CHEMICAL VAPOR DEPOSITED Al_2O_3 THIN FILM AS THERMAL INTERFACE MATERIAL FOR HIGH POWER LED APPLICATION

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Chemical vapor deposition method was used to synthesize Al_2O_3 thin film and used as thermal interface material for LED application. The Al_2O_3 thin film was deposited on Al substrate at various process time and undergone for post processing (annealing) at 3 different temperatures. The thermal transient analysis of LED was used to estimate the quality of Al_2O_3 thin film and observed low interface (thin film) resistance compared with air at bare Al substrates. High difference of rise in junction temperature (ΔT_J = 4.34°C) of LED was observed with 10 min synthesized samples annealed at 200°C. Annealed temperatures of Al_2O_3 thin film was not supported to reduce the total thermal resistance (R_{th-tot}) of the LED but synthesis time showed influence on changing the R_{th-tot} of the given LED at various driving currents. The surface analysis by atomic force microscopy was also supported the observed results in both thermal resistance and rise in junction temperature for various synthesis time and annealing temperature. Overall, CVD synthesized Al_2O_3 thin film could be a suitable alternative for thin film thermal interface material i LED applications.

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Keywords: CVD, Al_2O_3 thin film, thermal interface, LED

1. Introduction

Heat generated by an electronic packaging especially in light emitting diodes (LEDs) must be removed to the environment to maintain the temperature of the equipment within safe operating limits. This process often involves heat transfer from the LEDs to a heat sink that is one of the important elements in cooling the LEDs. Attaching the heat sink to the package surface requires the two to be brought into contact. When two such surfaces are joined, it may occur only at the high points of the both surfaces. The lower points will eventually create air-filled gaps. Such contact area can consist of more than 90% of air, in which represent a significant resistance to heat flow [1].

Thermal interface materials (TIMs) are used to eliminate those air gaps by conforming to the rough and uneven surfaces. As the TIM has a higher thermal conductivity than air [1], the resistance across the joint will decrease. A variety of material types have been developed in response to the changing needs for the LEDs system. Thermal paste is one among the thermal interface material (TIM) and having very low thermal conductivity. To increase the thermal conductivity, addition of high conductive material without affecting the physical nature to TIM is recommended. Among the fillers, Al_2O_3 is one of the most commonly used filler in polymer since it is widely available in market and has a good thermal conductivity. Al_2O_3 has been used as a filler to change thermal, dielectric and mechanical properties of the TIMs [1-3]. The same author group has already used metal oxide as filler in commercial thermal paste and achieved noticeable improvement in LED performance [4].

Among the available TIMs, Aluminium oxide (Al_2O_3) thin films have received an increasing amount of interest in many applications as dielectric, protective and ion barrier layers

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because of its excellent properties. In particular, it presents a high chemical stability, high radiation resistance [5] at the same time possess a good thermal conductivity [6]. This characteristic makes Al_2O_3 a good candidate for applications on metal-oxide semiconductor (MOS). Since, Al_2O_3 thin films have a thermal conductivity of 1 W/mK for thickness of 140nm, it will allow a better thermal path for high power LED to dissipate heat.

So far, most Al_2O_3 thin films have been deposited by physical vapor deposition (PVD) and it requires high production cost. But chemical vapor deposition (CVD) gives more conformal coverage of a surface. As one of the deposition techniques, CVD offers several distinct advantages which include depositing thin and high quality films with well-defined chemical composition and structural uniformity. No results have been reported so far on synthesis of Al_2O_3 thin film by Chemical Vapor Deposition method and used as thermal interface material. In this work, Al_2O_3 thin film was used as TIM, synthesized by CVD method and the effect of deposition time on the performance of LED is investigated. Al_2O_3 thin film was deposited on Al substrate and used as thermal substrate or heat sink for high power LED. The performance of the LED was tested at various driving currents and derived thermal resistances, rise in junction temperature for discussion.

