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Influence of Molar Concentration: Sol-Gel Synthesized Magnesium Oxide Thin Films for High Power Light Emitting Diode Thermal Management

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Abstract Sol-gel technique was used to synthesize 0.6 M and 0.8 M molar concentrations of magnesium oxide thin on aluminium substrate and enforced into heat spreader material in thermal management of lighting devices. X-ray diffraction analysis confirm the films to be polycrystalline cubic in nature with (200), (220) and (222) orientations. 20 layers from 0.8 M showed higher intensity from the X-ray analysis. Relative thermal conductivity measurement of 20 layers from 0.6 M showed a value of (26.3 W/mK) which appears to improve over other layers. At 700 mA operating current, 20 layers from 0.6 M showed higher junction temperature difference of 37.16 °C as well as higher thermal resistance difference of 5.9 K/W when compared to bare aluminium substrate. All coated films are within the accepted color correlated temperature safe operating range (< 3300 K) of light emitting diodes, addition to this, 20 layers MgO thin films from 0.6 M displayed a recommendable illuminance as well as color correlate temperature performance from optical characterization. Atomic force microscope surface analysis for both molar concentrations showed an improvement in surface smoothness over bare substrate. Overall, magnesium oxide thin films can be perfectly enforced as heat spreader for high power light emitting diodes.

1. Introduction

Light emitting diode (LED) have taking the place of conventional light sources in street lighting, traffic signals, residential, automobile, outdoor and advertisement board lighting [1-3]. Numerous advantages of LEDs, such as, miniaturization of their package, low-voltage power supply demand for their operation, no ultra - violet (UV) or infrared light emission, environmentally friendly, output energy efficient, long lifespan, flexible display design, resistant to shock, resistance to vibration as well as impact, all these features makes them beneficial over conventional lighting [1, 4]. Despite all those advantages of LEDs, miniaturization of LEDs resulted in generation of excessive heat within the package, thereby resulting in rise of junction temperature, thermal runaway, shorten of the LEDs lifetime, poor efficiency and overall failure of the LEDs [2]. Therefore, need arises to properly address the heat management of the LEDs, so that, the rapid heat generated within them be remove via a suitable thermally conductive path. Doing so will keep the device components in a safe operating temperature condition.

Spin coated MgO thin film is a suitable alternative to replace the existing BN, BeO, AlN, SiC, Al_2O_3, ZnO thin films that are used as thermal interface materials (TIM) or heat spreaders [1, 3], due to some of its excellent properties, among which are; good chemical and thermal stability, electrical insulation, high thermal conductivity (40 W/mK), large band gap of 7.8 eV. The properties include,   dielectric constant (9.8), as well as 12 MV/cm breakdown field and good technology applicability [5, 6]. Findings from other researchers in the same group reported a lower junction temperature (T_j) of 66.30 °C using Al_2O_3 filled epoxy TIM [1], and lower thermal resistance of 13.81 K/W and lower T_j of 47.32 °C from silver doped ZnO thin film TIM [7]. This study is aimed in using MgO thin film as a heat spreader between LED package and heat sink. Doing so will provide efficient high thermally conductive path, complete removal of excessive heat generated within the LED package, reduction in both LED's junction temperature and thermal resistance, solution to LEDs thermal management difficulties, improvement of LED lifespan, brightness, and performance. Various deposition methods were used in synthesis of magnesium oxide thin films, these includes, chemical vapor deposition, spin coating, spray pyrolysis, sputtering technique and vacuum evaporation [5, 8, 9]. Spin coating is used in this work due to its advantages of cost effectiveness, large and difficult area coating, stoichiometry control and uniformity of the films over the substrates during coating [5].

This paper reports the effects of molar concentration, variations in number of layers, film thickness, surface roughness level, thermal conductivity measurements, thermal transient analysis, structural and optical analyses as parameters that will brings about production of impressive MgO thin film heat spreader.

