

## Performance of Chemical Vapor Deposited ZnO thin film as thermal interface material on optical properties of LED

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**Abstract**—Chemical Vapor Deposition (CVD) was used for the synthesis of ZnO thin film on Al substrates at various flow rates of O<sub>2</sub> gas. ZnO thin film coated substrates were tested as thermal substrates on influencing the optical properties of high power LED at various operating currents. Spectrometer analysis showed that ZnO thin film prepared at 10 sccm O<sub>2</sub> flow rate showed better performance by reducing the Color Correlated Temperature (CCT) at driving currents. CCT values were maintained with respect to driving currents by ZnO thin film interface at all driving currents than air interface (bare Al substrate). On luminous flux analysis, 5 sccm samples showed good performance on increasing the light of the give LED at all driving currents than bare Al boundary condition. The observed results were evidenced with help of particle size distribution analysis on all film surface using Nanoscope software. Overall, ZnO thin film deposited at low O<sub>2</sub> flow rate would be an alternative to solid thin film interface material in electronic packaging applications.

**Keywords**—Thin film, ZnO, Interface material, LED, Optical properties

### I. INTRODUCTION

Over the years, lighting industry has gained rapid development in improving the performance of LEDs. This is due to the innovation of LED devices that has been proven to be better performance than the other general lightings such as incandescent lamps and compact fluorescent lamps (CFL). LEDs are proven to be more stable, longer life span, cost efficient and environmental friendly [1]. Despite the rapid development of the LED, there are some major drawbacks that can cause failure in the device such as over-heating of LED when driven at higher input current [2]. Moreover, the miniaturization of the LEDs significantly disrupts the effective heat dissipation in which large amount of heat accumulates in the device and eventually affects the thermal performance of the device [3]. Hence, thermal management solutions are vital to ensure the sustainability of the LEDs technology.

Recently, LEDs are being utilized in many industries such as automotive, advertising, medical and many more. Understanding the luminance and the light rendering performances of LEDs are important aspects in maintaining the performance and quality of the LED [4]. To sustain the light output performance, it is important to maintain the thermal management. One of the solutions that have recently gained interest among researchers is by employing thermal interface material (TIM) as the medium to help in dissipation of heat from the device. Commercially available TIM such as thermal grease, thermal pad, thermal paste and polymer based adhesives such as epoxy and silicone had causes problem such as pump out and dry out of the material [5]. Thermal greases or thermal pastes are usually not preferred recently as they are often inconvenient and messy when being used [6]. It has also

been reported that thermal pads are performing inefficiently due to the thickness of the pad that can increase the stack up tolerance [7]. Meanwhile, it has been reported elsewhere that adhesives at high temperature may cause delamination issues [8].

Other alternative that can be applied to replace the commercially available TIM is by using solid thin films as TIM. Many researches have been carried out in investigating the uses of thin film as TIM. Park et. al. reported the usage of TiW-Au-TiW thin film layer as TIM in the design of thermometer [9]. Shanmugan et. al. investigated the use of Zn thin film as TIM in electronic packaging application [10]. Meanwhile, Ong et. al., stated that boron doped aluminium nitride thin films are effective TIMs for high power LEDs [11]. Based on the earlier findings and to the best of the author's knowledge, zinc oxide has been proven as a best material for thin film application since it has electrical and optical transparency [12]. It is also an n-type semiconductor with direct band gap energy of 3.37 eV at room temperature and 60 meV of free exciton binding energy which helps to efficiently excite the emission process at room temperature that enable the devices to function at lower threshold voltage [13]. Recent report by Shanmugan et. al. showed that the RF sputtered ZnO thin film on Cu substrate was used as TIM and improved the luminance (LUX) and reduced the CCT of high power LED noticeably [14]. Mutharasu et. al. has already reported that ZnO thin film as TIM was enhanced the LUX of LED and suggested as an alternative to commercial thermal paste [15]. There are various deposition techniques to deposited zinc oxide thin film such as Chemical vapor deposition (CVD) [16], sputtering [17], spin coating [18], electron beam deposition [19] and spray pyrolysis [20]. Among these techniques, CVD had shown to produce high quality and uniform thin film [16,21].