2. Experimental Procedure

2.1 Synthesis of Al_2O_3 Thin Film

Al_2O_3 thin film was deposited on Al substrates by chemical vapor deposition method using 3 zones CVD furnace. In order to obtain a various elemental compositions and also optimization of the thin films, time taken for the deposition process was manipulated from 10 to 30 mins. The parameters of the process used in this study are provided in Table 1.

Table 1: Deposition parameters of Al_2O_3 thin film coating using CVD

Parameter	Condition
Precursor	Aluminium acetylacetonate
Precursor temperature	300°C
Substrate	Aluminium 2.5cmx25cm
Substrate temperature	450°C
N_2 flow rate	10 sccm
Deposition time	10 mins, 20 mins & 30 mins
Annealing condition	100°C, 200°C & 300°C

Aluminiumacetylacetonate [$Al(acac)_3$] was used as precursor for thin film deposition. Approximately 4g of $Al(acac)_3$ powder was filled in a boat type crucible and placed at the left zone of the furnace while the Al substrates were in the middle zone. At the outset, CVD tube was evacuated initially by rotary vacuum pump followed by filling inert gas (Argon) to maintain the inert atmosphere inside the tube. The precursor temperature was set to be at 300^0C. N_2 gas with a fixed flow rate of 10 sccm was used as carrier gas in which its flow rate was controlled by digital mass flow controller. N_2 gas was released into the tube from left zone, pushing in the $Al(acac)_3$ into the middle zone where the Al substrates were located with a fixed temperature of 450^0C. The coating process time was varied (10, 20 and 30mins) and coated Al_2O_3 thin film under atmospheric pressure. After the synthesis completed, the coated substrates underwent annealing process at 3 different temperatures (100^0C, 200^0C and 300^0C) for about 1 hour in separate tube furnace at ambient condition.

2.2 Thermal Transient Analysis

In order to test the performance of Al_2O_3 thin film as TIM, bare and Al_2O_3 thin film coated Al substrates were used as heat sink or thermal substrates and tested for the performance of LED package. Thermal Transient Tester (T3Ster) was used to perform thermal transient analysis in still air box at room temperature under three different driving currents (100, 350 and 700 mA). Initially, the LED was forward-biased for 900s and the transient cooling curve of the package was captured for another 900s. With the aid of Trister Master Software, all results were processed for structure function analysis. The LED was thermally calibrated using T3Ster as the power supply before we start the real measurement. The rise in the device junction temperature is produced from the product of K and the difference in temperature sensing voltage (ΔV_F).

$$\Delta TJ = \Delta V_F K$$

The LED was driven with a low operating current of 1mA at room temperature to prevent self-heating effect at the junction during the calibration process and the voltage drop was recorded once the LED reaches thermal equilibrium with the thermostat temperature. The ambient temperature of the LED was varied from 25^0C to 75^0C at 5^0C step size and the voltage drop was recorded at each temperature. From the graph of junction voltage (voltage drop) against ambient temperature, the K-factor of the LED was determined as 1.512 V/^{0}C.

2.3 Optical Measurements

The optical performances of the LED were also captured using optical spectrometer (MK350 LED meter) made by UPRtek in determining the optical parameters such as Correlated Color Temperature (CCT), Color Rendering Index (CRI) and luminosity (Lux) in still air box at room temperature. The experiment was carried out using Thermal Transient Tester (T3Ster) with three different driving currents (100, 350 and 700 mA). During the measurement, the LED was forward-biased for 900s at regulated currents.

