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Characterization of Zinc Oxide (ZnO) piezoelectric properties for Surface Acoustic Wave (SAW) device

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Abstract. In fabricating Surface Acoustic Wave (SAW) biosensors device, the substrate is one of important factors that affected to performance device. there are many types of piezoelectric substrate in the markets and the cheapest is zinc Oxide substrate. Zinc Oxide (ZnO) with its unique properties can be used as piezoelectric substrate along with SAW devices for detection of DNA in this research. In this project, ZnO thin film is deposited onto silicon oxide substrate using electron beam evaporation (E-beam) and Sol-Gel technique. Different material structure is used to compare the roughness and best piezoelectric substrate of ZnO thin film. Two different structures of ZnO target which are pellet and granular are used for e-beam deposition and one sol-gel liquid were synthesize and compared. Parameter for thickness of ZnO e-beam deposition is fixed to a 0.1kÅ for both materials structure and sol-gel was coat using spin coat technique. After the process is done, samples are annealed at temperature of 500°C for 2 hours. The structural properties of effect of post annealing using different material structure of ZnO are studied using Atomic Force Microscopic (AFM) for surface morphology and X-ray Diffraction (XRD) for phase structure.

1 Introduction

Surface Acoustic Wave (SAW) sensors have been successfully enhanced in its technology almost in every type of application including temperature, pressure, telecommunication and biochemical sensing. Relevantly, material of substrate will give an effect towards the sensing performance in biosensor field [1,2]. Today, many technology SAW has been used in Biosensor detection. SAW biosensor basically consists of three main parts which is piezoelectric substrate, a pair of metallic InterDigital Transducers (IDTs) and sensing area [1-3]. Operation of SAW biosensor normally depends on the output frequency response for characterization. It is operated by generating input IDT and output IDT will then detect the acoustic waves by employing the piezoelectric effect. Surface acoustic wave is generated from the input IDT and transducer mechanical wave into electrical signals by the output IDT [4]. Piezoelectric layer is very important in fabricating the SAW biosensor. This thin film is one of the important factors that improve the performance of the SAW device in term of sensitivity and accuracy in detection [5].

Piezoelectricity is an electrical charge which produces when certain material experience mechanical stress. Alternately, the mechanical force will be created when electricity applied on the same material. The commonly used piezoelectric substrate materials are Quartz (SiO2), Lithium Niobate (LiNbO3), Lithium Tantalate (LiTaO3), and zinc oxide (ZnO) [4,5]. Each of them has different properties such as cost, piezoelectric coupling coefficients, attenuation, temperature sensitivities and propagation velocity.

However, in this study ZnO is chosen as it is a material with great advantages for piezoelectric substrate. ZnO is a n-type semiconductor material and used for several sensor devices considered its sensitivity and easy doping method to improve sensing performance [6]. It is characterized with its direct wide band gap of 3.37 ev at temperature room, high excitation binding energy of 60 mev allows it to have some specific behaviour in physical and chemical properties [1,4,6,7]. This II-IV compound semiconductor is crystallized in hexagonal wurtzite structures [6,8]. With these bunches of outstanding characteristics, ZnO is believed to an exhibition of strong piezoelectric properties [1,4,8]. ZnO is said to be high in piezoelectric coupling factor and a good bonding to varying substrate such as silicon, silicon oxide and silicon nitride [8].

Thus, in this research ZnO thin film is deposited onto silicon oxide surface to ensure good bonding and prevention of eroded during further fabrication step such as lift-off process. There are various technologies that can be used for ZnO deposition onto different substrate material. Currently, variety deposition techniques available are Pulse Laser Deposition (PLD) [2,6], Chemical Vapor Deposition (CVD), RF Sputtering [2,6,9,10], Sol-gel method [6] and Electron beam evaporation [9,10]. The substrate temperature, coating

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conditions, annealing and thin film thickness will eventually effect on the optical and physical characteristics of ZnO thin films grown [11]. In this study, ZnO thin film is evaporated by reactive electron beam technique in oxygen environment and prepared by sol-gel technique to compare the best roughness for SAW device. The analysis will be done by characterized using AFM and XRD.

