Effect of Nano-Filler Loading on the Dielectric Layer Properties

Aimi Bazilah Binti Rosli
NANO-ElecTronic Centre (NET),
Faculty of Electrical Engineering ,
Universiti Teknologi MARA Malaysia,
40450 Shah Alam, Selangor, Malaysia
amy_limited@hotmail.com

***Abstract*— This paper reports on the effects of the nano-filler ranging from 0.0-3.0 wt% on the dielectric layer properties of magnesium oxide film by sol-gel spin coating technique. The dielectric layer properties are studied in terms of electrical, relative permittivity and it's morphology. The electrical and permittivity characteristic were characterized using 2-pointprobe *I-V* measurement and impedance spectroscopy while its morphology and topology is characterized using field emission scanning electron microscopy (FESEM) and Atomic force microscopy(Park system-XE 100) respectively. Relative permittivity of MgO films are decrease while the resistivity is increasing as the MgO nano-filler content is increasing. The best weight percentage of nano-filler is 1.0 wt%. The increases of the nano-filler content greater than 1.0 wt% resulted in high porosity.**

***Keywords-component; Nano-filler, Magnesium Oxide, spin coating, Dielectric, Morphology*.**

I. INTRODUCTION

Magnesium Oxide has been used as a dielectric material instead of SiO_2 for both low[1] and high power applications devices[2]. This alternative material can ensure an adequate large band offsets with Si, ranging from 7.3eV to 7.8eV[1, 3]**.** The growth method and annealing conditions are influential in determining the dielectric permittivity κ, generally ranging from 6.7 to 10 [3, 4] for magnesium oxide. Large power dissipation can be achieved by using magnesium oxide due to its high thermal conductivity[2]. In addition, chemical inertness of magnesium oxide also facilitates to minimize the formation of an interfacial layer in contact with Si [1].

Nanotechnologies are available in a lot of domain since they are a great source of innovation. It has been reported that a few percent of nano-filler can improved mechanical characteristic [5], permeability characteristic[6] and electrical properties [7].Although the inclusion of nano-filler could lead to an excellence thin film properties, the source of the improvement is still unclear [8]. There is also lack of investigation on the effect of MgO nano-filler on its morphology.

Therefore, in this research, the nano filler magnesium oxide films were sequentially grown on glass substrates using sol-gel spin-coating technique. The weight percentage of MgO nano filler was systematically studied to investigate its effect on dielectric layer properties of the MgO films.

II. EXPERIMENTAL DETAILS

Nano-filler magnesium oxide films were prepared by sol-gel spin-coating method. Prior to the deposition process, the substrates were subjected to cleaning process in ultrasonic bath for 10 min at 50 ˚c using acetone, methanol and deionizer water (DI) to remove dirts and contaminants.

In this research, nano-filler magnesium oxide solution was prepared using magnesium acetate tetrahydrate $(CH_3COO)_2$ as a precursor,Ethanol (C_2H_5OH) as a solvent, Nitric Acid (HNO_3) as the stabilizer and nano MgO powder as the nano-filler. The nano-filler magnesium oxide powder were varied between 0.0-3.0 wt%. The solution was then sonicated at 50 ˚C for 20 minutes and stir-heated at 80 °C for 3 hour to yield a clear and homogeneous solution. The solution was then under goes aging process at room temperature and kept for 24 hour to achieve complete reaction prior to deposition process.

Nano-filler magnesium oxide solution was spin-coated on glass substrate at 3200 rpm for 30 second and dried at 200 ˚C in thermal furnace for 10 minutes. In order to achieve the desired film thickness the spin-coating process was carried out for 10 times. After the coating process, the films were finally annealed at 500 ˚C for 1 hour.

The growth of nano-filler MgO films was measured using Surface profiler (VEECO). I-V measurement was conducted by 2-point probes (Solar Simulator).The dielectric constant of nano-filler MgO films were measured using impedance spectroscopy (Solatron SI 1260-1296). The morphology and topology of nano-filler MgO films were examined using field emission scanning electron microscopy (FESEM) and Atomic force microscopy(Park system-XE 100). The deposition and characterization process are summarized in the flow chart shown in Fig 1.