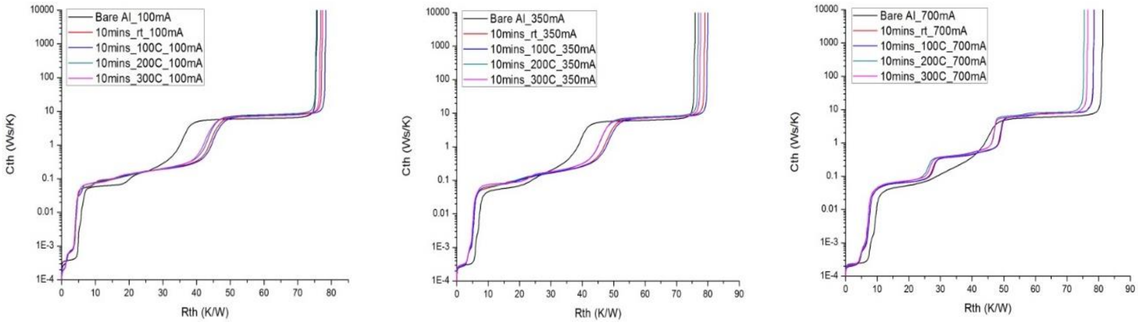
2.4 Surface analysis

Since the surface properties of two contact area is playing key role on conducting heat, the surface analysis is required to understand the behavior of contact surface and hence Atomic Force Microscopy (Model: ULTRA Objective, Surface Imaging Systems, GmbH) is used to study the surface behavior of the CVD synthesized Al_2O_3 thin film.

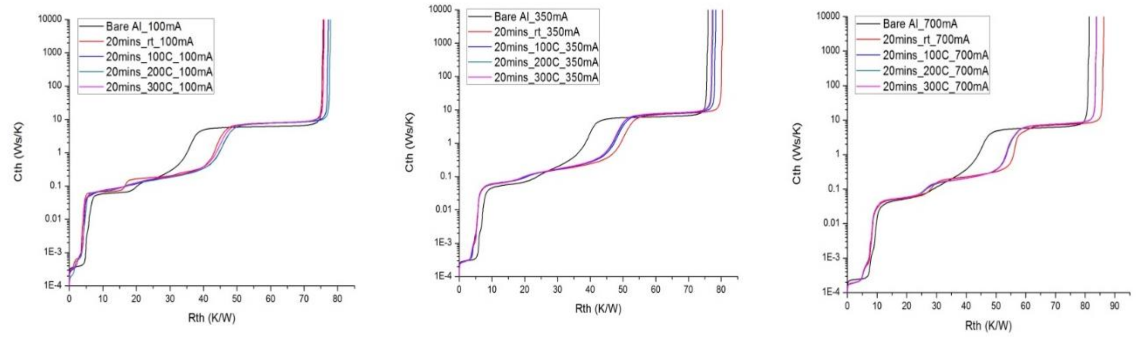
3. Results and Discussion

3.1 Thermal Transient Analysis

The thermal transient analysis was carried out for the given LED where Al_2O_3 thin film was used as thermal interface material. The thermal transient curve was recorded at 3 different driving currents for each sample and cumulative structure function curve was also derived using T3ster Master Software as shown in Figure 1 (a), (b) and (c). The variations of the cumulative structure function with respect to the annealing temperature were also included in the figures for discussions. The effect of annealing process on the samples will demonstrate its effect on heat conducting behavior of the Al_2O_3thin film.



(a) (b) (c)



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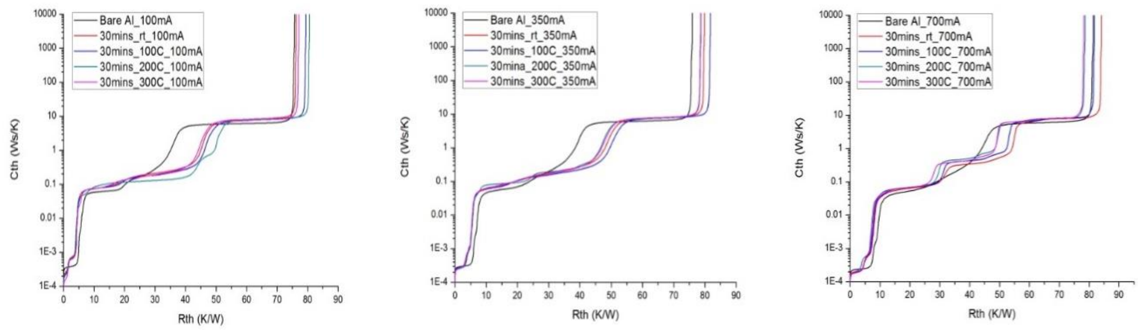


(a) (b) (c)

Fig. 1. Cumulative structure function of LED interfaced with Al_2O_3 thin film prepared at various process time and annealed temperature recorded at various driving currents.

