Sol-Gel Derived Nano-Magnesium Oxide: Influence of Drying Temperature to the Dielectric Layer Properties

Z Habibah1,*, K A Yusof1, L N Ismail1, R A Bakar1 and M Rusop1,2

1NANO-ElecTronic Centre (NET), Faculty of Electrical Engineering, Universiti Teknologi MARA, 40450 Shah Alam, Malaysia
2NANO-SciTech Centre (NST), Institute of Science, Universiti Teknologi MARA, 40450 Shah Alam, Malaysia

*E-mail: habibahzulkefle@yahoo.com

Abstract. Nano-magnesium oxide (Nano-MgO) had successfully deposited on glass substrate using sol-gel spin coating technique at different drying temperature (100, 200 and 300°C). The dielectric layer properties of deposited nano-MgO films were investigated in terms of its resistivity, leakage current density, relative permittivity, topology and morphology respectively. Changing in drying temperature resulted in variation of nano-MgO films properties and film dried at 200°C has compact, uniform, less porous and in nano size that lead to the enhancement in relative permittivity value.

1. Introduction
Over past decade, magnesium oxide (MgO) which is inorganic compound with NaCl structure had attracts great attention in many applications [1-3]. MgO also is a suitable candidate to replace existing dielectric layer material which is silicon dioxide (SiO2) due to its unique properties such as high dielectric constant (9.8), wide band gap (7.8eV) and high breakdown field (12MV/cm) [4, 5]. MgO films can be prepared using several methods such as physical deposition and chemical deposition method and among other methods, chemical deposition via spin coating technique has its own advantages which are large area deposition, simple and also cost effective. There are quite number of researches had been done in order to investigate the effect of deposition parameters on the properties of MgO films [6, 7]. Heat treatment which includes drying temperature is the important deposition parameters that will influence the production of high quality MgO films. Since the effects of drying temperature on the properties of sol-gel derived MgO thin films is not well understood yet, it is therefore a need to carry out a details study on this matter. This paper reports on the effect of drying temperature on electrical, structural and dielectric properties of nano-MgO films deposited using sol-gel spin coating method.

2. Experimental Procedure
During MgO solution preparation, magnesium acetate tetrahydride ((CH3COO)2 Mg*4H2O) was dissolved in ethanol (C2H5O) with small amount of nitric acid (HNO3) which act as the stabilizer. The solution was then sonicated for 20 mins and stirred at 80°C for 3 hrs. Next, MgO solution was undergoes aging process at room temperature for 24 hrs. Afterward, the prepared MgO solution was deposited on cleaned glass substrate where the deposition parameter was set at 3200 rpm for 30 secs.
Next the deposited films were dried at different temperatures (100, 200 and 300°C) for 10 mins. The deposition and drying processes were repeated for several times to achieve desired film thickness and finally the deposited nano-MgO films were annealed at 500°C to restructure the MgO particles.

There are three main characterizations have been done in this research work which are electrical, dielectric and structural characterizations. Electrical characterization was conducted using two-point probes $I-V$ measurement (Bukoh Keiki) while dielectric properties were determined by impedance analyzer (Solartron S1 1260A-1296). Surface topography and morphology of the deposited nano-MgO films were observed using atomic force microscopy, AFM (Park System, XE-100) and field emission scanning electron microscope, FESEM (JEOL JSM 7600F) respectively. The thickness of deposited films was determined using surface profiler (Veeco).

3. Results and Discussion

3.1. Electrical Properties

Fig. 1a and 1b shows the electrical behaviour of the deposited MgO films in terms of resistivity, conductivity and leakage current density respectively. As shown in Fig. 1a, the resistivity values were increased as the drying temperature increased. Rise in resistivity value was due to the small grain and high porosity [8] film that lead to the increase of carrier scattering and low carrier mobility [9]. Besides, it was also due to thicker film produced as the drying temperature increased. The resistivity values obtained are $1.08 \times 10^4$, $1.28 \times 10^4$ and $1.74 \times 10^4 \ \Omega \cdot \text{cm}$ for films dried at 100, 200 and 300°C respectively which are in same magnitude with work done by S. Lee et.al which their resistivity was around $10^4 \ \Omega \cdot \text{cm}$ [10].

![Figure 1](image.png)

Figure 1. Electrical properties in terms of (a) resistivity and conductivity, (b) leakage current density of MgO films dried at different temperatures.