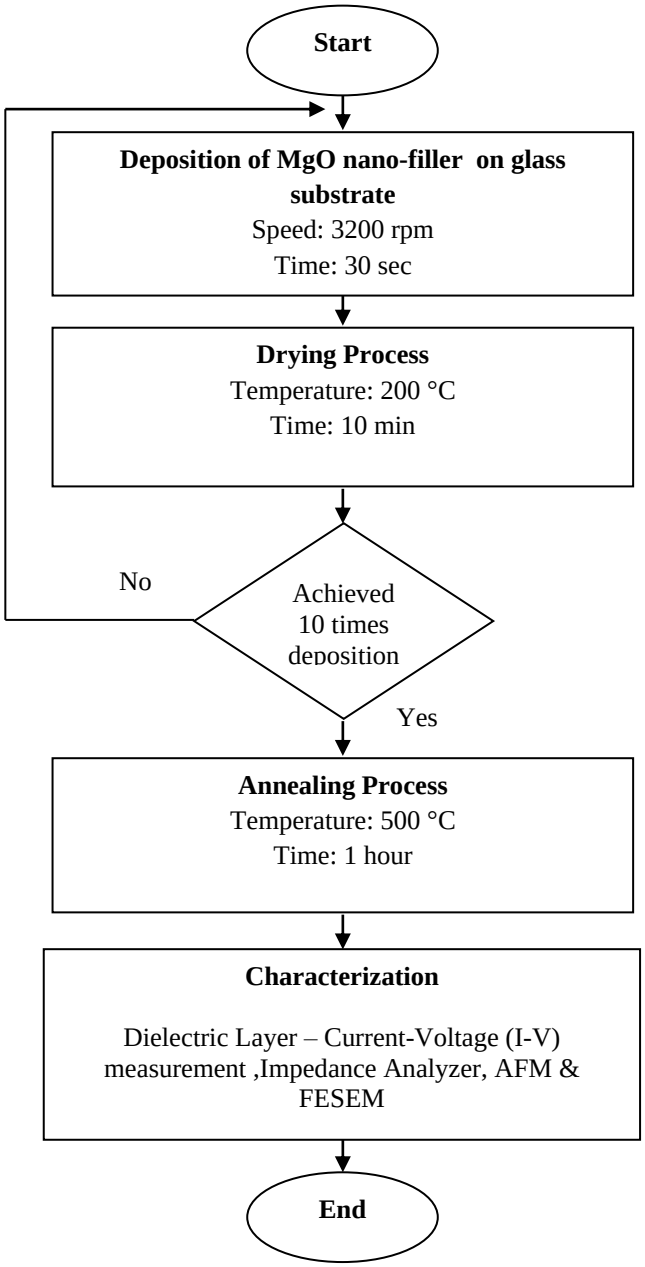


Figure 1: Preparation of MgO films at different nano-filler loading using sol-gel spin-coating technique.

III. Results and Discussion

A. *I-V Measurement*

Fig. 2 shows the I-V characteristic of deposited MgO films at different weigh percentage of nano-filler. From the slope of the graph, it can be seen that increasing of nano-filler loading ratio caused the resistance to be increased.

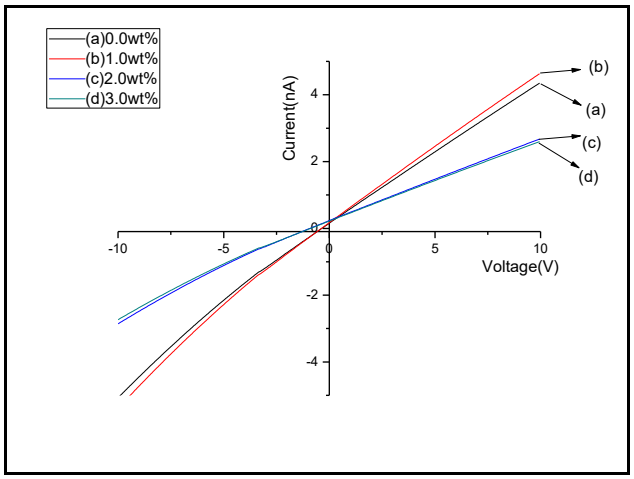


Figure 2: I-V curves of deposited MgO films at different nano-filler loading ratio

From the I-V measurement, the electrical resistivity, ρ was determined by using the following equation:

$$\rho = \frac{1}{\sigma} = \frac{Rwt}{l} (\Omega.\mathrm{cm}) \quad \text{----------} \rightarrow (1)$$

where *w* and *l* are the width and length of metal contact, *t* is the film's thickness, and *R* is the film's resistance.

Figure 3 shows the plotted result of resistivity and also conductivity of deposited MgO at different nano-filler loading ratio. From the plotted graph, it can be seen that the resistivity increases as the Mgo nano-filler increases. The values of resistivity increased from 47.7×10^3 to 525×10^3 Ωcm for 0.0 wt% to 3.0 wt% of nano-filler loading respectively.

