Digest Journal of Nanomaterials and Biostructures Vol. 12, No. 4, October-December 2017, p. 1119-1126

Ar GAS FLOW RATE INFLUNCEMENT ON THE STRUCTURE PROPERTIES AND ELECTRICAL BEHAVIOR OF NEW SENSING MEMBRANE ZnO/CU/ZnO (ZCZ) EXTENDED GATE FIELD EFFECT TRANSISTOR (EG-FET)

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In the last three decades, It has been proposed Zinc Oxide (ZnO) at the in front of metal oxide semiconductors to be used in various ions detection fields. This is due to its distinctive properties. ZnO/Cu/ZnO(ZCZ) multilayers structures as an extended gate were deposit on glass substrate using RF and DC magnetron sputtering at different ratio of Ar gas sputtering (6,12) sccm, XRD and AFM results were used to analysis the structural properties of multilayers. The average roughness of multilayer surface at Ar ratio 6sccm =0.55 nm less than multilayer film at 12 sccm. The pH sensitivity was calculated and found 30 mV/pH, 0.52 $(\mu A)^{1/2}$/pH for new sensing membrane multilayer at Ar =6sccm best than multilayer deposit at gas sputtering 12 sccm which had the value 25 mV/pH, 0.35 $(\mu A)^{1/2}$/pH this study focusing on the relation between the Ar gas flow rate with roughness and the electrical behavior represented by pH sensitivity, hysteresis voltage) as EGFET pH sensing membrane.

(Received October 5, 2017; Accepted November 17, 2017)

Keyword: ZnO, Cu, multi-layer, EGFET, pH, Ar gas flow rate

1. Introduction

Transparent conductive oxide an important material and studied extensively because it has rang of application such as solar cell [1], flat panel display [2], pH sensing [3]. Among these oxides a binary oxide in its properties (Zinc Oxide) (ZnO) such as cheap [4], non toxic, high stability in a hydrogen plasma[5], high transmission approximately 90% with high resistivity [6] .

Recently most useful method to reduce the resistivity of ZnO is inserting layer of metal between two layers of ZnO the metal layer proposed to improve the conductivity of the stack. This multi-layer system (ZnO/metal/ZnO) based on semiconductor ZnO and metal such as Ag, Cu, Pd used to improvement the properties of ZnO resistivity in balance with its transmission, Many previous studies about multilayer structure with different mid layer metal such as Ag [7, 8], Mo [9], Au [10, 11], and Cu [12, 13].

Mihenea *et al.* [14] were prepared multi-layers ZnO/Cu/ZnO by using RF and DC magnetron sputtering from ZnO target and Cu target ,the multilayer deposit on (PEN) flexible substrate. The structural, optical and electrical properties of the multi-layers was investigated, the morphology of the multi-layers were demonstrated and indicated that the structure of was nanostructure. Also, the study was reported that the transmission of the multi-layer was decrease with increase the Cu metal thickness. The sheet resistance of the multi-layers ZnO/Cu/ZnO almost 100 Ω/sq when used Cu metal as a mid-layer.

Daeil Kim[15] polycarbonate substrate (pc) have been used to deposit ZnO/CuZn/ZnO multi-layers via RF and DC magnetron sputtering by ZnO and CuZn target. The SEM image analysis and cross section were indicated that the multilayer had a normal grain growth with 100 nm, where sheet resistance of the multi-layers ZnO/CuSn/ZnO was 44 Ω/sq.

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Extended gate field effect transistor (EG-FET) device presented as alternative to ion sensitive felid effect transistor (ISFET). First operation of extended gate field effect transistor (EGFET) pH sensors was reported by Van De Spiegel *et al*. in 1983 and based on ISFET operation principle except that the gate out of metal oxide semiconductor felid effect transistor (MOSFET) and connected to sensing membrane which immersed in pH buffer solution.

The configuration of EGFET had many advantages like insensitivity to light and temperature, cheap, flexible shape of extended gate, Especially the protection of MOSFET because it far away from the sensing area, so we can use it a lot times .[3]

The measurement of ion content obeys the quantum theory (Site binding model) surface potentialφ as seen in equation (1):[16]

$$2.303\left(pH_{pzc} - pH\right) = \frac{q\varphi}{KT} + \sinh^{-1}\frac{q\varphi}{KT\beta} \quad (1)$$

Where pH_{pzc} is pH value at point of zero charge, K Boltzmann constant, T absolute temperature.

