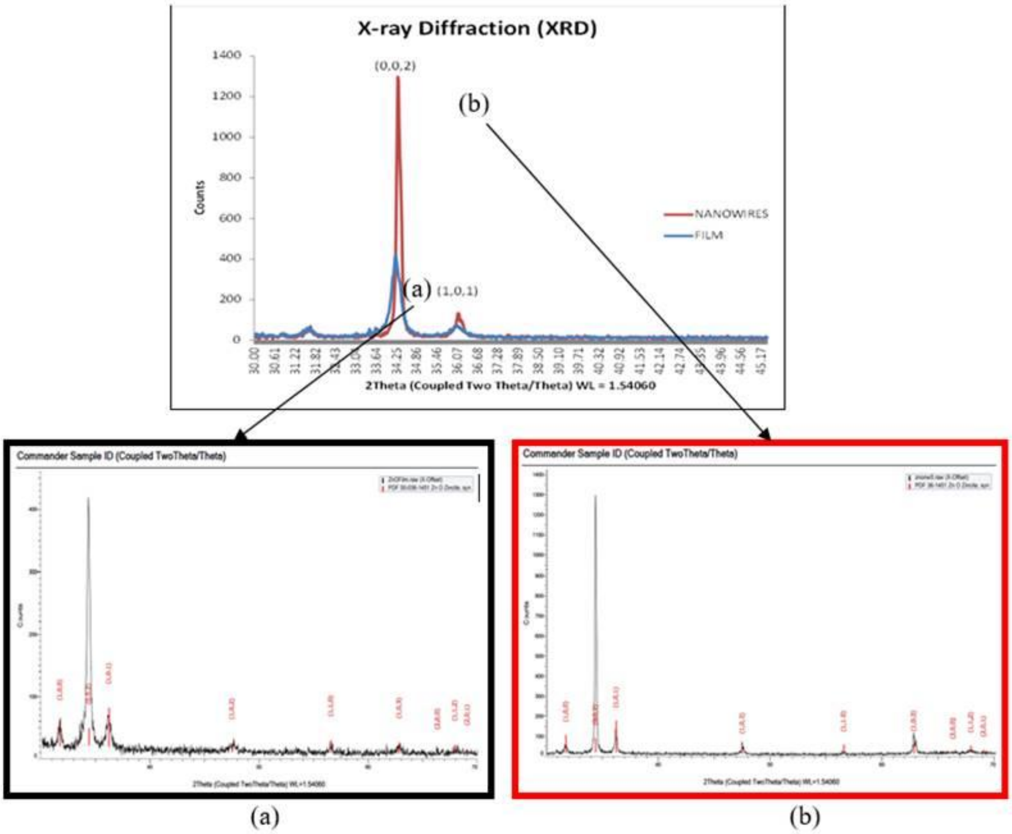

Figure 6. XRD patterns of synthesized ZnO film and ZnO nanowires, (a) ZnO film coated on SiO_2/Si wafer, and (b) ZnO nanowires grown on SiO_2/Si wafer.

3.4 Electrical Characterization of Developed SAW Devices

The electrical test is to observe the behavior of resistance and capacitance-voltage characteristics. The measurement instrument used for this electrical test is Keithley 4200-SCS Semiconductor Parameter Analyzer (SPA). The electrical measurements were conducted by probing the aluminium pads at IDTs at room temperature. The C-V characteristic test is being conducted in order to prove the result of frequency response compare with result test by using network analyzer. Base on the measurement data of C-V characteristic test, the frequency response then was calculated by using equation 2. The C-V curve of the IDTs with aluminum as sensing area design that had been tested is shown in Figure 7. The measurement was carried out at voltage range of -7 V to 7 V. It was clearly observed that the capacitance has the constant value of 4.17 *n*F. The frequency frequencies of the propagating wave along the surface of an elastic substrate are calculated by Equation 1. By assuming that the velocity of SAW on ZnO is 4200 m/s and λ is 800 *μ*m [11, 23]. Therefore, the expected frequency is around 5-7 MHz. The values of center frequency that gain from the calculation in equation 2 is about 6.12 MHz. Both results then were compare in result from network analyzer. From result, the center frequency has gain in arrange 6.0 MHz to 6.5 MHz. From all results, show still in the range of wanted frequency.

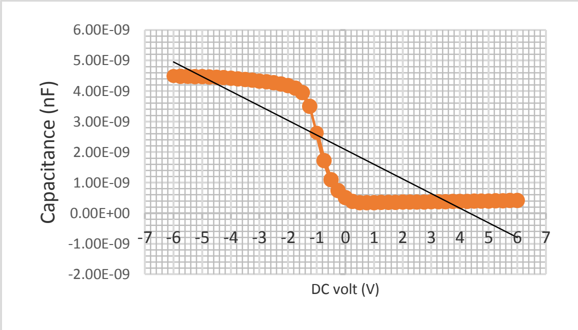

Figure 7. The graph of C-V characteristic of IDT design.

The Agilent E5062A network analyzer is used to conduct frequency response measurement and compare result of sensing area with Aluminum film and ZnO nanowire. From the result also, the center frequency and insertion loss can be evaluated. The measurement frequency range for these fabricated SAW devices is within 300 KHz to 3 GHz. As shown in Figure 8, the center frequency for ZnO nano wires and aluminium as sensing area are approximately 8.34 MHz and 6.36 MHz, respectively. It shows have shift frequency response for both materials as sensing area with Δf_o2MHz. While, the SAW device with ZnO nano wires has less insertion lost which is -37.07dB as compare to aluminium film. The insertion lost may due to the impedance mismatch of input and output IDT [15]. Both SAW device shows difference value in center frequency and insertion lost. Therefore, both SAW devices exhibit difference sensitivities at difference frequency range.

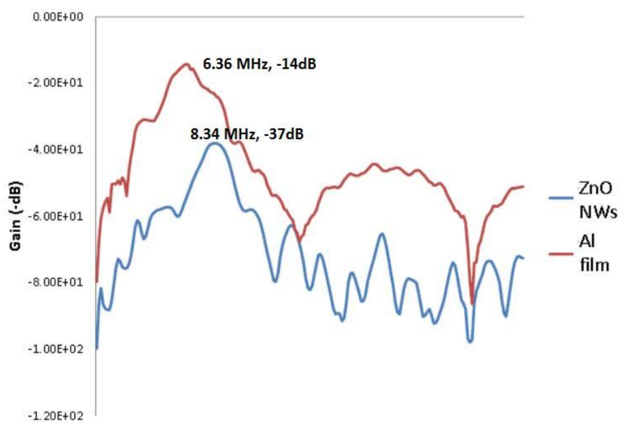

Figure 8. Frequency responses for SAW device with difference sensing material.

4. CONCLUSION

The SAW sensor device was successfully fabricated using lift-off photolithography process coupled with sol-gel method. ZnO nanowires modification on SAW sensing area was performed using dip coating process based on chemical reaction. The characterization of ZnO nanowires using AFM, SEM and XRD indicated that the nanowires structures with diameter from 100 nm to 500 nm and few micrometer length was perfectly grown on the sensing area of SAW device. Enhanced in electrical properties has been observed through the measurement of capacitance and frequency response for the devices that consists of ZnO nanowires as sensing area, suggesting its potential platform in application of a low-cost biodiagnostic devices. From frequency response shows the sensing area with ZnO nanowire are higher compare to Aluminum thin film, hence the sensitivity of the device is better.

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