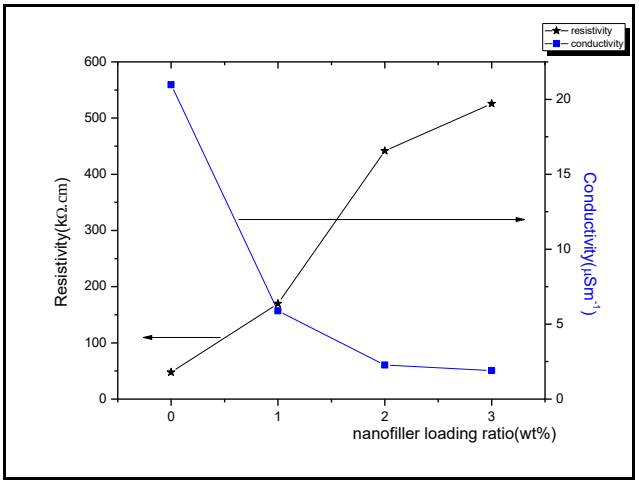


Figure 3: Resistivity and conductivity of MgO films at different nano-filler loading ratio

As suggested by Y.Murakami et.al, the resistivity property of thin films is related to the charge mobility and

charge density. Lack of charge mobility and charge density resulted in greater resistivity [9]. Besides that, as describe in equation 1, it could be said that resistivity is linearly dependent on the thickness of the film. As summarized in table 1, as the weight percentage of the nano-filler increases, the thickness of the films increases that contributes to the high resistivity value.

The relationship between resistivity and conductivity is shown by the following equation:

$$\sigma = \frac{1}{\rho}. \qquad \text{----------} \rightarrow (2)$$

By referring to equation (2) the conductivity of MgO nano-filler films is a reciprocal of its resistivity and it is proven by the graph shown in Fig.3,where the value of conductivity decreased from 21.0×10^6 to 1.9×10^6 Sm^{-1} for 0.0 wt% to 3.0 wt% nano-filler.

B. Relative permitivity

Fig. 4 shows the plotted result of relative permittivity versus frequency in range a range of 0-70 kHz for different weight percentage of nano-filler measured by impedance spectroscopy. At low frequency region (0-10 kHz), the value of relative permittivity is drastically decreased. However, a constant value of relative permittivity is achieved when frequency start to reach 25 kHz. The measured values are 8.18, 7.12, 6.92, 7.22 for nano-filler loading of 0.0, 1.0, 2.0 and 3.0 wt%.

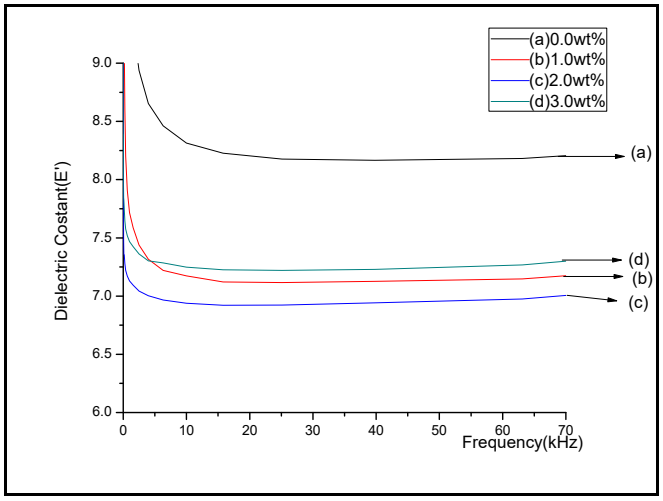


Figure 4:Plot of dielectric constant versus frequency at different weight percentage nano-filler

The graph shows that as the nano-filler loading increases, the relative permittivity decreases. As reported by Zhao et.al, the value of relative permittivity is morphology dependent, suggesting that the increasing in surface roughness leads to the formation of low relative permittivity films [10].Besides that, the reduction in the relative permittivity is also due to films porosity [11].