2 Experimental Details

2.1 Silicon Wafer sample preparation

Pure Silicon substrate wafer (100) is first cleaned using Buffered Oxide (BOE) to clear any unwanted native oxide. Then, the wafer is checked using Film Spectrophotometer to ensure zero oxide on the surface of the wafer. Thermal oxidation process is done onto bare wafer in which silicon will react with oxygen under high temperature to form a layer of silicon dioxide. Usually the thickness of silicon dioxide depends on the oxidation rate whereby oxidation type such as dry or wet oxidation, temperature and flow rate in the furnace is considered. Wet oxidation is used in this process as it is aiming to grow high quality insulator with parameter of 1000°C for 45 minutes using oxidation furnace. For resistivity improvement, SiO_2 was growth to a range of 3000-5000µm respectively. ZnO target is then deposited onto SiO_2 by using different structures which is pellet and granular and sol gel ZnO liquid.

The deposition is set by e-beam evaporator Korean Coating Material & Component (KCMS) KES-200D with a parameter of 0.1kA for thickness deposition. An electron source in a vacuum will generate electron beams in order to irradiate any evaporate material. Then, the evaporated material will be heated and evaporated to ensure evaporated material forms a thin film on subjects samples. The chamber gauge is evacuated to a pressure of 3.5^{-5} to 4.5^{-5} Torr and the high voltage of about 7.3kV. The film growth rate of deposition is varied along the time within the range 0.01Ås^{-1} to 0.10Ås^{-}1 with a duration of 2 hours. However the parameter is depending on the substrate temperature, oxygen pressure and filament current of the electron beam [12].

This is followed by thin film characterization using Atomic Force Microscopy (AFM) and X-ray diffraction (XRD) for surface morphology and composition studies.

2.2 Pellet source for E-Beam Evaporator

To perform an e-beam technique, ZnO pellets firstly needs to be prepared. The conventional sintering technique is used to form undoped ZnO pellets using zinc oxide powders with high purity of ≈99%. ZnO powder was first balanced for 1g on the scales with up to ±0.02g uncertainty. ZnO powder is then milled in an agate mortar by using a pestle and mortar in the Sustainable Engineering Research Center. Manual Hand Pellet Presser was used to form pellets disks with approximate pressure of 500psi. Then, the pellets were sintered at various temperature of 700°C-1000°C in ambient air for 3 hours. Lastly, the pellets is quenched to normal room temperature to freeze the structure formed. The two pellets of 1cm x 1cm diameter size sintered pellets used for one deposition of ZnO thin film using e-beam. Figure 1 defines the undoped ZnO pellet formed.

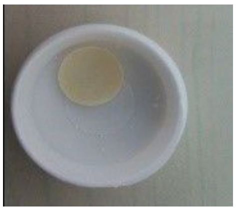

Fig. 1. Undoped ZnO pellet

2.3 Granular sources for E-Beam Evaporator

As for granular source, which is provided directly from the manufacturer, it is characterized by a small size of approximately of 1-3mm and with high purity of 99.99%. An amount of ZnO pellet is loaded into boat for E-beam evaporation. Figure 2 shows an amount of ZnO pellet granular source.

Fig. 2. ZnO pellet granular source

2.4 Sol Gel Zinc Oxide Preparation

Zinc oxide sol-gel solution was prepared before spin coat onto substrate. A 880mg of zinc acetate dihydrate (ZAD) is measured using an electronic mass scale and mix into a beaker of 20 ml of dehydrated 2-Methoxyethanol solvent. The mixture stirred at 1000 RPM on a magnetic stirrer for 20 minutes at 60 ºC. After that, a total of 244 µl of diethanolamine (DEA) is added drop by drop every 10 minutes into the mixture in the beaker on the magnetic stirrer for the next 2 hours. Finally, the mixture is left for 24 hours at room temperature for aging time before the solution is used. Steps for ZnO preparation as shown in Figure 5.