The chemical sensitivity β depended on the number of surface site per unit area N_s according to the equation2:

$$\beta = \frac{2N_s q^2 (k_b/k_a)^{\frac{1}{2}}}{KTC_{DL}} \quad (2)$$

where K_a and K_b are constant, C_{DL} the electrical capacitance.

From equation 2 high N_s leads to higher β.

This study focusing on the effect of gas sputtering flow rate on the sensitivity of multi-layer (ZnO-Cu-ZnO) extended gate field effect transistor (EGFET) with optimum thickness of the multi-layer (60-20-60) nm Cu has been chosen as metallic mid layer because cheaper than many metals such as Ag. In additional, the multi-layer sensing membrane device with commercial MOSFET is suitable for biomedical application and cheaper than disposable chemical sensor.

2. Experimental work

2.1 Multi-layer film preparations

Microscope slides glass substrate (1cm × 3cm) dimension was used to deposit ZCZ multi-layer thin film; glass slide was ultrasonically cleaned in acetone de-ionized water, and then dried with nitrogen gas. The deposition was carried out using (HHV Auto 500 vacuum coater with turbo molecular pumping system, RF and DC magnetron sputtering system. All multi-layers were fabricated used ZnO target (99.999% purity) and Cu target (99.999% purity).

Thickness of multi-layers ZnO/Cu/ZnO controlled by sputter time and set to be (60/20/60) nm at two value of argon gas sputtering flow rate (6, 12) sccm (standard cubic centimeters per minute). The base pressure was 4.1 ×10-5 mbar. The pressure during working for ZnO target was 6.75×10-3 mbar and for Cu target was 5.1×10-3 mbar. RF and DC power, it has been fixed at 100 W. Figure 1 illustrated the growth process.

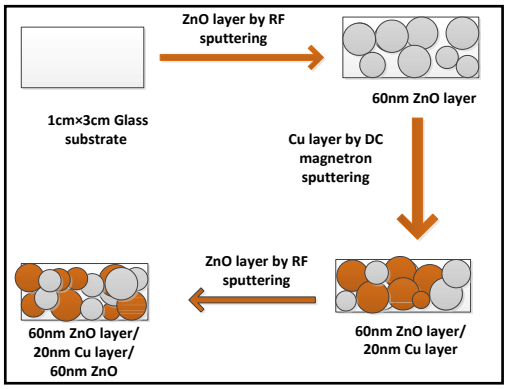


Fig. 1. (ZCZ) multi-layer growth process

2.2. ZnO/Cu/ZnO multilayers film characterization

The crystalinity structure of ZCZ multilayer film was investigated by PANalytical using X-ray diffractometer (XRD) (intensity -2theta model) equipped with CuKα source (λ=0.15418 nm). Nanoscope analysis dimension edge, bruker Atomic force microscopy (AFM) used to determined the morphology and roughness of the multilayer film.

2.3. pH sensing system

The (I_{DS}-V_{DS}),(I_{DS}-V_{REF}) characteristic of ZnO/Cu/ZnO multilayers film EGFET were measured using pH system as shown in Figure 2.

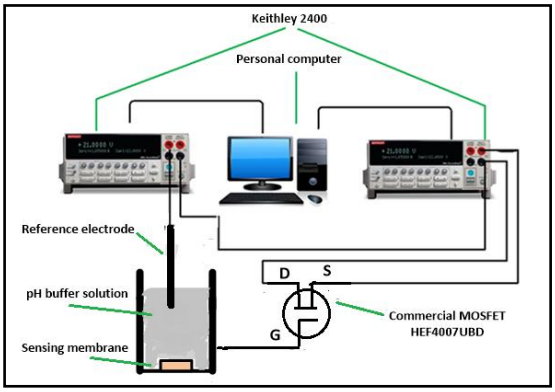


Fig. 2. pH system setup

pH system as seen in figure 2 consist of two Keithley 2400, personal computer (pc) to analysis and save data (I-V) curves, commercial MOSFET HEF4007 UBD, Ag/AgCl reference electrode connected to the gate of MOSFET, cavity of sensing film membrane ZCZ films and set of pH buffer solutions which have pH range (2-12).

3. Results and discussion

3.1. Multi-layer structures

X-ray diffraction analysis have been used to examined the crystalinity and phase of ZnO/Cu/ZnO multilayers thin film prepared at two condition of gas sputtering (Ar=6 sccm,12sccm), all films had same phase that (002), (101), (102), (103), (004) of ZnO with hexagonal structure[17, 18]compared with small peak for Cu metal (111) [19] was observed in Figure 3.