2. Experimental Details
0.6 and 0.8 molarity were used to prepare MgO solution separately in this study. Magnesium acetate tetrahydrate, calculated nitric acid and diethanolamine were dissolved in ethanol. The solution was sonicated at 50 °C. The solution was heated as well as stirred simultaneously for the period of 3 hrs using 60 °C. The solution was then kept for 24 hrs to undergo ageing.
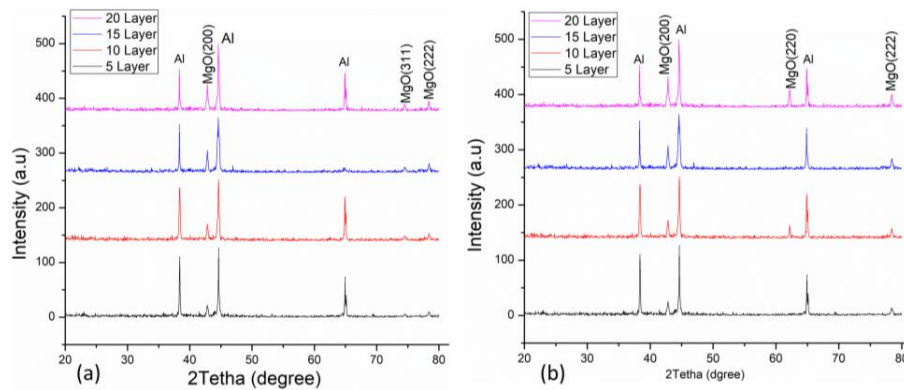
Al5052 substrate was used in this study. Detergent, acetone, methanol along with dionised water were used to clean the substrate. The solutions were then deposited on the cleaned substrates, spun using 3000 rpm and 30 s parameters, followed by drying for 5 min at a temperature of 200 °C. The circle is then repeated for several times (5, 10, 5, 20) in order to have different film thickness. After that, the coated MgO were annealed at a temperature of 500 °C for 2 hrs.

Thermal performance of the MgO thin films as heat spreader for CREE (3.6 W) white LED CXA 1304 was tested by thermal transient tester (T3ster), X-ray diffractor (Bruker D8) was used to study the crystallinity of the films. Atomic force microscope (AFM) (ULTRA objectives surface imaging system, GmbH) was applied to study the morphology of the samples. Thermal conductivity and optical analysis were carried out respectively using HOT DISC method and spectrophotometer.

3. Results and Discussion
3.1. XRD Analysis
X-ray diffraction analysis for both 0.6 M and 0.8 M were recorded. The studies showed the films to be polycrystalline in nature. Diffraction peaks of 42.82°, 74.51°, and 78.44° were recorded for 0.6 M, similarly 42.82°, 62. 16° and 78.44° for 0.8 M. The peaks correspond to crystallographic orientations of (200), (311), (222), and (220) for 42.82°, 74.51°, 78.44°, and 62.16° respectively. (200) crystallographic orientation appears to be the strongest diffraction peak among others.
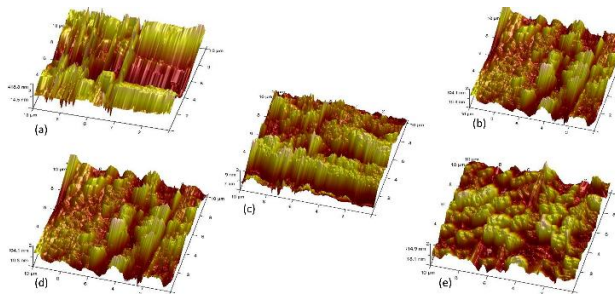
Figure 1. XRD spectra of spin coated MgO thin film, (a) 0.6 M and (b) 0.8 M