The ZnO is deposited using a conventional deposition process where the ZnO solution will be dropped on the silicon oxide surface using a dropper. The substrate will be spun using spinning to get a uniformity form. The zinc oxide sol-gel is dropped and coat uniformly onto the sample using the photoresist spin coater. After that, the sample will be pre-heat on the hot plate for 10 minutes to remove the sol-gel solvent followed by cooling down on a ceramic plate. The coating process is repeated until the desire number thin film layers achieved. Next, the samples are annealed at 500 °C for 2 hours with nitrogen gas to decrease the sheet resistance of the piezoelectric layer as well as to improve the surface quality of the zinc oxide. Since more zinc oxide sol-gel solution is needed to coat the samples for 25 layers, 40 ml of zinc oxide sol-gel solution is prepared instead of 20 ml. The target is piezoelectric zinc oxide thin film with a thickness of 0.5 µm. Figure 3 shows steps for ZnO sol-gel technique.

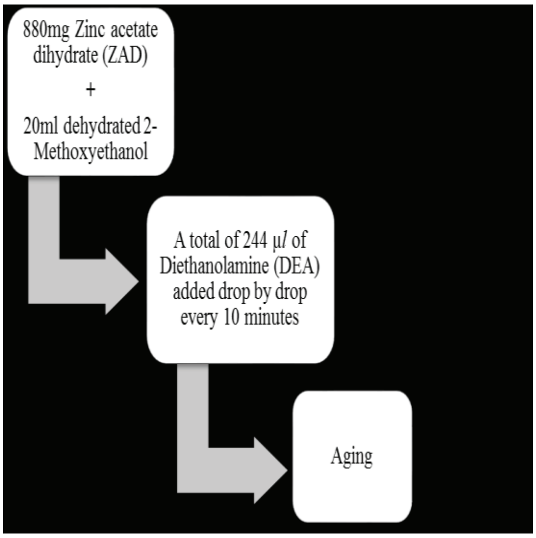

Fig. 3. Steps for ZnO Sol-grl preparation.

3 Results and Discussion

3.1 AFM Analysis

The surface morphology of deposition using different structures of ZnO is analyzed using Scanning Probe Microscope SPA400 (AFM). Figure 4(a) shows a 3D image of ZnO thin film using pellet source. Cone-like topography can be seen clearly from the certain expected of good uniformity scan area. The 3D image illustrates the light and darker color of the non-uniformity with highest peak is reach up to 100nm. Average roughness Ra of the sample is 20.39 nm and a peak to valley roughness P-V of 185.5nm shows that the surface of the sample got a more significant deviation between the distance of the highest peak and the lower valley of the grains. The small grain size of the sample is equivalent to 546nm with RMS roughness of equal to 25.71 nm. Crystalline structure and rough surface of ZnO sample is slightly better after annealing process.

Figure 4(b) defines the 3D image of ZnO thin film using granular source for deposition of e-beam. The highest peak is 150nm with average roughness of 28.74 nm. Grain size is equivalent to 1029.926 nm with P-V of 246.9 nm. Sample defines a slight roughness and crystalline structure using ZnO granular source after annealing process is far better compared to the ZnO pellet source as it is good in uniformity.

When coated for 25 times and anealing, the sample surface exhibit bigger ZnO grains based on the 3D images with 40 nm peak in Figure 4(c). Surface analysis confirms that average roughness Ra increases with the increasing number of coating in average 19.3 nm with P-V 106 nm. ZnO grain size also has been observed to increase with the increase in thickness of the film with more sol-gel coatings. The grain size has increase from average 170 nm. R. Gayen et al. [13] also reported that there an increase in grain size with an increase in zinc oxide thin film layer

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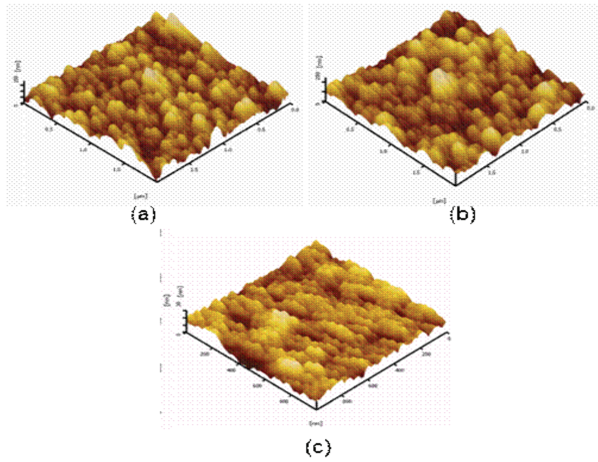

Fig. 4. The AFM 3D image of ZnO thin film a)using pellets and b) using granular source c) sol-gel

Surface analysis for all three techniques are summarized in Table 1. From results shows that the ZnO granular source has better quality compare to others samples in term of surface roughness and thickness. In reference [14], A. Zaki has claimed 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 can enhance the performance of piezoelectric substrate.