In this research, ZnO is prepared on Al substrate by CVD method at various flow rate of O<sub>2</sub> gas. The ZnO thin film deposited Al substrate was used as a heat sink. The optical performance of the LED is focused and tested at various driving currents. The changes in optical properties such as color rendering index (CRI), Correlated Color Temperature (CCT) and the LUX are observed and discussed.

### II. MATERIALS AND METHODOLOGY

Al substrates (2.5 x 2.5 x 1.5 cm) were initially cleaned in ethanol bath using ultrasonic process for about 10 minutes at 50°C using Ultrasonicator. To obtain ZnO thin films, CVD furnace equipped with three zone was used in which Al substrates were placed at the center zone as shown in fig. 1. Zinc acetate dehydrate powder (5 g) was used as precursor and maintained the temperature at 300°C to initiate the process at

the left zone (see fig.1). During synthesis, the substrate temperature was kept at 400°C. Oxygen was introduced in the chamber and its flow was controlled by mass flow controller. Before deposition, the CVD tube was kept in vacuum condition during the heating process. To optimize and study the influence of gas flow rate on ZnO thin film quality and growth, O<sub>2</sub> gas flow rate was varied from 5 sccm to 40 sccm and deposition time was fixed at 30 minutes. The parameters used in the experiment are shown in the Table-1. The surface quality of the prepared film was verified and analyzed by Nanoscope software using AFM images. The optical characterizations of high power LED (OSRAM Golden Dragon, 3.7W, White) attached on ZnO thin film coated Al substrate was measured by LED spectrometer (MK350, UPRtek). Still air chamber (300 x 300 x 300mm) was used for all experiments to satisfy the JEDEC JESD 51-2A standards on natural air convection measuring method [22] at room temperature of 25°C ± 1°C. it is recommended method for testing LED for their thermal performance in controlled environment. The ZnO thin film interfaced LEDs were forward biased for 900 seconds and captured the light emitting spectrum using spectrophotometer at three different driving currents (100 mA, 350 mA, 700 mA).

TABLE I. SYNTHESIS MATERIAL AND PARAMETERS FOR CVD PROCESSED ZnO THIN FILM

Parameter	Condition
Precursor	Zinc Acetate Dihydrate
Precursor temperature	300°C
Substrate	Aluminium (2.5 cm x 2.5 cm)
Substrate temperature	400°C
O <sub>2</sub> flow rate	5sccm, 10sccm, 20sccm, 30sccm, 40sccm
Deposition time	30 ins

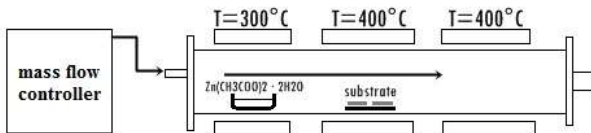


Fig 1. Schematic Diagram of CVD system

III. RESULTS AND DISCUSSION

A. Optical properties

1) Color Correlated Temperature analysis

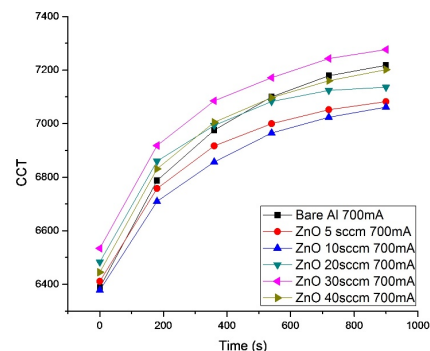
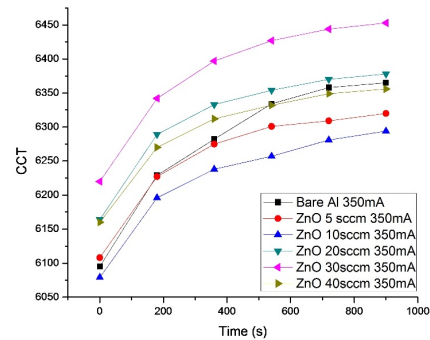
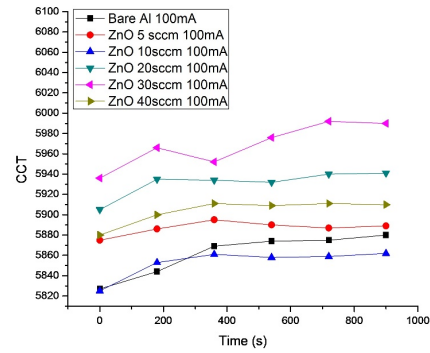


