Effect of MgO Immersion Time on Dielectric Layer Properties of ZnO/MgO Films

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***Abstract*— This research work, focuses on the deposition of multilayer ZnO/MgO using immersion method deposited at different immersion time (2, 4, 6 and 8 hours). The resistivity values obtained were varied in the range of 12.5 to 20.0kΩ.cm which is due to the changes in carrier mobility and scattering. It was also found that, the leakage current, *J* was below than 10^{-8} A.cm^{-2} which is suitable for dielectrics. Some surface modification observed as the immersion time increased from 2 to 8 hrs which also reflects to the variation in resistivity, leakage current and *k* values obtained. The formation of flakes like structure was observed for multilayer films with 4 hrs immersion time which leads to the enhancement in *k* value at high frequency region.**

Keywords — Multilayer ZnO/MgO; Immersion time; Dielectric properties; Flake structure

INTRODUCTION

Nowadays, people are interested in low cost deposition techniques which also reflected to the deposition multilayer thin film. Multilayer thin film consists of alternating layers of two different materials [1]. Multilayer thin film are recently use for microelectronic fabrication, packaging and protection or for modifying the optical properties of a surface [2]. There are many materials that have been used to form multilayer such as multilayer ZnO/MgO and Polyimide (PI) multilayer. Recently, Sujira Promnimit and Joydeep Dutta have fabricated the multilayer thin films by Layer by Layer (LBL) process method and they conclude that by increasing the number of deposition cycles, the film thickness could be increased which lead to a reduced roughness induced by filling defect structures in the films [2].

Dielectric is an electrical insulator that can be polarized by an electrical field. Glass, paper and ceramic are the dielectric material. Zinc oxide, ZnO which has wide bandgap (3.37eV) and large excitonic binding energy of approximately 60meV [3]. Based on these properties, ZnO is suitable for dielectric material. Currently, ZnO has been used in many applications including new optical displays, MEMS related devices, light emission devices and solar cells [3]. Since today, ZnO had been used for the research such as Sharul Ashikin Kamaruddin et.al which investigate ZnO structure using Sol-Gel immerse technique [3] and A.E. Rakhshani has conclude that ZnO are stoichiometric, intrinsically n-type and consist of crystallites when deposited by immersion method [4]. Magnesium oxide, MgO is the important materials which has a large band gap with range is 7.3 eV to 7.8 eV. MgO has a dielectric permittivity *k* range from 6.7 to 10 which depends on the growth method and annealing conditions. It also has a high thermal conductivity and it's a chemical inertness, which minimizes the formation of an interfacial layer in contact with Si [5]. Based on all the properties above, MgO is the best material to be current dielectric material.

There are several methods to deposite multilayer thin film such as pulsed deposition, electron beam evaporation, chemical vapor deposition and sol-gel techniques [4]. However, sol-gel method provides its own advantages which are lower crystallization temperature, ability to tune microstructure via sol-gel chemistry, conformal deposition ability, compositional control, large surface area coating capability and low cost [3].

Immersion of sol-gel also known as the chemical bath method was recently used in film deposition [7,8]. This is because immersion method is a promising method to growth nanostructures such as prepare aligned Al-doped ZnO nanorod thin film for sensor applications while Guosong Wu et.al have conclude in their report that a facile solution-immersion method had formed a honeycomb-like layer, mainly composed of $Mg(OH)_2$ on its surface [7,8].

Therefore, in this study, the objective is to deposite multilayer dielectric in nanometer dimension using immersion method. Lastly, after completing the experiment, the properties of the deposited multilayer were verified for the dielectric application.

METHODOLOGY

Substrate cleaning process, solution preparation and multilayer film deposition are the steps that have been involved in this experiment. In this study, glass with dimension of 2.5cm x 2.5cm was used as the substrate. First of all, substrate cleaning process was performed in order to remove all impurities on the glass. The acetone, methanol and DI water were used as the cleaning material.