Table 1. Comparison result of AFM for ZnO

Characteristic	ZnO Pellet Source	ZnO granular source	Sol gel (25 coatings)
Root mean square (rms) (nm)	25.71	4.55	23.29
Peak to valley height(P-V)(nm)	185.5	246.9	106
Average roughness (Ra) (nm)	20.39	28.74	19.37
Thickness (nm)	100	200	20

3.2 X-ray Diffraction (XRD) Studies

To confirm the thin film coating of the ZnO sol-gel in consistency with crystalline species of the ZnO material, the samples were tested using diffractometer. D2 Phaser desktop diffractometer from Bruker AXS were used and operated at 30 kV and 10 mA with a Cu radiation source. 2θ/θ scan type is employed with scan angle 20° to 60°. All diffractograms were analyzed using DIFFRAC.EVA software from Bruker AXS and undergo material matching and correction processes such as smoothing and baseline correction. The characterization of XRD focuses on the range of 30° to 40° as it shows the most eligible range for ZnO and matched the reference pattern perfectly. Figure 5 illustrates a comparison of XRD result for e-Beam deposition for using ZnO pellets and granular sources after annealing process. Result granular source (line blue) shows the highest peak intensity up to 654 with at an angle of 34.43° with preferred (111) crystal axis orientation whereas the weak peak responded to an angle of 32.25° and 36.60° respectively. For preparation ZnO pellet source shows an average counts through angle 30° to 40° and sudden peak at angle 34.39° with a value of 466. Others weak peak are none clearly seen and have an average intensity along the tested of 2θ. Both of data within the range angle of 30°-40° matched perfectly referring to a PDF database of Cui Shouxin, et.al (2009) [15]. Using granular source indicates the greater intensity compared to the pellets with same parameter and condition applied. Since samples had undergone annealing process, when temperature increases thus pressure also increases. After annealing, wurtzite ZnO is compressed which reduced the height and shifted down energy [15].

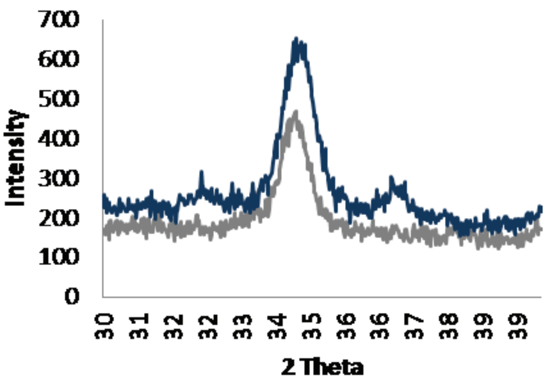

Fig. 5. Comparison of XRD using ZnO pellets and granular source

For the Sol-Gel analysis the samples exhibit 2θ peak at 31.8°, 34.3° and 36.4°, and 47.6°. However, the strongest peak intensity of the X-Ray diffraction was observed at 34.3° that related to preferred (002) c-axis ZnO crystal orientation. Other weak peak intensity corresponds to ZnO crystal orientation are for (100) at 31.8°, (101) at 36.4° and (102) at 47.6°. Therefore, the ZnO sol-gel coatings has been confirmed to produce preferable c-axis orientation for piezoelectric thin film with the presence of peaks of reflections of the ZnO wurzite structure with lattice constant a= 3.242 and c=5.176. A review on zinc oxide sol-gel done by L. Znaidi [16] also reported that a similar finding on the preferred zinc oxide thin film with (002) crystal orientation.

