STUDY OF METAL IMPREGNATED ZnO THIN FILM AS THERMAL INTERFACE FOR HIGH POWER LED APPLICATIONS

NUR HASYIMAH BINTI HASHIM

UNIVERSITI SAINS MALAYSIA

2019

STUDY OF METAL IMPREGNATED ZnO THIN FILM AS THERMAL INTERFACE FOR HIGH POWER LED APPLICATIONS

by

NUR HASYIMAH BINTI HASHIM

Thesis submitted in fulfilment of the requirements
for the degree of
Master of Science

August 2019

ACKNOWLEDGEMENT

Alhamdulillah, my utmost gratitude is to Allah SWT whose grace and blessings are upon me through the years of my post-graduate research. Whom also provide me strength, wisdom and guidance to complete this thesis.

I would like to express my sincere gratitude and deep appreciation to my main supervisor, Prof. Dr. Mutharasu Devarajan for his valuable guidance and big effort in supporting and motivating me throughout the whole period of the research. I also give my sincere thanks to Dr. Shanmugan Subramani for his many guidance and valuable inputs in this research project. I give my special thanks to all lab technicians from Nano Optoelectronics Research Lab (NOR) for the technical support on the research equipment. Unforgotten to give my gracious thanks to my lab colleagues, Anitha, Dheepan, Jass, Wei Qiang, Mah, Muna and Idris who's together facing both hardness and joy in research. Thanks for the kindness and hospitality in practical works and sharing ideas and experience together throughout the years of the research. Special thanks to my parents and sisters, Hashim Nor, Siti Halijah and Siti Hamidah for their inseparable support through the peak and valleys of my postgraduate life.

Additionally, I owe my gratitude to Akram Zaki who continuously give various support to ensure I completed Master's degree. Not forgetting, thanks to my Bakti Angels for their friendship, moral support and stood by me during this postgraduate study. Finally, I would like to thank my sweetheart who always there for me and cheer me up. Thank you.

TABLE OF CONTENTS

CHAPTER 5 - CONCLUSION

LIST OF TABLES

LIST OF FIGURES

LIST OF ABBREVIATIONS

LED	Light emitting diode
PCB	Printed circuit board
MCPCB	Metal core printed circuit board
TIM	Thermal interface material
BLT	Bond line thickness
XRD	X-ray diffraction
AFM	Atomic force microscopy
FESEM	Field emission scanning electron microscope
FWHM	Full width half maximum

LIST OF SYMBOLS

R_{th}	Thermal resistance
T_J	Junction temperature
ΔT_J	Rise in junction temperature
Q	Heat source
T	Temperature
A	Cross-sectional area
k	Thermal conductivity
R_c	Contact resistance
P_I	Input electrical power
P_{opt}	Output optical power
P_H	Heat dissipation power
$P_{thermal}$	Thermal power
V_f	Forward voltage
D	Crystallite size
λ	Wavelength of radiation
β	Full width at half maximum (FWHM)
θ	Bragg diffraction angle of (hkl)
ε	strain