Figure 1 show improvement in (200) peak orientations intensity as numbers of MgO film layers increases from 5 to 20, so also as the molar concentration increases from 0.6 M to 0.8 M. Improvement in the peak intensity resulted to appreciable performance of 20 layers of the films from 0.6 M and 0.8 M in heat spreading, these are in same magnitude with research carried out by S. Valaranasu et al. [8], where film with higher intensity performs better.
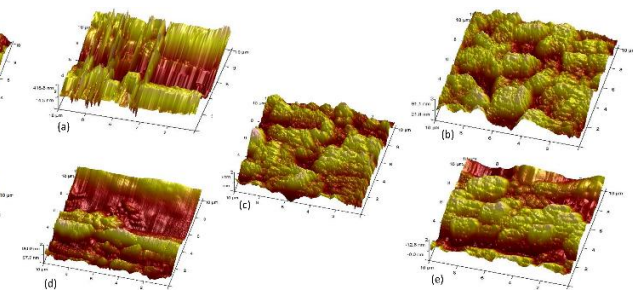
3.2. Thin Film Thickness and AFM Analysis

Thin film thickness is one of the key features of TIMs and heat spreaders. Thin film thickness of various layers of the MgO from 0.6 M and 0.8 M concentration were measured and presented in table 1. The thickness increases as the number of layers increases from 5 to 20 layers, similarly, the thickness increases as the molar concentration increases from 0.6 M to 0.8 M. Films with thickness in the range of 300 nm to 800 nm are expected to give recommendable results [10]. Therefore, 20 layers are expected to be best for the study purpose.

Spin coated MgO thin films surface roughness from 0.6 M and 0.8 M were carried out using AFM machine. Figure 2 and 3 show the 3D surface nature of the analysis. The surface roughness measured (root mean square) values are 79, 81, 113 and 121 nm for 5, 10, 15, and 20 layers from 0.6 M. While for 0.8 M are 61, 94, 116, and 130.2 nm for 5, 10 15 and 20 layers respectively.

Figure 2. AFM images of 0.6 M (a) bare, (b) 5L, (c) 10 L, (d) 15 L and (e) 20 L

Figure 3. AFM images of 0.8 M (a)bare, (b) 5L, (c) 10 L, (d) 15 L, and (e) 20 L

The measured values were summarized and presented in table 1. Table 1 showed that, 20 layers appears to have higher values of surface roughness from both concentrations, these was due to higher crystallite size and agglomeration of particles [5, 11].   Research had proven that, thin films with smooth surface do possess higher thermal conductivity and perform more efficiently [10], however, research carried out here showed that, 20 layers from 0.6 M with surface roughness (121 nm) performs better, this was due to influence of the film thickness and thermal conductivity of MgO (~40 W/mK).

Table 1. Showing size of the films, root mean square values along with thermal conductivity values for spin coated MgO thin films on Al5052.
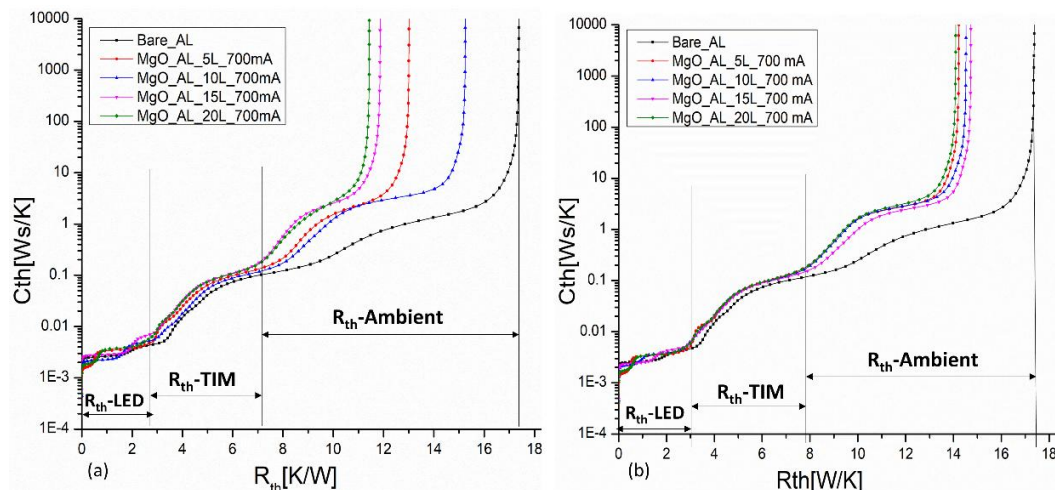
Samples	Thickness (nm)		Root mean square (nm)		Thermal conductivity (W/mk)	
	0.6 M	*0.8 M*	*0.6 M*	*0.8 M*	*0.6 M*	*0.8 M*
Bare			150	150	17.23	17.23
5 Layers	156.8	174	79	61	20.7	19
10 Layers	183	189.91	81	94	18.6	18.4
15 Layers	289.08	296.23	113	116	23.7	25.3
20 Layers	301.7	319.7	121	130.2	26.3	24