During solution preparation, two types of solutions have been synthesized which are ZnO and MgO. For ZnO solution, the precursor is Zinc Acetate dehydrate and 2-Methoxyethanol as a solvent while the stabilizer was Monoethanolmine. MgO solution was synthesized using magnesium nitrate, sodium hydroxide and DI water. The prepared solution was then sonicated for 10 minutes with

the applied temperature of 50^0C. Next, the mixed MgO solution was stirred at 50^0C for 30 minutes. The ZnO coated glass was then immersed in the 100ml prepared MgO solution at different immersion time with temperature of 95^0C. The immersion time was varied at 2, 4, 6 and 8 hours respectively. Then, the samples were removed and rinsed with DI water to eliminate residual salts and followed by an annealing process where the samples were annealed at 500^0C temperature for 1 hour.

When all the process complete, the characterization process was followed. There are three factors for characterizing the sample which are electrical properties, dielectric properties and structural properties. The summary of deposition and characterizations are as follows:

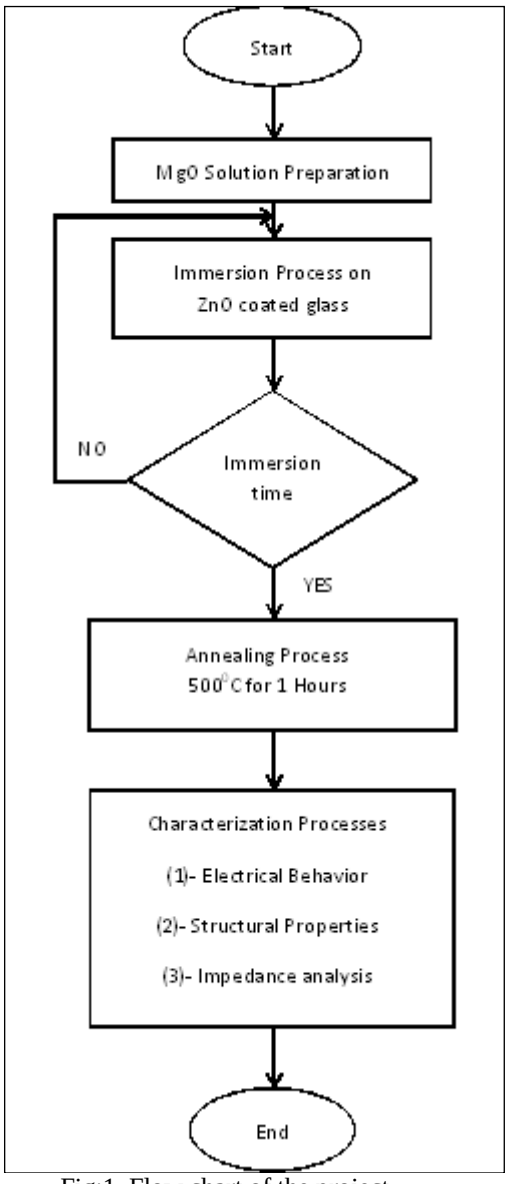


Fig:1. Flow chart of the project.

RESULTS AND DISCUSSION

ELECTRICAL BEHAVIOR

Electrical properties of the sample were performed using Bukoh Keiki two point probes *I-V* measurements and multilayer Zno/MgO was used as the test structure. Fig.2 shows the *I-V* curve of multilayer Zn0/MgO with different immersion time at a sweep voltage from -10V to 10V. Based on the figure, multilayer ZnO/MgO metal contacts form Ohmic characteristic which obtained for all samples after the measurements. From the graph, it shows that from 2 to 4 hours of immersion time, the resistance is increase while for 6 to 8 hours the resistance is vice versa. When the resistance increase due to voltage increase, it means that the small current were formed and its one of the insulator properties. From the plotted *I-V* curve, the resistivity of multilayer ZnO/MgO was calculated using the following equation:

$$p = \frac{wt}{\iota} R \qquad (1)$$

where *p* is the resistivity and the unit is Ω.cm, *w* is the width, t is the thin film thickness and ι is the length of the metal contact, and *R* is the resistance of the films.