KAJIAN TENTANG FILEM NIPIS ZnO BERISI LOGAM SEBAGAI LAPISAN HABA ANTARA MUKA UNTUK APLIKASI LED KUASA TINGGI

ABSTRAK

Isu terma dalam LED kuasa tinggi sangat memberi kesan kepada prestasi pencahayaan LED dari segi keutuhan dan ketahanannya. Bahan antara muka termal telah disyorkan untuk meningkatkan pelesapan haba kerana ia berkebolehan mengendalikan haba secara berkesan antara sumber haba dan penyerap haba. Dalam projek ini, filem nipis ZnO dan dua jenis ZnO berisi logam (Kuprum dan Perak) telah diendap pada substrat aluminium melalui teknik salutan putaran sol-gel. Parameter pemendapan ZnO seperti bilangan kitaran salutan dan kepekatan sol-gel telah diubah untuk memperolehi filem tipis ZnO yang berkualiti tinggi. Selepas pengoptimuman filem nipis ZnO, filem nipis logam diisitepuan ZnO dihasilkan pada kepekatan 0.03M, 0.06M, 0.09M. Ciri-ciri struktur filem nipis telah dikaji menggunakan XRD manakala morfologi dan topologi permukaan filem dianalisis melalui FESEM dan AFM. Prestasi terma filem nipis diuji menggunakan T3Ster dimana LED kuasa tinggi dilekapkan pada substrat bersalut. Keputusan belauan sinar-X menunjukkan tiga puncak utama ZnO (100, 002, 101) dengan struktur kristal heksagon pada semua sampel bersalut. Struktur analisis filem nipis ZnO menunjukkan peningkatan saiz kristalit daripada 13.80 nm hingga 34.35 nm manakala FWHM berkurang daripada 0.631° hingga 0.254° apabila bilangan kitaran salutan meningkat daripada 5 salutan kitaran kepada 20 salutan kitaran. Walaubagaimanapun, apabila kepekatan ZnO meningkat daripada 0.5M kepada 3.0M, saiz kristalit berkurang daripada 34.35 nm kepada 29.04 nm manakala FWHM meningkat dari 0.254° kepada 0.301°. Puncak intensiti ZnO telah menurun secara drastik apabila kedua-dua logam dimasukkan ke dalam ZnO. Filem nipis Ag-ZnO mengalami pengurangan saiz kristalit daripada 38.14 nm (3% Ag: ZnO) kepada 28.63 nm (9% Ag: ZnO) manakala filem nipis Cu-ZnO menunjukkan peningkatan daripada 33.34 nm (3% Cu: ZnO) kepada 37.48 nm (9% Cu: ZnO). Pengembangan terikan kekisi telah dilihat pada semua filem nipis ZnO berisi logam. Mikrograf FESEM untuk filem nipis ZnO menunjukkan keseragaman morfologi apabila salutan kitaran meningkat tetapi kepekatan tinggi ZnO menunjukkan ketidakseragamam taburan butir. Filem nipis Ag- ZnO mempamerkan saiz butir tidak sekata daripada 51.98 nm (3% Ag) kepada 55.54 nm (6% Ag) dan kemudiannya berkurang kepada 50.52 nm (9% Ag) kerana agregat. Walaubagaimanapun, filem nipis Cu-ZnO menunjukkan peningkatan saiz butir daripada 48.50 nm kepada 56.34 nm apabila peratusan Cu meningkat. Mod torehan AFM mendedahkan bahawa filem nipis ZnO mempunyai kekasaran permukaan yang rendah dengan nilai 17.80 nm pada filem nipis 20 kitaran salutan dan 0.5M ZnO. Purata kekasaran permukaan filem nipis Ag-ZnO meningkat daripada 23 nm kepada 46.90 nm apabila kepekatan bertambah dimana boleh dikaitkan dengan topologi yang tidak seragam. Peningkatan taburan butir oleh filem nipis Cu-ZnO mendorong kearah pengurangan kekasaran permukaan daripada 14 nm kepada 12 nm. Pengujian termal fana pakej LED terhadap salutan substrat telah dikaji untuk mengetahui rintangan haba dan suhu simpang. Keputusan menunjukkan filem nipis 3% Ag-ZnO dan 9% Cu-ZnO mempunyai prestasi termal yang baik. 3% Ag-ZnO memperolehi 10.67 K/W dan 68.32 ºC manakala 9% Cu-ZnO memperolehi 8.66 K/W dan 55.84 ºC untuk rintangan haba dan suhu simpang. Selain itu, rintangan haba telah berkurang sebanyak 22.23% dan 27.73% untuk kedua dua filem nipis berbanding filem nipis ZnO.