3.3. Thermal Conductivity

The bulk thermal conductivity measurement of the MgO thin films was performed using transient plane source (TPS 2500 S) Analyzer. The derived conductivity values are presented in table 1. The table showed that, 20 layers from 0.6 M concentration possesses higher values of the thermal conductivity of 26.3 W/mK. This was due to appreciable thickness and lower surface roughness of 20 layers from 0.6 M compared to that of 0.8 M. Thickness and smooth surfaces plays a significant role in improving thermal conductivity of devices [10].

3.4. Thermal Transient Analysis

A perfect heat spreader is expected to have high thermal conductivity, low thermal resistance (R_{th}) as well as low junction temperature (T_j) values [1]. MgO thin film where spin coated and used as a heat spreader between CREE (3.6 W) LED package and Al5052 substrate. Thermal resistance together with junction temperature measurements of LED mounted on MgO heat spreader were conduct using (350, 500 and 700) mA input currents. Figure 4 represent the analyzed thermal resistance ranges of LED fixed on bare as well as different layers of MgO thin films and powered at 700mA.

Figure 4 showing, analyzed thermal resistance ranges of LED mounted on un-coated substrate and spin coated MgO thin films at (a) 0.6 M and (b) 0.8 M

The figure showed an improvement in performance of the LED by reduction in the R_{th} of the coated films compared to that of the bare substrate.
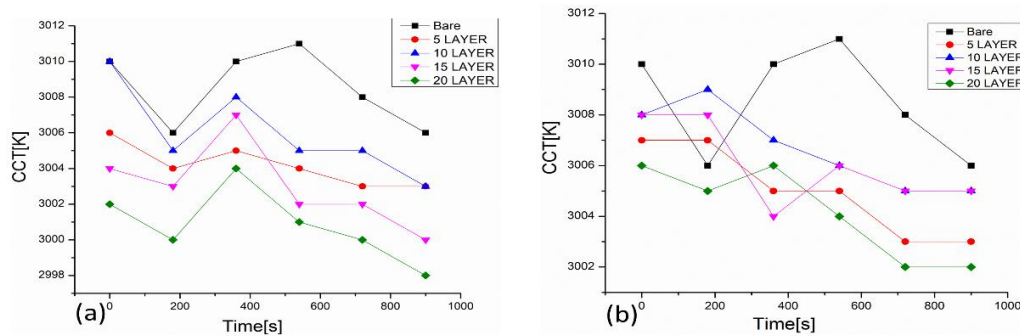
Table 2. shows extracted values of R_{th} and T_j of LED mounted on un-coated substrate and synthesised MgO thin films.

		0.6 M				0.8 M			
	Bare Al	5 L	10 L	15 L	20 L	5 L	10 L	15 L	20 L
Driven Current (mA)									
Thermal Resistance [(R_{th}) (K/W)]									
350	12.4	10.9	11	10	9.8	12	12	12.3	11.7
500	13.3	11.7	11.6	10.8	10.4	12.7	13	13.2	12.7
700	17.3	13	15.2	11.8	11.4	14.2	14.5	14.7	14.1
Junction Temperature [T_j (°C)]									
350	61.9	57.5	56.8	55	54.2	60.8	61	61.5	59.9
500	83.7	77	76.7	72.9	71	81.6	82.6	83.4	81
700	136.3	103.6	123.5	100.9	98.8	116.4	118	119.4	115.2