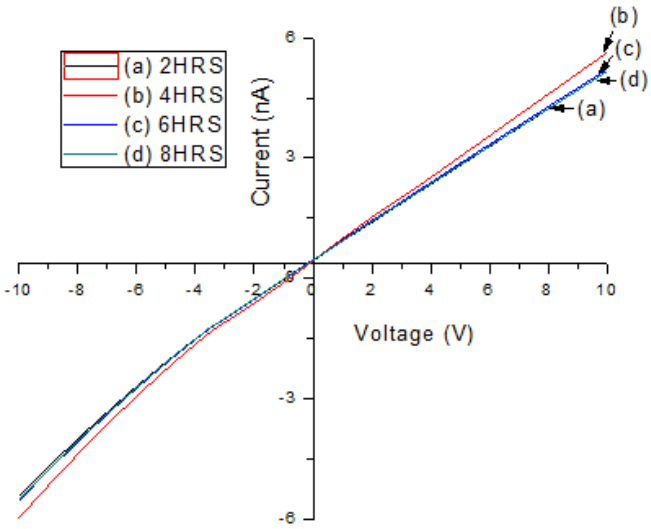


Fig:2. *I-V* curve of multilayer ZnO/MgO films.

Fig.3 shows the influence of immersion time to the resistivity. It reveals that the resistivity was decreased when the immersion time reach at 4 hours where the resistivity value is about 14.29 kΩ.cm. However, the resistivity value was then increased up to 20.0kΩ.cm for 8 hours immersion time.

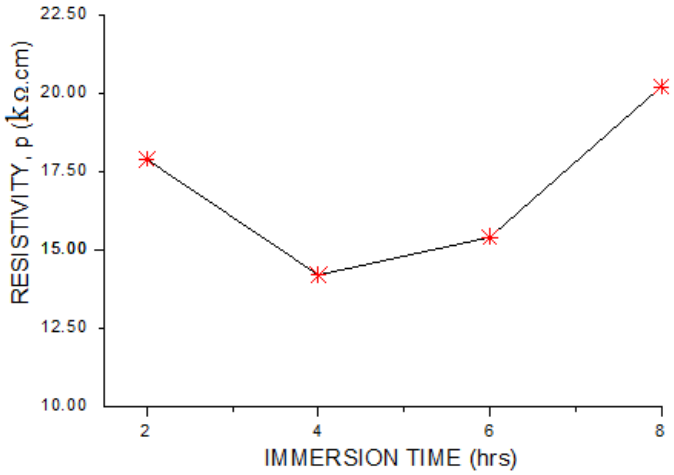


Fig 3. The graph of resistivity versus immersion time.

At 4 hours immersion time, the resistivity value was found to decrease because of the small amount of segregation of grain boundry that causes carrier mobility increase. This was proven by FESEM image on Fig.5(b) that the film morphology is more compact and uniform which in low carrier scattering. When immersion time was above than 4 hours, the resistivity value were increase due to the thicker thickness produced for the prepared multilayer ZnO/MgO. This is prove by the equation (1) where the resistivity are directly proportional to the thickness of thin films. Recently, Hong-ming Zhou et.al, concluded that the resistivity value was depend on the mobility of the carriers that caused by the grain boundry [10]. Toshinori Tagawa et.al on the aothe hand had been concluded on their research that, the resistivity became larger with increasing the thickness of film [11].