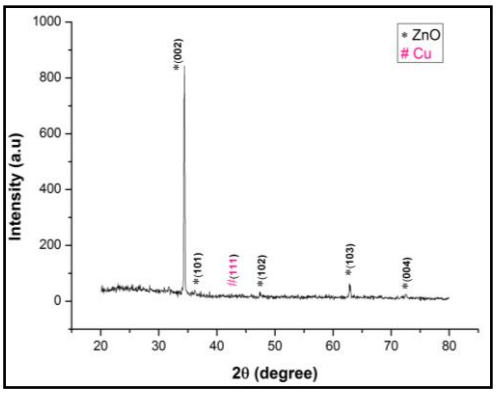


Fig. 3. ZCZ multi-layer XRD pattern (Intensity – 2theta) scan

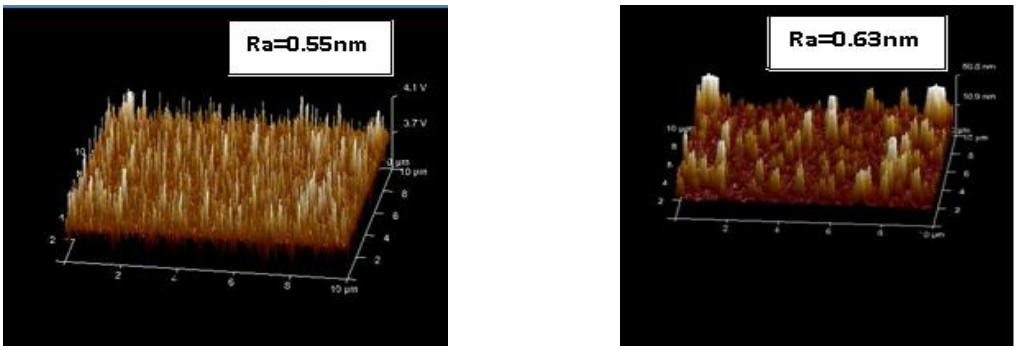
The results indicated that the ZCZ multilayers that polycrystalline, no observation effect of gas sputtering on phase of multilayers films.

3.2. Roughness and 3D image of multi-layer ZnO/Cu/ZnO

ZnO/Cu/ZnO multilayers thin film were deposit on glass substrate by using physical method RF and DC sputtering system which it provides the film many advantages such as good homogenous and durability, to determine the quality of deposition technique to achieve the desirable film smoothness for suitable application AFM is used for characterization of surface topography with resolution up to angstroms in height and demonstrated the deviations of surface such as hillocks and valleys.

One of surface deviations surface roughness is can characterized as the degree of unevenness along out of plane of the film surface or surface irregularity it is provides a measure of how smooth (or rough) the surface of a film is as showed in Figure 4 (a,b). The root means square roughness (Ra) values for ZnO/Cu/ZnO multilayer thin film prepared at different ratio of gas flow rate Ar increasing with increase the Ar ratio. [20]

Surface roughness play a key role in study the crystalinity of material roughness is determined by weakens of oscillations this fact agreement with XRD results.

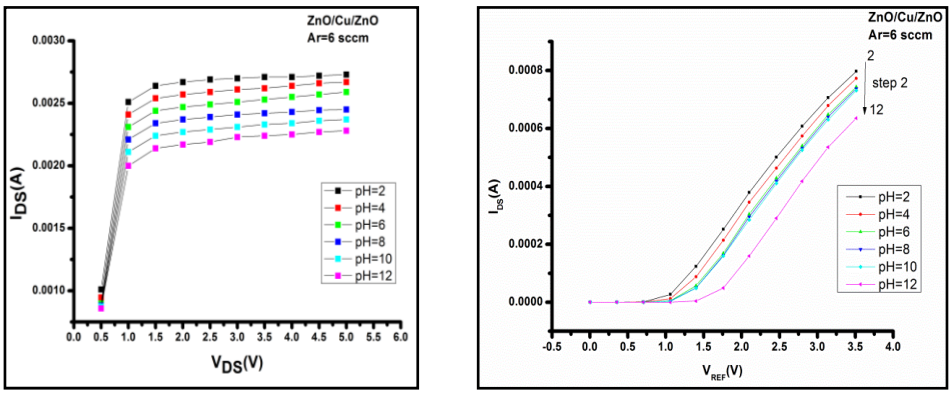


a) b)

Fig.4. Ttopographic 3D images of ZCZ multilayer prepared with gas flow rate Ar (a) 6 sccm (b) 12 sccm