Table 2 present the extracted values of the transient analysis at all the driven currents. The table showed 20 layers from 0.6 M to have lower R_{th} values in the range of 9.77, 10.43 and 11.44 K/W at 350, 500 and 700 mA respectively. At 700 mA, 20 layers from 0.6 M possess higher R_{th} difference value of 5.9 K/W when compared to bare substrate. This difference appears to be greater than that of 20 layers (3.24 K/W) from 0.8 M and other layers from both concentrations. Similarly, 20 layers of 0.6 M at 700 mA displayed higher T_j difference of (37.44 °C) when compared to other layers and bare substrate. From both R_{th} and T_j of the transient analysis, 20 layers from 0.6 M at 700 mA appears to display higher difference values, these was due to its low surface roughness value, appreciable thickness and higher thermal conductivity compared to 20 layers from 0.8 M. This show more preference of 20 layers from 0.6 M as heat spreader over 20 layers from 0.8 M and other layers towards solving LED thermal management difficulties.
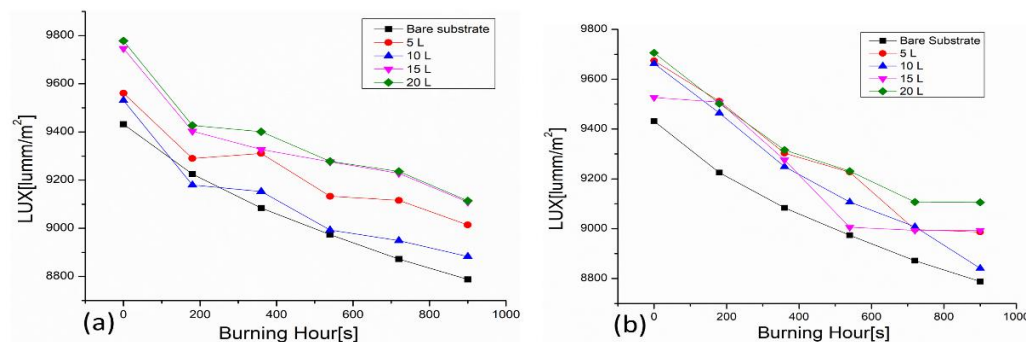
3.5. Optical Analysis
Optical properties for LEDs like Color-Correlated Temperature (CCT) in connection with luminance (LUX) were carried out using hand hold spectrometer. A good TIM or heat spreader are expected to maintain low CCT values to enable them stay within the safe operating range [12]. The measured CCT values at 700 mA for 0.6 M and 0.8 M are presented in figure 5. The overall LEDs CCT performance values should not exceed 2700 – 3300 K, according to the data sheet to avoid degradation of the LEDs. The figure shows that all the samples are within the limit, with 20 layers from both concentrations showing lower values than the bare and other coated layers. However, 20 layers from 0.6 M recorded the lowest values as can be seen from the figure, indicating that, it is more suitable as a heat spreader in providing smooth heat transfer from the LEDs package to the ambient efficiently.

Figure 5 Colour-Correlated Temperature (CCT) images of LED mounted on bare and various layers of MgO at (a) 0.6 M and (b) 0.8 M

Removal of excessive heat generated within the LED package can help the LEDs to always show high illuminance (LUX) an maintaining their brightness all the time [12]. These can only be achieved by using TIMs or heat spreader in between the device package and heat sink. At 700 mA, 20 layers from 0.6 M displayed a remarkable performance in maintaining the brighter light output nature of the LED fixed on it, this was attained through higher values of LUX displayed by the sample when used as heat spreader.

Figure 6 Variation of Lux of LED fixed on MgO thin film as heat spreader at (a) 0.6 M and (b) 0.8 M, all at 700 mA driving current

Considering all the results displayed by 20 layers of 0.6 M from all the analysis carried out, it proved its practical capability of been used as heat spreader toward improving thermal management of LEDs packaging system.