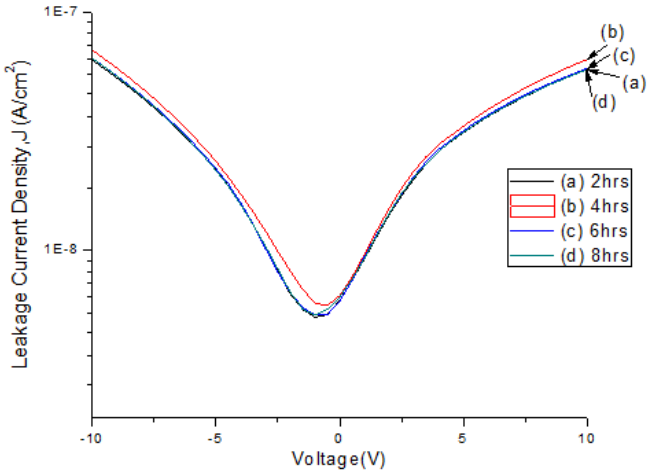


Fig.4: Leakage current density versus voltage.

The leakage current behavior was calculated using following equation:

$$J = I / A \qquad (2)$$

Where, *I* is current and *A* is area electrode. Fig.4 shows that the graph of leakage current density of prepared multilayer ZnO/MgO for -10V to 10V. From the Fig.4, *J* increases as the immersion time increases from 2 to 4 hours. However, as immersion time greater than 4 hours, *J* was decreases. This is because from 2 to 4 hours, the film starting to restructure and the particle are in compact nanostructure on the surface which were influenced by the increasing of carrier mobility as shows on Fig.5(b).

STRUCTURAL PROPERTIES

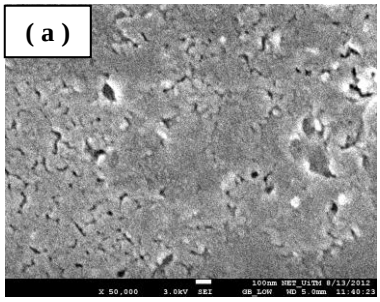
Structural properties of the deposited multilayer ZnO/MgO were characterized using surface profiler, field emission scattering electron microscope and also atomic force microscopy. The obtained thickness and roughness of the prepared multilayer ZnO/MgO was tabulated in Table 1 and it revealed that immersion time was directly proportional to the thickness where as the immersion time increases from 2 hours to 8 hours, the thickness of the films became thicker. As stated by S. Kuai et.al, high immersion duration results in thicker surface which due to the higher filling fraction [12].

TABLE 1: The thickness and roughness of the deposited multilayer ZnO/MgO.

IMMERSION TIME (hrs)	THICKNESS (nm)	ROUGHNESS(nm)
2	528.57	21.555
4	535.95	64.342
6	587.95	57.062
8	594.93	46.102

In Table 1 also reveals that the roughness of prepared multilayer ZnO/MgO are drastically increase for film immerse at 2 to 4 hours. However, above 4 hours immersion time the roughness of the prepared films start to reduce from 64.342nm to 46.102nm.This is because of changes in carrier grain size which at 2 to 4 hours the film was starting the restructure, so the roughness is higher due to its growth of the particles. However, at 4 to 8 hours the roughness is decrease bit by bit, because at this period the film structure is became agglomerate structure. This reason are proven by the images of doing FESEM.

FESEM images in Fig.5 show that the change in immersion time result in surface modification of the deposited multilayer film. We observed that at 2 hours immersion time the particle not growth well, but at 4 hours immersion time, the film start to restructure and it form flake-like structure. For multilayer films immersing at 6 to 8 hours, the particles produced was in agglomerate structure with some porosity as compared to others.