STUDY OF METAL IMPREGNATED ZnO THIN FILM AS THERMAL INTERFACE FOR HIGH POWER LED APPLICATIONS

ABSTRACT

Thermal issue in high power LEDs greatly impacts the performance of LED lighting in terms of its reliability and durability. Thermal interface materials have been recommended to improve heat dissipation as they are capable in conducting heat effectively between heat source and the heat sink. In this project, zinc oxide (ZnO) and two types metal impregnated ZnO (Copper and Silver) thin films were deposited on Aluminium substrates via sol-gel spin coating technique. The deposition parameters of ZnO thin films such as number of coating cycle and sol-gel concentration were varied to obtain good quality ZnO thin films. After the optimization of ZnO thin films, metal impregnated ZnO thin films were produced at concentration 0.03M, 0.06M, 0.09M. The structural properties of the thin films were investigate using XRD while surface morphology and topology of films were analysed through FESEM and AFM respectively. Thermal performance of the thin films were tested using T3Ster where high power LEDs mounted on coated substrates. X-ray diffraction results showed three major ZnO peak (100, 002, 101) with hexagonal crystal structure for all coated samples. Structural analysis of ZnO thin films showed the increment of crystallite size from 13.80 nm to 34.35 nm while FWHM reduced from 0.631° to 0.254° when number of coating cycles increased from 5 coating cycles to 20 coating cycles. However, as the ZnO concentration increased from 0.5M to 3.0M the crystallite size reduced from 34.35 nm to 29.04nm while FWHM increased from 0.254° to 0.301°. The intensity of ZnO peaks were decreased as both metal introduced into ZnO. Ag-ZnO thin films showed decrement of crystallite size from 38.14 nm (3% Ag: ZnO) to 28.63 nm (9%

Ag: ZnO) while Cu-ZnO thin films showed increment from 33.34 nm (3% Cu: ZnO) to 37.48 nm (9% Cu: ZnO). The expanded in lattice strain has been observed for all metal impregnated thin films. FESEM micrographs of ZnO thin films indicated uniform morphology as number coating cycles increased but, high concentration ZnO showed disordered grain distributions. Ag- ZnO thin films exhibited anomalous trend of grain size from from 51.98 nm (Ag 3%) to 55.54 nm (Ag 6%) and then decreased to 50.52 nm (Ag 9%) due to aggregates. However, Cu-ZnO thin films showed increment of grain size from 48.50 nm to 56.34 nm upon increasing Cu percentage. AFM tapping mode revealed that ZnO thin film had low surface roughness with value 17.8 nm at 20 coating cycles and 0.5M ZnO thin films. Average surface roughness of Ag-ZnO thin films were increased from 23 nm to 46.9 nm when the concentration increased which can related with non-uniform topology. The improvement of grain distribution for Cu-ZnO thin films led to decrement of surface roughness from 14 nm to 12 nm. Thermal transients testing of LED package on coated substrate were investigated to identify the thermal resistance and junction temperature. The results showed that 3% Ag-ZnO and 9% Cu-ZnO thin films had good thermal performance. 3% Ag-ZnO obtained 10.67 K/W and 68.32 ℃ while 9% Cu-ZnO obtained 8.662 K/W and 55.84 ℃ for thermal resistance and junction temperature. Besides that, the thermal resistance had reduced 22.23% and 27.73% for both thin films respectively compared to ZnO thin films.

CHAPTER 1

INTRODUCTION

1.1 Overview

This chapter describes the introduction of LEDs and the current issues faced in this field. Besides that, the contributions and output from this research project also have been addressed. Finally, the detailed objective of present work and outline of this thesis were highlighted.