Figure 2. Variation of LED CCT values at various driving currents for CVD processed ZnO thin film interface under various flow rate of O<sub>2</sub>

The optical properties of the LED such as CCT and LUX were studied using spectrometer at various driving currents to evaluate the performance of CVD processed ZnO thin film as thermal interface material application. The recorded CCT values are plotted against various driving currents as shown in fig. 2. It shows that the O<sub>2</sub> flow rates affect the LED performance by changing CCT considerably. The observed CCT values are between 5820 and 7250 °K. As the driving currents increases, the CCT values are also increases with respect to O<sub>2</sub> flow rate. Moreover, the CCT values are increasing with measuring time increasing. It is noticed that the sample prepared at 30 sccm O<sub>2</sub> flow shows high value in CCT than all other boundary conditions. Lowest value in CCT could be observed for the sample prepared at 10 sccm flow rate for

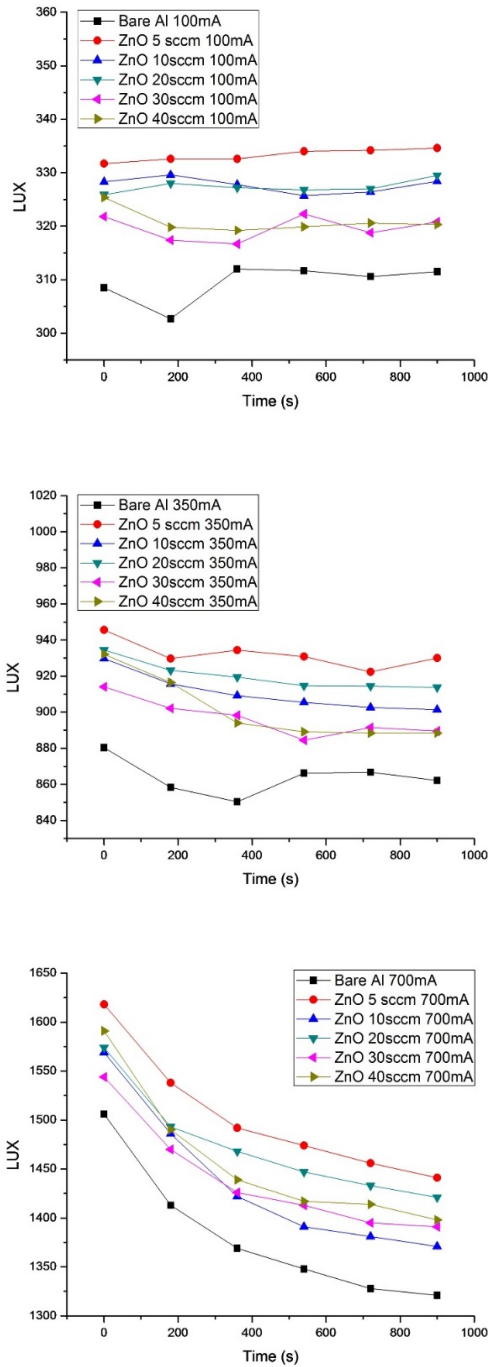


Figure 3. Variation of LED Lux values at various driving currents for CVD processed ZnO thin film interface under various flow rate of O<sub>2</sub>.

all driving currents. Next to 10 sccm flow rate sample, low CCT values are noticed with 5 sccm flow rate at > 350 mA. The LED tested at high driving current shows that the bare Al

boundary condition affect the LED performance by increasing the CCT with measuring time increasing when compared to ZnO thin film coated samples.

Manufacturers produce white LEDs with outputs classed as “warm white” (2,600-to-3,700 K correlated color temperature or CCT), “neutral white” (3,700-to-5,000 K CCT) and ‘cool white’ (5,000-to-8,300 K CCT) grouped into “bins” comprising devices of very similar color output [23]. From our results, the color of the LED is estimated to be cool white and slightly shift towards the blue color. This shift is not dominating in ZnO thin film interfaced LED. Overall, the proposed ZnO thin film helps to control the change in lattice constant of the LED die by reducing the die temperature or junction temperature which is evidenced by observing a small shift in CCT value for the tested LED [23].