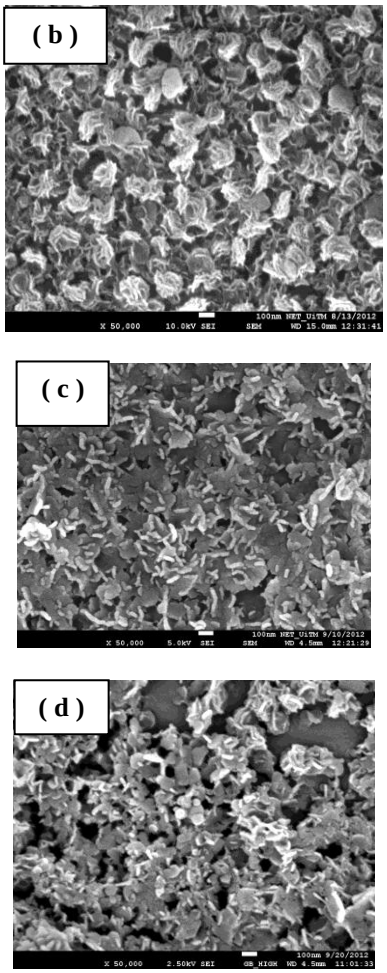


Fig.5. Surface morphology of multilayer ZnO/MgO for vary of immersion time. (a) 2 hours, (b) 4 hours, (c) 6 hours (d) 8 hours.

IMPEDANCE ANALYSIS

Impedance spectroscopy analyzer was used to measure the relative permittivity dielectric constant, *k* for the prepared multilayer ZnO/MgO films. From Fig.6, it shows that the dielectric constant of the prepared multilayer ZnO/MgO was frequency dependence. At low frequency region (up to 100kHz), *k* drastically decreased with frequency. This is because of the inhomogeneities in the structure due to the space charge polarization [13]. At the high frequency region, *k* is became constant with frequency. In this case, it causes by no space charge polarization where dipoles fail to follow the change in electric field and there is no dispersion [13]. It also shows that *k* increase from 7.3 to 7.5 for sample immerse at 2 hours and 4 hours respectively. Besides that, at 4 hours above of immersion time, the *k* decrease from 7.5 to 6.5. This causes by the structure of the film which at 4 to 8 hours the structure had porosity at certain area. The increase of porosity will decrease the *k*. Due to research that state that the low value of *k* can be produced by having a porous structure [13]. However, *k* value at 4 hours immersion time was the highest compared to others. This is because of the formation of flakes like structure in the film which increase the polarization in the film.

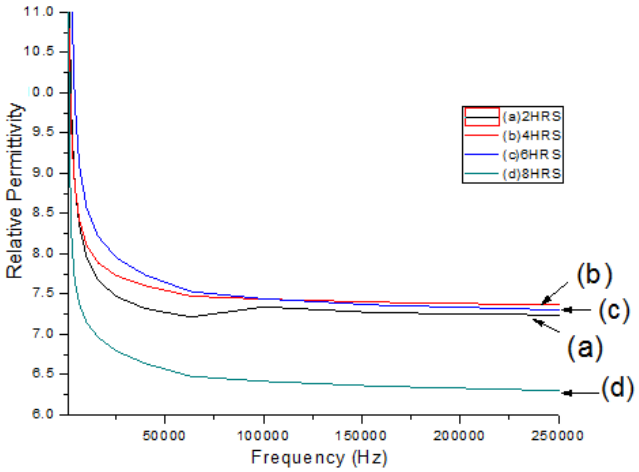


Fig 6. Dielectric constant of the multilayer ZnO/MgO.

CONCLUSION

Multilayer ZnO/MgO were successfully prepared using a low cost method which is immersion method deposition. The varied deposition parameter was in terms of immersion time which is from 2 to 8 hours with 2 hours interval. The properties of prepared multilayer films were observed in electrical behaviour, structural properties and impedance analysis. From the electrical characteristics obtained, the resistivity and leakage current values varied with immersion time. The resistivity values obtained were in a range of 12.50 to 20.00kΩ.cm while leakage current was found to be around 10^{-8} A.cm^{-2}. Variation in resistivity and leakage current values were caused by charges in the surface morphology (uniformity and porosity) of the prepared multilayer films. The multilayer film with 4 hours immersion time was suggested suitable to be used as dielectrics because it is high in resistivity and good in leakage current. From impedance analysis, the *k* value for this film was highest compared to others that cause by formation of flakes like structure in the film which increase the polarization in the film.

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