4. Conclusion

500 °C and 2 hours were annealing temperature and annealing period used to synthesized 0.6 M and 0.8 M magnesium oxide thin films using spin coating technique. They were used as heat spreader between CREE 3.6 W light emitting diode and aluminum substrate. XRD analysis revealed all the films were polycrystalline cubic with (200), (220) and (222) orientations. The intensities of the films increase as the number of layers increases from 5 to 20 layers, similarly it increases from 0.6 M to 0.8 M. 20 layers MgO film from 0.6 M was suggested suitable to be used as heat spreader between light emitting diodes package and metal substrate for thermal management of LEDs due to its thermal conductivity (26.3 W/mK), higher thermal resistance difference (5.9 K/W) and higher junction temperature difference (37.16 °C) . Beside these, it showed a lower surface roughness (121 nm) compared to that of 20 layers from 0.8 M and optical performance of LED fixed on it was within the safe operating range (<3300 K), were it showed appreciable performance in both color correlated temperature and illuminance.

References

[1]   Anithambigai P, Dheepan C M K, Mutharasu D, Huong L H, Zahner T, Lacey D and Kamarulazizi I 2016 Potential thermally conductive alumina filled epoxy composite for thermal management of high power LEDs *J. Mater. Sci. Mater. in Electron* **28(1)** p 856–67

[2]   Chakravarthii D M K, Devarajan M, Suvindraj K and Choo C L 2017 Thermal characterization of LEDs mounted on substrates with converging–diverging and diverging–converging channels. *IEEE Trans. Electron Devices* **64(8**) p 3308–15

[3]   Jamaludin N, Anithambigai P, Shanmugan S and Mutharasu D 2014 Performance and thermal analysis of aluminium oxide filled epoxy composite as TIM for LEDs. *Mater. Sci. Res.* **11(1)** p 35-41

[4]   Arik M, Becker C A, Weaver S E and Petroski J 2004 *Thermal management of LEDs: package to system* vol 5187, ed N Ferguson, S Narendran, S DenBaars and J Carrano (Califonia: Unote States International Society for Optics and Photonics) p 64

[5]   Habibah Z, Raudah A, Mohamad R M and Ismail L N 2011 *Molar concentration effect on MgO thin films properties* Proc. IEEE Symp. Ind. Electron Appl. (ISIEA2011) (Langkawi: Malysia) p 468-471

[6]   Habibah Z, Arshad A N, Ismail L N, Raudah A and Mohamad R M 2013 *Chemical solution deposited magnesium oxide films: influence of deposition time on electrical and structural properties.* Procedia 5th BSME Inter. Conf. on Thermal Eng. (Bngladish) vol 56 (Amsterdam: North-Holland/American Elseveir) p 737-742

[7]   Mah J W, Subramani S, Devarajan M and Fauziah S 2017 I*nvestigation of rheological, structural, surface, optical and thermal properties of low temperature produced silver doped ZnO thick film as thermal interface material in lighting application* J. Mater. Sci. Mater. Electron **28** p 10112-121

[8]   Valanarasu S, Dhanasekaran V, Karunakaran M, Vijayan T A, Chandramohan R and Mahalingam T 2014 *Microstructural, optical and electrical properties of various time annealed spin coated MgO thin films* J. Mater. Sci. Mater. Electron **25(9)** p 846–53

[9]   Moses E R A, Nehru L C, Jayachandran M and Sanjeeviraja C 2007 *Spray pyrolysis deposition and characterization of highly (100) oriented magnesium oxide thin films* Cryst. Res. Technol. **42(9)** p 867–75

[10]   Jamaludin J N, Subramani S and Devarajan M 2018 *Thermal and optical performance of chemical vapor deposited zinc oxide thin film as thermal interface material for high power LED* AIMS Mater. Sci. **5(3)** p 402–13

[11]   Hopkins P E 2013 *Thermal Transport across Solid Interfaces with Nanoscale Imperfections: Effects of Roughness, Disorder, Dislocations, and Bonding on Thermal Boundary Conductance.* ISRN Mech. Eng. **2013** p1–19

[12]   Ki-Duck C, Tae H and Sung-Il C 2003 *Improvement of color temperature using independent control of red, green, blue luminance in AC plasma display panel* IEEE Trans. Electron Devices **50(2)** p 359–65