2) Luminous Flux analysis

To strengthen the above observation, luminous flux of high power LED is also recorded for all boundary conditions at various driving currents. The observed data are plotted against the time duration as shown in fig.3 and exhibits clearly about the influence of O<sub>2</sub> flow rate on the light output of the LED with respect to the driving currents. It clearly reveals that the ZnO thin film coated Al substrates perform well on increasing the light output and hence high lux value could be observed for ZnO thin film boundary conditions than bare Al boundary conditions. On considering the driving currents, the output is much affected with measuring time for low driving currents (100 mA). Noticeable fluctuation in light output is observed as the driving current increases (>350 mA) and especially, a decrease in lux is recorded with measuring time increase at 700 mA. Among all other O<sub>2</sub> flow rates, ZnO thin film deposited at 5 sccm O<sub>2</sub> flow rate boundary condition shows higher light output and degrade the same in small amount than bare Al boundary conditions. The variation in the observed CRI values is ±4. Since there is no significant change in CRI values and not addressed in detail here.

3) Surface Analysis

The above observation has been verified with the help of particle size analysis derived from surface analysis done by Nanoscope analysis software using AFM images of ZnO thin film prepared by CVD method at various flow rate of O<sub>2</sub> gas. The surface depth profile was captured in histogram format and presented in Fig. 4. It depicts that the histogram of 5 sccm samples shows some flatness and the particle size range is between 0.25 and 1.25 μm. It is feasible for making good thermal contact with the surface and reduces the contact resistance. Thus, the surface of 5 sccm samples would be the better surface quality and hence improve the device performance by removing the heat from device to ambient via Al substrates efficiently. This observation is supported the above discussion of improved lux with ZnO thin film prepared at 5 sccm flow of O<sub>2</sub>. From the fig., ZnO thin film prepared at 30 sccm O<sub>2</sub> flow shows the histogram of particle size