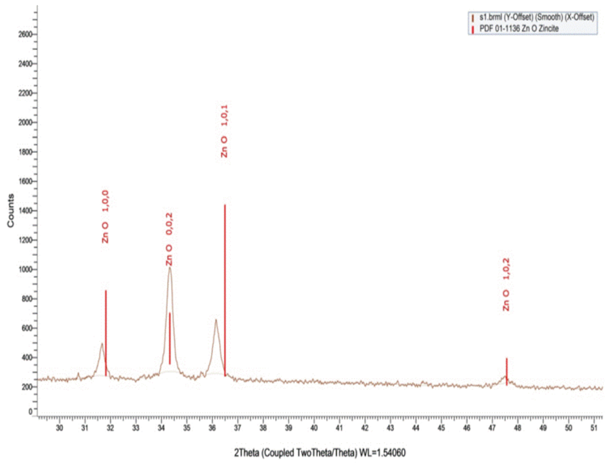

Fig. 6. Caption of the Figure 1. Below the figure.

3.3 UV-VIS Analysis

UV-Vis transmittance spectra analysis of the ZnO prepared by granular deposition and sol-gel coating are shown in Figure 7 after heat treated with annealing 500 C temperatures. The range of the wavelength is taken from 300nm to 800 nm. The ZnO thin film absorbs UV light in the wavelength from range 300-400 nm for both results. From the graph, the potential cut-off wavelength takes place from 370nm – 385nm and no other peak was observed in the spectrum confirms that the synthesized are pure ZnO and its wide-band-gap is about 3.3eV. [17,18] The band gap energy can be evaluate by substituting the value of the absorption peak at a given wavelength in the following equation [19]

$$E_g = h\nu_g = hc/\lambda_g \quad (1)$$

Where h = 4.14x10^{-15} eVs, c = 2.99x10^{8} m/s, and λ is referred in Figure 6.

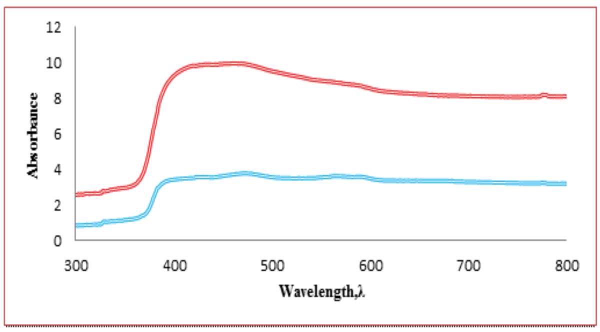

Fig. 7. UV-vis absorption spectrum of ZnO nanoparticles.

3.4 SEM Analysis

Figure 8 shows the SEM images of the granular source samples that undergo different annealing temperature. All the results taken were measured by the average applied voltage 15kV and the beam diameter around 1μm while the magnification range applied ranging from 10000X to 50000X in order to observe the difference surface morphology of the thin films. Figure shows, the mean size and also size distribution of particles are increasing with increasing annealing temperature. The important results concern the smoothest surface grain size of ZnO. The results are in agreement with the previous work reported [20], demonstrated that the annealing effect will improve the structure of ZnO form nanostructure ZnO thin film

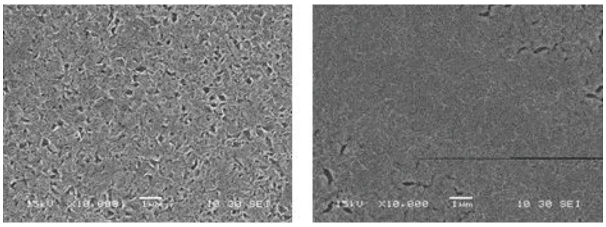

Fig. 8. SEM image of granular deposition ZnO a) before annealing b) after anealing.

4 Conclusion

Nanostructured ZnO thin films have been deposited using E-Beam and 25 layers coated by sol-gel technique. The samples were characterized by XRD, SEM, AFM and UV-VIS. The XRD analysis confirmed that the thin film was ZnO nanostructure. The thin Film produces preferable c-axis orientation for piezoelectric thin film with the presence of peaks of reflections of the ZnO website structure. From SEM analysis shows that the average particle size of ZnO increases with increasing annealing temperatures. UV-VIS analysis shows the range wavelength in range 300nm and 400nm with band gap 3.3eV. From the result, granular source shows the best layer in term of roughness and absorbance. These ZnO Thin films can be used in the SAW Biosensor application.

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