In order to get the details on the performance of the LED, the rise in junction temperature (T_J) was derived from transient cooling curve and the observed values are presented in Table - 2. The interface thermal resistance ($R_{th\text{-}b\text{-}hs}$) was also evaluated from the cumulative structure function curves and summarized in Table -3.

Table 2 Rise in junction temperature of LED fixed on CVD Al_2O_3 thin film deposited Al substrates in various process times and annealing temperatures

Driving current (mA)	**Bare Al (K/W)**	**Room temperature (K/W)**	**Annealed at 100⁰C (K/W)**	**Annealed at 200⁰C (K/W)**	**Annealed at 300⁰C (K/W)**
		10 mins			
100	20.78	21.22	21.46	20.73	21.09
350	77.56	80.57	81.59	79.17	81.48
700	173.69	170.35	170.71	169.35	170.42
		20 mins			
100	20.78	21.34	21.17	21.31	20.88
350	77.56	81.70	79.88	78.89	78.84
700	173.69	183.26	179.10	178.68	179.87
		30 mins			
100	20.78	20.91	21.75	22.24	21.14
350	77.56	80.88	82.75	82.78	80.03
700	173.69	177.22	175.82	174.23	170.26

From the table, the variation in T_J is as expected ie when the driving current increases the rise in junction temperature increases. On considering 100 mA, there is not good difference in T_J with respect to change in process time and also annealing temperature. At 350 mA, an unexpected result is noticed as the bare Al substrates show low value in T_J than all other samples. On focusing the Al_2O_3 thin film at the same driving current (350 mA), thin film synthesized in 10 min duration and annealed at 200°C shows low value in T_J than all other boundary conditions. This is because of thermal mismatch of Al_2O_3 and Al at the moderate heating power. It is due the difference in thermal conductivity of Al_2O_3 and Al. At high driving current (700 mA), CVD synthesized Al_2O_3 thin film shows good performance on reducing the T_J value of the given LED considerably and observed the difference (ΔT_J) of 4.34°C when compared with bare Al boundary condition and recorded as high ΔT_J than all other boundary conditions considered in this analysis. The observed difference in ΔT_J is high when compared with sputtered Al_2O_3 thin film as TIM in another study by same author group [6]. It indicates the effect of heat during the burning time of LED on the thermal conductivity of Al_2O_3 thin film.

As noticed from the change in T_J, it is attributed to the effect of thermal resistance of the interface (CVD Al_2O_3) used in this study. As observed from T_J, $R_{th\text{-}tot}$ values are contradicting with the expected results and showing high value than the expected values. It may be due to the current crowding effect and results in the reduction of power dissipation area, which causes the thermal resistance of a semiconductor to increases [7,8].

It is also observed that the bare Al substrates show good results and low than all boundary conditions. Moreover, the annealing process of CVD Al_2O_3 thin film shows good results on reducing the $R_{th\text{-}tot}$ of the LED as compared to the non-annealed samples. It is attributed to the effect of structural changes and hence huge difference in $R_{th\text{-}tot}$ is observed with annealed Al_2O_3 thin film boundary conditions. High driving currents give more heat to the material and hence high value in $R_{th\text{-}tot}$ is noticed with high driving currents. There are few reasons for this observation for annealed Al_2O_3; i) There may be thermal mismatch between the thermal conductivity of Al_2O_3 and Al, ii) lattice mismatch between Al_2O_3 thin film and Al substrate [9-11]. The results are not given for discussion here.