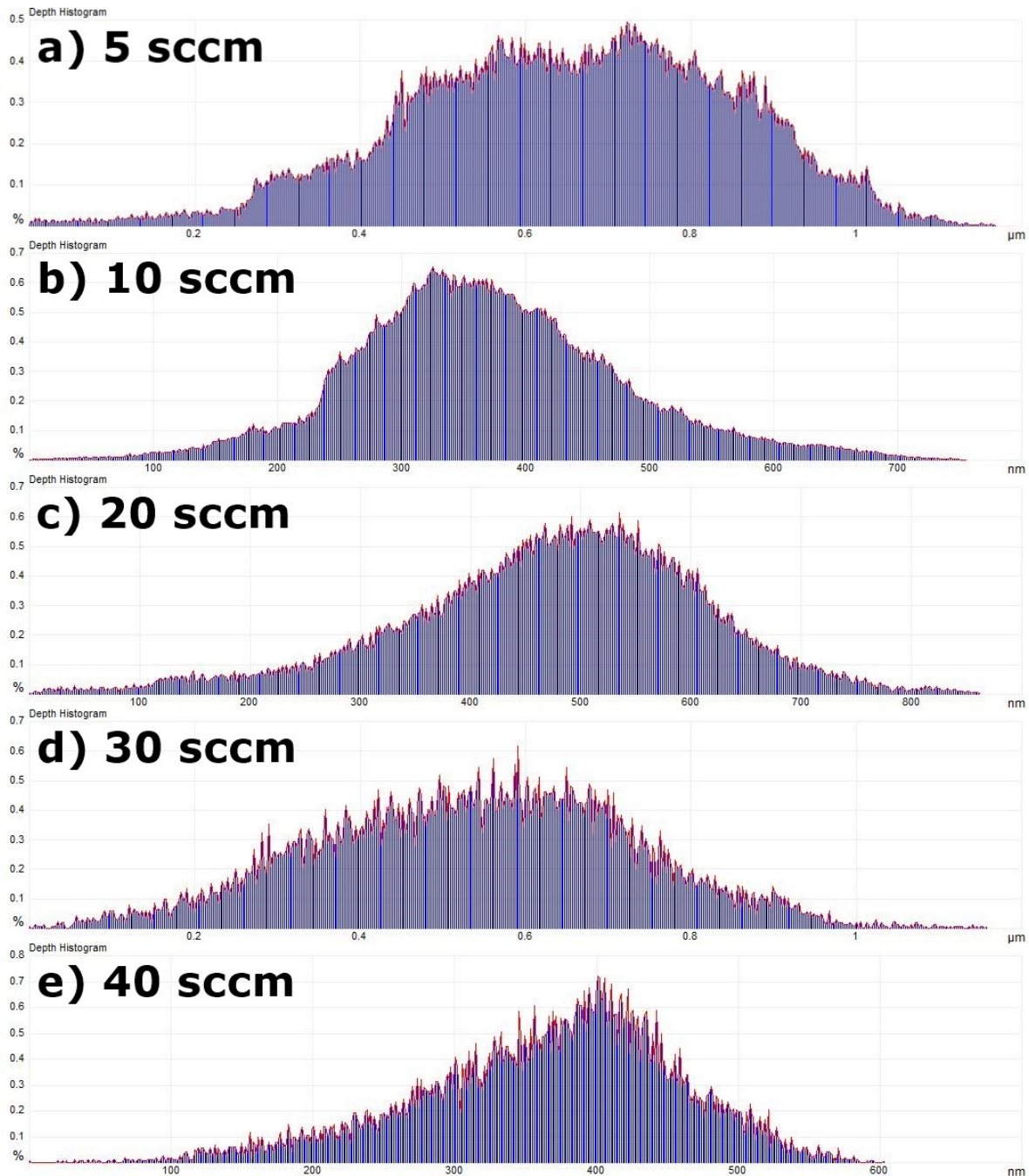


Fig. 4. Particle size distribution of ZnO thin film on Al substrates at various flow rate of  $O_2$  gas

distribution like 5 sccm sample but the high of the histogram is not good as we noticed with 5 sccm. There is an irregular pattern which indicates the possibility of air interface at the joint between LED and ZnO thin film coated Al substrates. It reflects the poor performance in thermal conduction and hence low light output than 5 sccm samples. Overall, it concludes that the uniform particle size distribution is expected with 5 sccm samples and favor the heat conduction with less thermal resistance.

#### IV. CONCLUSION

The optical properties of LED were tested using ZnO thin film as thermal interface material synthesized by CVD method at various flow rate of  $O_2$  gas. ZnO thin film prepared at low  $O_2$  flow rate showed better performance on maintaining the CCT values of the LED and reduce the risk on color shifting. High level lux was noticed for the LED interfaced using ZnO thin film prepared at 5 sccm flow of  $O_2$  gas. The results were

evidenced by observing optimum level particle size on the surface which make good thermal contact conductance with the LED package. It is concluded that the ZnO thin film prepared at low  $O_2$  flow rate by CVD process would be recommended for solid thin film thermal interface material application in solid state lighting applications. A detailed analysis on thin film material quality and their reliability is preferred for further development of ZnO thin film in thermal management