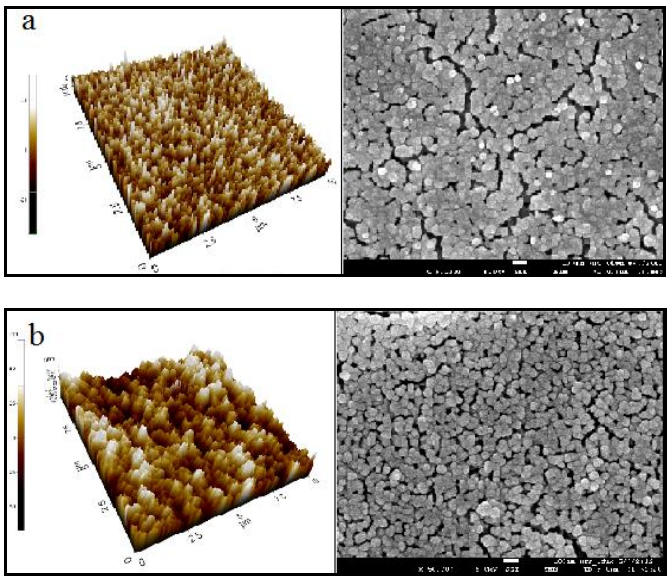
C. Surface Topology and Morphology

Table 1 shows the thickness of nano-filler MgO films deposited using different weight percentage of MgO nano-filler. The thickness of the films was measured by surface profiler (VEECO). As summarized in table 1, greater weight percentage of MgO nan0-filler resulted in thicker MgO films. The thickness of MgO films with 0.0 -3.0 wt% nano-filler increases from 140.76 nm to 1054.07 nm. As suggested by C. Ye et.al, increases in the thickness of films is due to increases of growth rate of film [12].

TABLE 1 AVERAGE THICKNESS NANOFILLER MGO THIN FILMS AT DIFFERENT NANO- FILLER LOADING RATIO

Weight Percentage (wt%)	Average of Thickness (nm)
0.0	140.76
1.0	359.10
2.0	680.15
3.0	1054.07

Figure 5 shows the topology and structural properties of deposited MgO films using different nano-filler loading ratio. As can be seen in figure 5, the roughness of MgO films is increased with increasing of Mgo nano-filler weight percentage. The roughness value of MgO films for 0.0 wt%, 1.0 wt%, 2.0 wt% and 3.00 wt% are 6.812, 15.283, 9.299, 9.938 nm respectively. As reported by Thomas Andritsch, the size of agglomeration tends to increase with increase the MgO nano-filler content that will subsequently form rougher film [13].It is also suggested that the incomplete penetration of MgO nano-filler increase the roughness of MgO film [14].



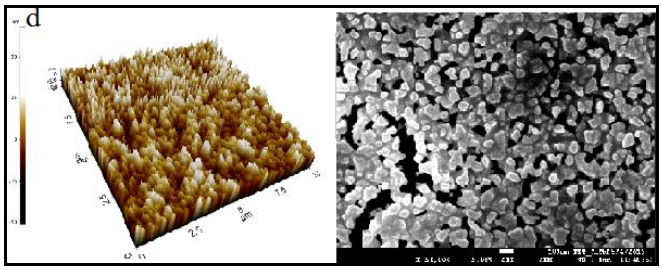


Figure 5:AFM and FESEM image of MgO film at different nanofiller of (a) 0.0 wt% (b) 1.0 wt% (c) 2.0 wt% and (d) 3.0 wt%

The FESEM images in fig.5 shows that the MgO films without nano-filler produced small particles with some porosity. However, the addition of MgO nano-filler at 1.0 wt% resulted in larger particle size, uniform and nonporous in structure. Nevertheless, as the nano-filler loading reached 2 wt%, the surface morphology become rougher, large grain boundary and high in porosity. Thus it is suggested that these MgO films are not suitable for capacitor application due to high porosity. As reported by Dong Tang et.al, when the MgO nano-filler increases, the MgO films were relatively compact. The nitric acid which acts as stabilizer cannot stabilize the MgO particle growth and increase the porosity form [15].

IV. CONCLUSION

MgO films at different MgO nano-filler weight percentage loading have been grown successively on glass substrates using sol-gel spin coating method. The MgO films with nano-filler loading leads to the variations of electrical, relative permittivity and morphology properties of MgO films. The best prepared MgO film was at 1.0 wt% where particle size and thickness of MgO films are 64.4 nm and 359.10 nm. The value of the resistivity and dielectric constant of MgO thin film at 1.0 wt% is 170 $x10^3 \Omega$.cm and 7.25 respectively. It is also good in morphology which are uniform, small particles (64.4 nm) and less porosity which good for capacitor application.

V. FUTURE RECOMMENDATION

In the future, the development of MgO thin film can be improved by using different filler such as polymer or ceramic. Besides that, deposition of thin film in multilayer form is preferable to enhance the thin film morphology properties.