Table 3. Interface thermal resistance of CVD Al_2O_3 thin film deposited Al substrates in various process times and annealing temperatures

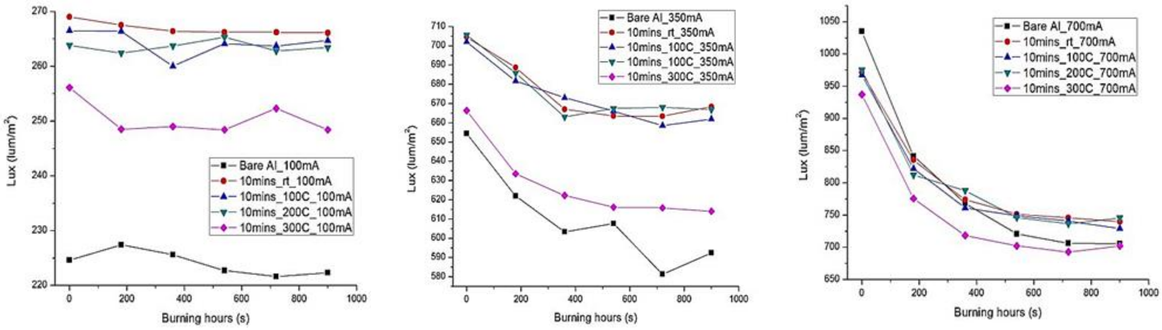
Driving current (mA)	**Bare Al (K/W)**	**Room temperature(K/W)**	**Annealed at 100°C (K/W)**	**Annealed at 200°C (K/W)**	**Annealed at 300°C (K/W)**
		10 mins			
100	37.00	30.15	30.28	28.43	30.96
350	33.65	28.66	28.92	27.55	29.50
700	32.80	28.48	28.72	27.72	29.14
		20 mins			
100	37.00	28.99	28.53	29.95	28.11
350	33.65	26.54	26.36	26.18	25.98
700	32.80	28.69	25.85	25.68	25.65
		30 mins			
100	37.00	26.91	29.68	27.72	28.27
350	33.65	27.26	28.12	28.33	27.79
700	32.80	28.19	27.19	28.07	27.75

Consequently, the interface (Al_2O_3 thin film) thermal resistance is evaluated from the cumulative structural analysis (Figure 1) and the values are summarized in Table 3. It shows that the Al_2O_3 thin film interface has a low value in the thermal resistance than that of bare Al (air interface). The differences in R_{th} ($\Delta R_{th\text{-}b\text{-}hs}$) of Al_2O_3 thin film interface keeps on decreasing from 100mA to 700mA as compared with the bare Al, but however the differences are still high (>3.6 K/W). This indicates that the thin film conducts more heat from the LED to the heat sink instead of air which act as thermal barrier. Overall, it specifies that the method of CVD deposition process of Al_2O_3 thin film coated on Al substrate is performing well on reducing the T_J of the LED at high driving currents and can be considered CVD as alternative synthesis method for Al_2O_3 thin film as TIM for high power LED application.

3.2 Optical Analysis

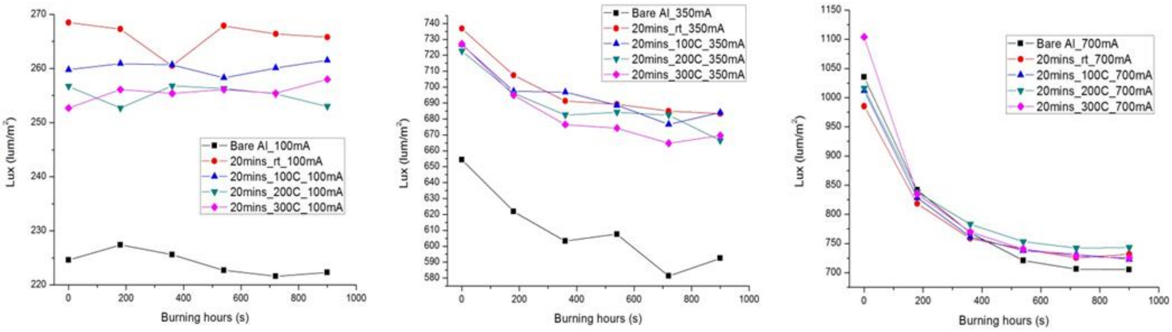
Color Rendering Index (CRI) is the most useful measure of a light source's color characteristics as it measures of a light source's ability to show how well its renders color. CRI is calculated from the differences in the chromaticity of 8 standard test color samples (TCS) when illuminated by a light source. In this study, the recorded CRI for the given LED for all process conditions were observed and it is noticed that the differences between the values are within ±5. According to a research, the changes in the CRI values of less than 5 points are not significant [7]. Hence the data are not provided and shown for discussion.