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## REFERENCES

- [1] Chang, M.H., Das, D., Varde, P.V. and Pecht, M., Light emitting diodes reliability review. *Microelectron. Reliab.*, vol.52, pp. 762-782 2012.
- [2] Routledge G. Lighting the way to a low-energy future. *IEE Rev.* vol 48, pp. 21-25, September 2002.
- [3] Nakayama W. Thermal management of electronic equipment- A review of technology and research topics. *Appl. Mech. Rev.*, vol. 39, pp. 1847-1868, December 1986.
- [4] Fontoynt M. Perceived performance of daylighting systems: lighting efficacy and agreeableness. *Sol. Energy.* vol. 31, pp. 83-94. August 2002.
- [5] Due J, Robinson AJ. Reliability of thermal interface materials A review. *Appl. Therm. Eng.* Vol. 50, pp. 455-463, 10 Jan 2013.
- [6] Gwinn, J.P. and Webb, R.L., Performance and testing of thermal interface materials. *Microelectron. J.*, vol. 34, pp.215-222, 2003.
- [7] Weixel, M., Hewlett-Packard Company, Composite thermal interface pad. U.S. Patent 6,037,659, 2000.
- [8] Li, Y. and Wong, C.P., Recent advances of conductive adhesives as a lead-free alternative in electronic packaging: materials, processing, reliability and applications. *Mater. Sci. Eng: R: Reports*, vol. 51, pp.1-35, 2006.
- [9] Park, Jong-Jin, and Minoru Taya. "Design of thermal interface material with high thermal conductivity and measurement apparatus." *J. Electron. Packag.* 128, 46-52, 2006.
- [10] Shanmugan S, Mutharasu D. "Thermal and optical performance of LED using Zn thin film as thermal interface material in electronic packaging application" *International Journal of Engineering Trends and Technology (IJETT)*, Vol. 39 no. 1, 2016.
- [11] Ong, Z.Y., Shanmugan, S. and Mutharasu, D., "Thermal performance of high power LED on boron doped aluminium nitride thin film coated copper substrates." *J. Sci. Res. Rep.*, vol. 5, pp.109-119, 2015
- [12] Shinde SS, Shinde PS, Bhosale CH, Rajpure KY. Optoelectronic properties of sprayed transparent and conducting indium doped zinc oxide thin films. *J. Phys. D: Appl. Phys.* Vol. 41, pp. 105109, May 2008.
- [13] Jagadish C, Pearton SJ, editors. Zinc oxide bulk, thin films and nanostructures: processing, properties, and applications. Elsevier; 10 October 2011.
- [14] Shanmugan S, Yin OZ, Anithambigai P, Mutharasu D. Analysis of ZnO Thin Film as Thermal Interface Material for High Power Light Emitting Diode Application. *J. Electron. Packag.* Vol. 138, pp. 011001, 1 Mar 2016.
- [15] Mutharasu, D., Shanmugan, S., Anithambigai, P. and Yin, O.Z., Performance testing of 3-W LED mounted on ZnO thin film coated Al as heat sink using dual interface method. *IEEE Transactions on Electron Devices*, vol. 60 pp.2290-2295, 2013.
- [16] Minegishi K, Koizumi Y, Kikuchi Y, Yano K, Kasuga M, Shimizu A. Growth of p-type zinc oxide films by chemical vapor deposition. *Japanese Journal of Applied Physics.* vol. 36, pp. L1453, Nov 1997.
- [17] Sundaram KB, Khan A. Characterization and optimization of zinc oxide films by rf magnetron sputtering. *Thin Solid Films.* vol. 295, pp.87-91, 28 Feb 1997.
- [18] Kamaruddin SA, Chan KY, Yow HK, Zainizan Sahdan M, Saim H, Knipp D. Zinc oxide films prepared by sol-gel spin coating technique. *Appl. Phys. A: Mater. Sci. Process.* Vol. 104, pp. 263-268, 1 July 2011.
- [19] Aghamalyan NR, Gambaryan IA, Goulanian EK, Hovsepian RK, Kostanyan RB, Petrosyan SI, Vardanyan ES, Zerrouk AF. Influence of thermal annealing on optical and electrical properties of ZnO films prepared by electron beam evaporation. *Semicond. Sci. Technol.* Vol. 18 pp. 525, 28 Apr 2003.
- [20] Joseph B, Gopchandran KG, Manoj PK, Koshy P, Vaidyan VK. Optical and electrical properties of zinc oxide films prepared by spray pyrolysis. *Bull. Mater. Sci.* Vol. 22, pp. 921-926, August 1999.
- [21] Liu X, Wu X, Cao H, Chang RP. Growth mechanism and properties of ZnO nanorods synthesized by plasma-enhanced chemical vapor deposition. *J. Appl. Phys.* Vol. 95, pp. 3141-3147, 15 Mar 2004.
- [22] JEDEC Standard. Integrated Circuits Thermal Test Method Environment Conditions-Natural Convection (Still Air). JESD51-2A. 2007.
- [23] <https://www.digikey.com/en/articles/techzone/2013/may/thermal-effects-on-white-led-chromaticity>