3.3. pH sensing membrane characterization

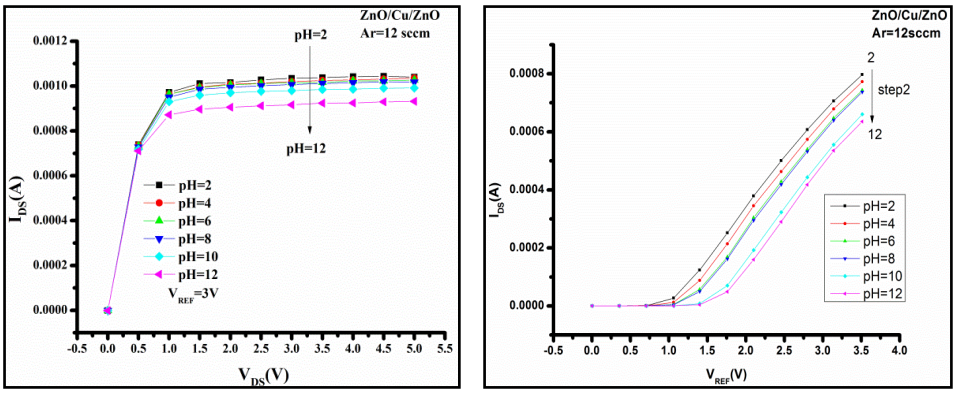
After growth of ZCZ multilayers at two ratio of gas sputtering (6, 12) sccm. electrical properties (I-V) curves of ZnO/Cu/ZnO multilayers as extended gate field effect transistor sensing membrane were characterized by using pH system showed in figure2. (The electrical properties of the two samples were characterized from (I-V) curves of EGFET by using the system showed in figure2).



a) b)

Fig. 5. (IDS-VDS),(IDS-VREF) Characteristic curves in (a) saturation regime(b) linear regime of ZCZ at Ar ratio 6 sccm

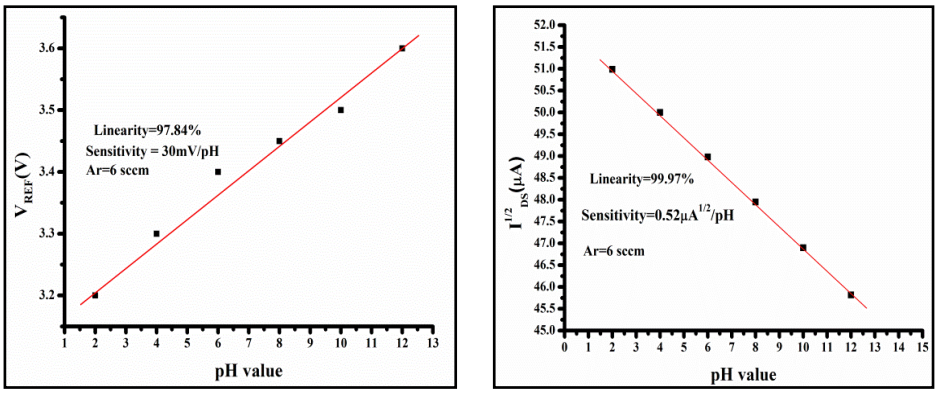
Fig. 5.a shows the (I_{DS}-V_{DS}) curves in saturation region and Fig5b (I_{DS}-V_{ref}) in linear region of multilayers ZnO/Cu/ZnO at gas flow rate 6 sccm, whereas I_{DS}, V_{DS}, V_{REF}, drain to source current, drain to source voltage and reference electrode voltage respectively. Figures 5(a) and 5(b) was showed that: there is an inversely proportional between I_{DS} and pH, it can observed that by saturation and linear region plots. From saturation region can observed that the V_{REF} rises with pH value because higher pH means more OH^- ions in solution which equivalent the (-V) applied on EGFET[3] .



a) b)

Fig. 6. (IDS-VDS),(IDS-VREF) characteristics curves for(a) saturation regime(b) linear regime respectively of ZCZ at Ar ratio 12 sccm