Lux value of the given LED was tested at various driving currents to see the performance of Al_2O_3 as thin film interface material prepared by CVD method. Lux of the given LED describes the effective light output for the given driving currents and is measured in lumen per square meter (lum/m^2). The measured values are plotted to see the influence of synthesis time and annealing temperature on the light output of the LED. The observed results were plotted against the burning time as shown in Figure 2- 4.



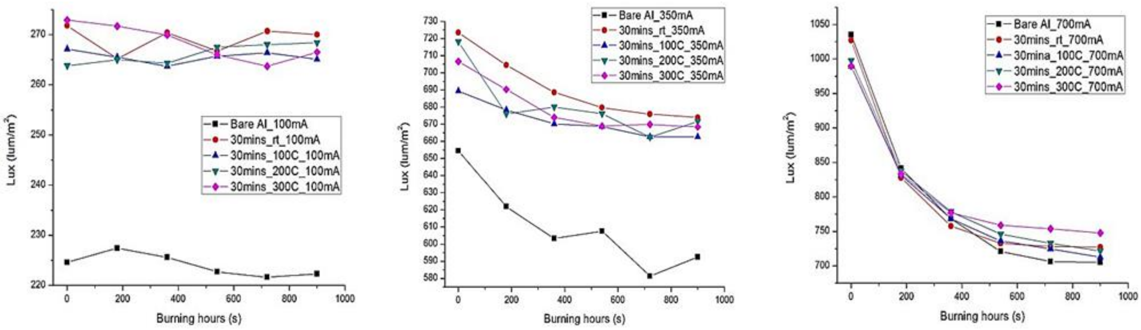
(a) (b) (c)

Fig. 2 Variation of Lux at different boundary conditions for Al_2O_3 thin film prepared at 10 mins process times measured at various driving currents



(a) (b) (c)

Fig. 3 Variation of Lux at different boundary conditions for Al_2O_3 thin film prepared at 20 mins process times measured at various driving currents



(a) (b) (c)

Fig. 4 Variation of Lux at different boundary conditions for Al_2O_3 thin film prepared at 30 mins process times measured at various driving currents

At low processing time (10mins), the CVD processed Al_2O_3 thin film perform very well on improving the light output in terms of lux as the LED running time increases. Moreover, as-deposited Al_2O_3 thin film performs much better than other boundary conditions. It clearly seen from the fig. 2 that the annealed Al_2O_3 thin film is not supporting the improved light output as the driving current increases from 100 to 700 mA. At high driving currents, a noticeable difference in lux value could be observed for all boundary conditions compared with bare Al substrate boundary conditions. The increased processing time (20 and 30 mins) for Al_2O_3 thin film is not showing much influence on changing the light output of the given LED. The observed lux values are in the range between 220 – 275 lum/m^2, 580 – 740 lum/m^2 and 700 – 1100 lum/m^2 for 10 mins, 20 mins and 30 mins processed samples respectively. At 300 mA driving current, the film processed at 20 min time duration is showing good performance on improving the light output as compared with 10 and 30 mins process timings. In addition to this, the annealing temperature is also influencing the light output of LED with respect to the processing time. Overall, the CVD synthesized Al_2O_3 thin film performs well on enhancing the optical output and hence it is proved as an alternative thermal interface material for LED application. This observation is correlated to the rise in the junction temperature of the LED as the decrease in the junction temperature will results in the increase of the lux value.