ACKNOWLEDGMENT

The author are grateful to supervisor, Miss Raudah Abu Bakar, PhD student, Habibah Zulkefle and all the members of NANO-ElecTronic Centre (NET), NANO-SciTech Centre, (NST) and Faculty of Electrical Engineering, UiTM Shah Alam for the assistance given to ensure the success of this work.

REFERENCES

[1] L. Yan, C. M. Lopez, R. P. Shrestha, E. A. Irene, A. A. Suvorova, and M. Saunders, "Magnesium oxide as a candidate high-k gate dielectric," *Applied Physics Letters*, vol. 88, pp. 142901-142901-3, 2006.

[2] A. Posadas, F. J. Walker, C. H. Ahn, T. L. Goodrich, Z. Cai, and K. S. Ziemer, "Epitaxial MgO as an alternative gate dielectric for SiC transistor applications," *Applied Physics Letters*, vol. 92, pp. 233511-233511-3, 2008.

[3] O.Madelung,"II-VI and I-VII Compounds; Semimagnetic Compounds," *Landolt-Börnstein, New Series Group III,* vol. 41B, 1999.

[4] E. Miranda, E. O'Connor, G. Hughes, P. Casey, K. Cherkaoui, S. Monaghan, R. Long, D. O'Connell, and P. K. Hurley, "Soft breakdown in MgO dielectric layers," in *Reliability Physics Symposium, 2009 IEEE International*, 2009, pp. 688-691.

[5] H. N. P.Reichert, S.Kllinkle,R.Brandsch,R.thoman and et.al, "Polyproplene organoclay nanocomposite formation:Influence of compatibilizer functionality and organoclay," *Macromolecular Material and Engeneering*, vol. 275, pp. 8-17, 2000.

[6] M. T. G.Gorrasi, V.Victoria, D.Kaempfer, "Transport properties of organic vapors in nanocompositesof organophilic layered silicate and syndioctactic polyprone," *Polymer*, vol. 44, pp. 3679-3685, 2003.

[7] D. K. C.Zilg, R.Thormann,R.Mulhaupt, "Electrical Properties of polymer nanocompososites based upon organicphilip layered silicates," *IEEE CEIDP*, pp. 46-49, 2003.

[8] K. Ishimoto, E. Kanegae, Y. Ohki, T. Tanaka, Y. Sekiguchi, Y. Murata, and C. Reddy, "Superiority of dielectric properties of LDPE/MgO nanocomposites over microcomposites," *Dielectrics and Electrical Insulation, IEEE Transactions on*, vol. 16, pp. 1735-1742, 2009.

[9] S. Okuzumi, Y. Murakami, M. Nagao, Y. Sekiguchi, C. C. Reddy, and Y. Murata, "DC Breakdown Strength and Conduction Current of MgO/LDPE Composite Influenced by Filler Size," in *Electrical Insulation and Dielectric Phenomena, 2008. CEIDP 2008. Annual Report Conference on*, 2008, pp. 722-725.

[10] Zhao, Y., Toyama, M., Kita, K., Kyuno, K., and Toriumi, 'Moisture absorption-induced permittivity deterioration and surface roughness enhancement of lanthanum oxide films on silicon', *Applied Physics Letters*,pp. 072904-072903,2006.

[11] K. M. Mikhail R Baklanov, "Porous Low Dielectric Constant Materials for Microelectronics," *Philosophical Transactions of the Royal Society A: Mathematical, Physical and Engineering Sciences*, vol. 364, pp. 201-215, 2006.

[12] C.Ye, X.M.Teng, and G. Lia, "Optical properties of MgO-TiO2 amorphous composite films," *Journal of Applied Physics*, 2007.

[13] T. Andritsch, R. Kochetov, P. H. F. Morshuis, and J. J. Smit, "Dielectric properties and space charge behavior of MgO-epoxy nanocomposites," in *Solid Dielectrics (ICSD), 2010 10th IEEE International Conference on*, pp. 1-4.

[14] R. O. Fatima S. Aguilera, Estrella Osorio, Pedro Moura, and Manuel Toledano, "Wetting ability of an acetone/based etch&rinse adhesive after NaOCl-treatment," *Biomaterials and bioengineering in dentistry*, p. 1.

[15] T. Dong, D. Rui-xue, Z. Yang, and N. Hong-jun, "Effect of MgO Dopant on Structure and Performance of Mesocarbon Microbead," in *Measuring Technology and Mechatronics Automation (ICMTMA), 2011 Third International Conference on*, pp. 7-10.