From Equation 1, it clearly shows that the conductivity is inversely proportional to the resistivity value and it is proven by conductivity graph shown in fig. 1a.

$$\sigma = \frac{1}{\rho}$$  \hspace{1cm} (1)

where $\sigma$ is the conductivity and $\rho$ is the resistivity of deposited MgO films. The leakage current density, $J$ was plot in Fig. 1b where the value was below than $10^{-9}$ at applied voltage of 2V which was five order of magnitude lower than common dielectric material (SiO$_2 \sim 4 \times 10^{-4}$ A/cm). Moreover, it also reveals that there were slightly decreased in the $J$ values as the drying temperature increased from
100 to 300°C. This was due to the reduction in grain boundary area produced in the film that reduced a direct path for the carrier to pass through the film.

3.2 Relative Permittivity Characteristic

The dielectric properties of MgO films with the influence of drying temperature were measured at frequency range from 0 to 40 kHz and their relative permittivity, $\varepsilon_r$ value was shown in Fig. 2. High $\varepsilon_r$ values obtained for all samples at low frequency region (below 10 kHz) and its value drastically decreased as the frequency increased. As stated by Shaikh et. al, decrease in $\varepsilon_r$ value with frequency is due to inhomogeneous film that cause the charge trapped at the interface structure [11]. At high frequency region (above 40 kHz), $\varepsilon_r$ value of each film decreased to a constant value which due to non-polarized charges [12].

![Figure 2. Relative permittivity, $\varepsilon_r$ as the function of frequency at various drying temperature.](image)

In addition, Fig. 2 also revealed that $\varepsilon_r$ value increases as the drying temperature increased from 100 to 200°C. However, as the temperature increase to 300°C, leads to the reduction of $\varepsilon_r$ value. The variation in $\varepsilon_r$ value for various drying temperature is related to the structural changes such as grain size and surface roughness of the film [13]. From the FESEM images (Fig. 3), it clearly shows that the MgO film dried at 200°C has a compact, uniform, small roughness and less porosity which contributes to the high $\varepsilon_r$ value produced.

3.3 Structural Properties

Deposited MgO thin films growth was characterized in term of their thickness using surface profiler and their values were tabulated in Table 1. As can be seen, rise in the drying temperature leads to the formation of thicker film.

| Drying Temperature (°C) | Average Film thickness (nm) | Root Mean Square, RMS Surface Roughness (nm) |
|-------------------------|-----------------------------|---------------------------------------------|
| 100                     | 137.49                      | 44.360                                      |
| 200                     | 192.97                      | 20.660                                      |
| 300                     | 224.72                      | 21.543                                      |

As stated by G. Hu et. al, film surface roughness is define as the measure of the surface texture of the film in terms of their RMS value of the surface height profile of the film [14]. The surface roughness of the deposited MgO films was determine using atomic force microscopy, AFM with scan
area of \((10 \times 10)\, \text{m}^2\). From the tabulated data, the RMS surface roughness value for MgO films shows an improvement in film uniformity as the drying temperature increased from 100 to 200°C, however it value slightly increase for film dried at 300°C.

Fig. 3 shows the FESEM images of deposited MgO films with magnification and voltage of 30k and 5kV respectively. From the images (Fig. 3), it indicates that the drying temperature affect the MgO film morphology. Increased in drying temperature from 100 to 200°C resulted to the formation of uniform structure with few defects. Nevertheless increase in drying temperature to 300°C produced a non-uniform and cracks film. This was due to the magnesium evaporation process [15].

MgO film dried at 200°C gives better electrical and dielectric properties compared to others which are related to it uniform and less porosity on the film. Besides particle size of MgO film produced was in nano-meter size (Fig. 3d) which in a range of 36.4nm to 73.1nm. P. K. Ghosh et.al stated that, the enhancement in polarization that lead to the increased in the \(\varepsilon_r\) value can be obtained with nano-meter particle size of the film due to its high surface area per unit volume [16]. Since the film dried at this temperature has good film properties, the EDS analysis (Fig. 3e) was then performed to confirm magnesium, Mg and oxygen, O\(_2\) element are exist in the film. Other than that, silicon, sodium and platinum element detected are due to the glass substrate and coating respectively. Thus, the film dried at this temperature is suitable to be used as a dielectric layer.

**Conclusion**
Sol-gel spin coating method was used as deposition technique of prepared nano-MgO films. The drying temperature was varied at 100, 200 and 300°C in order to investigate the effect of drying temperature towards dielectric layer properties of nano-MgO films. Dielectrics properties of deposited nano-MgO films were investigated in terms of its electrical, relative permittivity and structural properties. Changing in drying temperature applied resulted in the variation of nano-MgO film properties. Nano-MgO film dried at 200°C was suggested suitable to be used as dielectric layer due to
its high resistivity ($12.8 \times 10^4 \Omega \cdot \text{cm}$) and low leakage current density ($\sim 10^{-9} \text{A} \cdot \text{cm}^{-2}$). Besides, it also has merit in surface properties such as dense, homogeneous, less porous and nanometer particle size which resulted increment in relative permittivity.

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