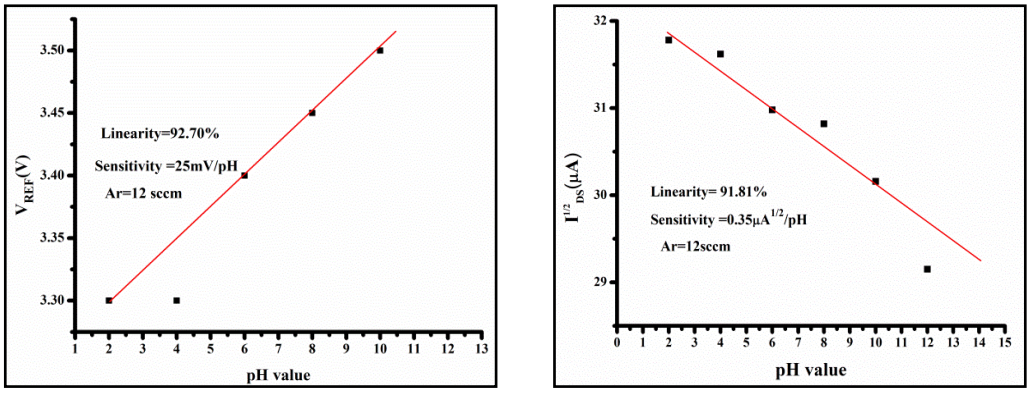
The sensitivity of (ZCZ) sensing membranes at saturation region was calculated from the linear relation between square root of I_{DS} versus pH. [20], The results showed that: the sensitivity of the first sample (6 sccm) was 0.52$(\mu A)^{1/2}$/pH. While I_{DS} sensitivity of the second one (12sccm) was 0.35 $(\mu A)^{1/2}$/pH. Reducing sensitivity value of attributed to increasing the ratio of Ar gas (Ar atoms increase) during the sputtering deposition of the meta



a) b)

Fig.7. Sensitivity plot of ZCZ multilayers sensing membrane at gas ratio 6 sccm (a) linear regime (b) saturation regime

In the linear region, the other function of pH sensitivity in terms of potential (equation 1) , which that is subject to Nernstian limit (59.2 mV/pH) [21] was calculated from the slop between VREF and pH. This was 30 mV/pH, 25 mV/pH for (ZCZ) multilayers membrane at gas ratio (6, 12) sccm respectively.



a) b)

Fig. 8. sensitivity plot of ZCZ multilayers sensing membrane at gas ratio 12 sccm (a) linear regime (b) saturation regime

From results above the best sensitivity for multilayers sensing prepared at Ar gas flow rate 6 sccm means that less Ar gas flow rate better to enhanced the sensitivity of (ZCZ) multilayers. this study discussed the effect of change Ar gas flow rate with sensitivity to pH sensor [21] Hsiao-Wen ZAN *et al* reported that the AIN gate leakage in OTFT transistor reduce by controlling the Ar/N_2 gas ratio during sputtering process. And denoted that the gas ratio affected on threshold voltage and leads to reduce it. Ali Sardarinejad *et al* [22] were prepared the RF sputtered RuO_2 thin films by using different gas ratio Ar/O_2 and then test the film as EGFET , the sensitivity of the RuO_2 EGFET was enhanced from 64.33–73.83 mV/pH when the Ar/O_2 ratio change from 10/0–7/3. This implies that low Ar gas flow rate leads to higher sensitivity, this results agreement with our results.

Table 1. Sensitivity comparison for many materials

Materials	Linear region		Saturation region		Reference
	Sensitivity (mV/pH)	Linearity %	Sensitivity ($\mu A^{1/2}$ /pH)	Linearity %	
ZnO/Si	46.25	99.02	0.73	99.03	[23]
Psi	66	99.3	0.76	99.4	[24]
ZnO/Cu/ZnO (Ar=6) sccm	30	97.84	0.52	99.97	This study
ZnO/Cu/ZnO(Ar=12)sccm	25	92.70	0.35	91.81	This study

Table 1 presented the comparison in sensitivity and linearity values for many materials with our study.

4. Conclusions

ZnO/Cu/ZnO transparent conductive multilayer were successfully deposit on glass substrate by using RF and DC magnetron sputtering system, the multilayers prepared at two condition of Ar gas sputtering flow rate (6, 12) sccm with fix thickness and power, the structural properties of the multilayers were investigated, AFM analysis indicated that the root mean square roughness increasing with increasing Ar flow rate, and then the multilayer check as pH sensing membrane for the first time, increasing the flow rate of Ar gas from 6sccm to 12 sccm affected on sensitivity pH of ZnO/Cu/ZnO EGFET and leads to reduce it.

Acknowledgment

We gratefully acknowledge the support of USM Research University grant 1001/PFIZIK/811220 and Malaysian Ministry of Higher Education Fundamental Research Grant 203/PFIZIK/6711349.

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