D. AFM Analysis

In order to understand the influence of different process time as well as the annealing temperature on surface morphology of Al_2O_3 thin film on Al substrates, the 3D surface images were captured by AFM as shown in Figure 7 - 9.

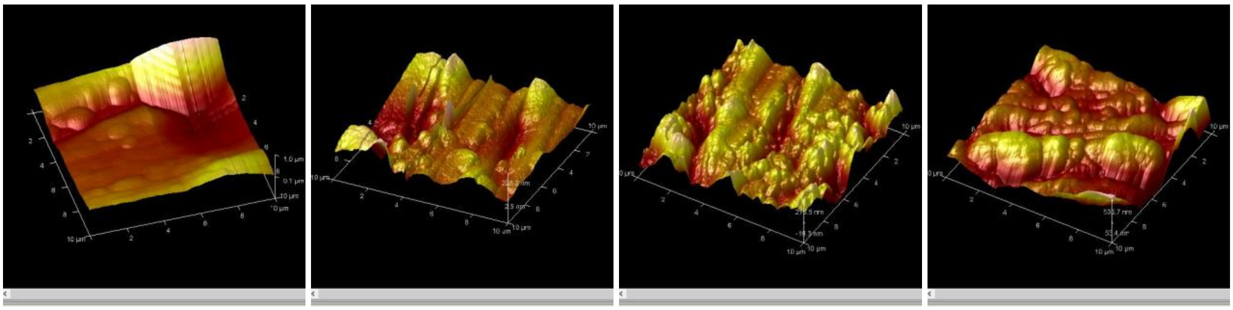


Fig. 7: AFM images of Al_2O_3 thin film synthesized in 10mins at (a) room temperature, and (b) annealed at 100 ℃, (c) 200 ℃ and (d) 300 ℃

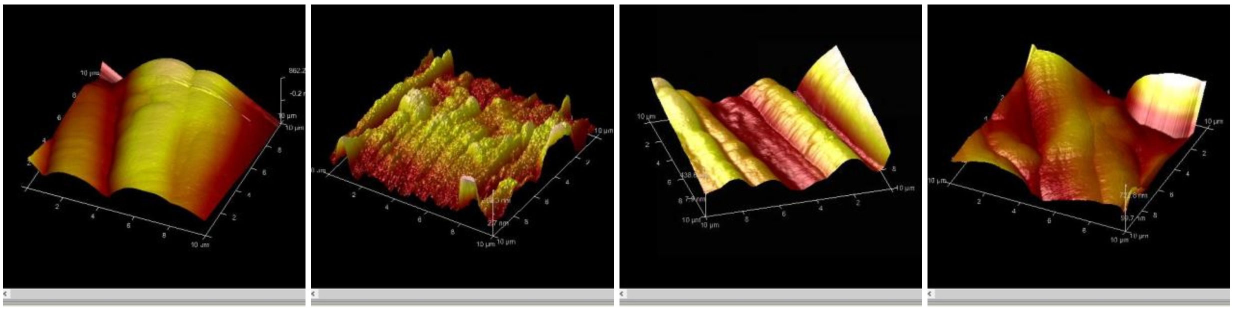


Fig. 8: AFM image of 20mins deposition samples: (a) room temperature, (b) annealed at 100 ℃, (c) annealed at 200 ℃ and (d) annealed at 300 ℃

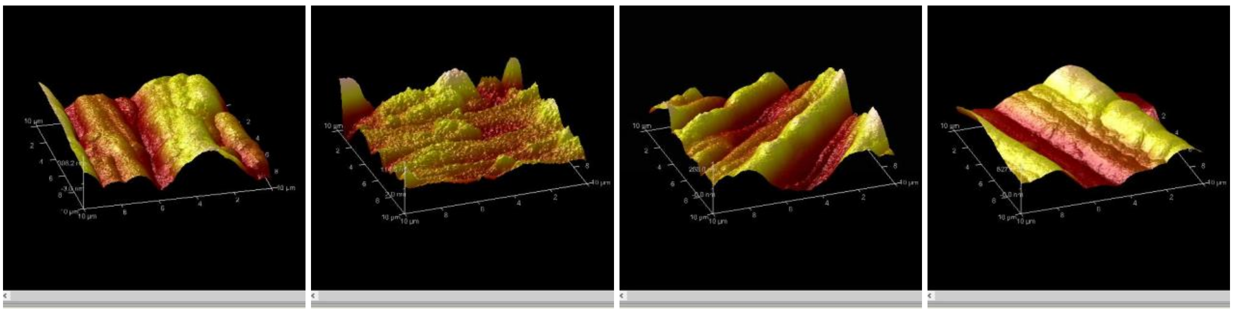


Fig. 9: AFM image of 30mins deposition samples: (a) room temperature, (b) annealed at 100 ℃,(c) annealed at 200 ℃ and (d) annealed at 300 ℃

As we can see from the picture, it clearly shows that the difference in surface morphology is due to the annealing temperature as well as the process time. A plate type structure on the surface can be observed mostly on samples (a) and (d) for all process time. Hence it is concluded that the process time and annealing temperature is influenced the surface of CVD synthesized Al_2O_3 thin film noticeably. To study in detail, all captured images were processed and analyzed using Nanoscope software and the surface parameters such as surface roughness and particle size were measured as shown in Table 4.

Table 4: Surface roughness and particle size of Al_2O_3 thin film prepared by CVD method

Time of deposition (min)	Roughness (μm)				Particle size (μm)			
	RT	100^0C	200^0C	300^0C	RT	100^0C	200^0C	300^0C
10	0.198	0.0414	0.461	0.0873	2.789	0.310	0.745	1.388
20	0.232	0.0185	0.125	0.114	9.595	0.501	0.767	0.229
30	0.0754	0.0182	0.0738	0.192	0.618	1.001	0.751	8.021

The samples of 30mins process time show the lowest surface roughness of all which may indicate that higher deposition time will result in lower roughness. For all process time, the samples annealed at 100°C show lower roughness than other processing temperatures and very low surface roughness value (18 nm) was noticed with 30 mins synthesized samples annealed at 100°C. Surface with low roughness conducts more heat than the surface with high roughness. It supports the observation made with thermal resistance analysis. But in the same case, it is also noticed that sample annealed at 300^0C has the highest value of roughness as compared to other two samples of the same annealed temperature. High roughness (461 nm) is also noticed with samples processed in 10 mins and annealed at 200°C. Overall, the starting surface roughness of annealed samples was low specifying the influence of Al and O diffusion onto the thin film surface. As the temperature increases, the roughness became high which is explaining the effect of diffusion at high temperature.

Considering the particle size, it reveals that the increase in the annealing temperature may contribute to the growth of a larger particle size except for the 20mins process time sample. The film prepared having a large particle size also present a high surface roughness. Moreover, the particle size is also a key factor for thermal resistance. Large grain/particle size have more contact surface area and hence conducts more heat than small particle size. Consequently, the as-deposited samples show larger particle size than annealed samples and provide poor contact with the surface as it is possible with minimum no. of contact points. As a result, high thermal resistance was noticed with as-deposited sample than annealed samples.

4. Conclusion

Al_2O_3 thin film was synthesized on Al substrate by CVD method at various process time and annealed at various temperatures. The synthesized samples were used as thermal interface material and tested their performance with LED at various driving currents. Al_2O_3 thin film interface LED showed lower rise in junction temperature and noticed the good difference with 10 min processed samples annealed at 200 °C when compared with bare Al substrates. As compared with air, Al_2O_3 interface showed lower interface resistance for all boundary conditions. CVD process time and annealing temperature were influenced on changing both thermal resistance and junction temperature of the LED at various driving currents. Surface analysis was also supported the observation by transient curves. Based on the results, it is concluded that the CVD synthesized Al_2O_3 thin film samples could be an alternative as thermal interface material for high power LEDs.

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