1.2 Introduction

The revolution in the lighting industry brings light emitting diodes (LEDs) in our everyday lives where clear advantages have been offered compared with the conventional lighting sources. Innovation in lighting technology also made LEDs as one of the semiconductors that always giving better light quality. Achievement of blue light emitting diodes by Isamu Akasaki, Hiroshi Amano and Shuji Nakamura the holders for Nobel Prizes in Physics 2014 showed that LEDs technology has been improved well (Nobel Media AB, 2014). LEDs are the excellent candidate for the future lighting application due to its high brightness, low power consumption, longer lifetime, high reliability and excellent colour saturation (Chang, Das, Varde, & Pecht, 2012; Izotov, Sitdikov, Soldatkin, Tuev, & Olisovets, 2014; Teeba, Anithambigai, Dinash, & Mutharasu, 2011). Furthermore, white LEDs twice more efficient than incandescent lamp which is very beneficial for everyday use. Domination of LED application has covered from entertainment lighting and display to general lighting purpose.

Even though LEDs have huge variety of benefits, overheating in LEDs has become critical issues. Ineffective heat dissipation could lead to to enormous rise in the junction temperature that causes thermal runaway and catastrophic failures (Poppe, Farkas, Szekely, Horvath, & Rencz, 2006). The excessive heat generated at the p–n junction eventually interrupts the efficiency of LEDs performance. This phenomenon is called the hot spot problem where it absolutely results in a measurable drop of LEDs brightness and lifetime. Currently, it has been reported that the average 80-85% of the electrical power of an LED has been converted into heat instead of optical power (Cheng, Huang, & Lin, 2012). This showed that cooling solutions of an LED did not solve the rapid increase of heat generated at the active region. Therefore, these bring challenges in the thermal management of LEDs as effective thermal dissipation is crucial to avoid major drawbacks in future. Better techniques in thermal management of LEDs technology must be implement to maintain the performance of LEDs at optimum level.

Due to the thermal issue occur in the LEDs, various heat transfer have been develop by researchers and industries such as heat sink and thermal interface material (TIM) to tackle heat accumulation inside the package. TIM getting significant interest in optoelectronics area because it act as connecting path between the devices to the heat spreader by filing in the air-gaps with high thermal conducting material(Razeeb, Dalton, Cross, & Robinson, 2018) . The combination of thermal resistance and interstitial air gaps eventually decrease the overall heat dissipation. Hence, a good thermal conductivity TIM could provide better thermal management in LEDs.

A wide band gap (3.37 eV) and large exciton binding energy (60 meV) at room temperature have established ZnO as promising materials that suited for UV light emitting diodes, laser diode, light emitting diodes and even transparent electrodes in solar cells (Janotti & Van de Walle, 2009). ZnO is transparent to visible light and can be made highly conductive by doping. Many attempts have been made or modify the properties of ZnO such as adding dopants or impregnated ZnO in other to increase the multifuctionality by change the phase of the material and hence the structural defects. Besides that, ZnO has wurtzite crystal structur where the zinc atoms are arranged in hexagonal close-packed lattice. This structure is relatively open with all the octahedral and half of the tetrahedral sites empty that made it easier to incorporate dopants in the zinc oxide lattice (Singh, Kumar, Khanna, Kumar, & Kumar, 2011). In most cases, the purpose of using doping is to modulate the properties of ZnO. Since TIM has to be an electrical insulator, the chemical and temperature stability of ZnO made it capable to operate at high temperature and as insulating layer in electronic devices. Table 1 summarized the types of ZnO thermal interface material. It is found that most literatures reported that ZnO TIM were thin films. As reported by Jassriatul et. al. and Shanmugam et. al. both ZnO thin films had low thermal resistance and junction temperature eventhough had been tested with high driving current at 700mA [7] [8]. This is because the thin films had uniform coating and good adhesion to various substrates compared to paste or thick films. The usage of thermal paste as TIM had caused problems such as dry out and pump out the during power cycling, resulting in increased thermal resistance (Due & Robinson, 2013). Therefore, selection of thermal interface material was important to improve heat dissipation to the ambient.

Table 1 List of thermal resistance and junction temperature of various ZnO thermal interface materials

TIM Material	Coating Technique	Thermal resistance (K/W)	Junction temperature (℃)	Remarks	Ref
ZnO thick film	Screen printing	8.6 (500mA)	14.7 (500mA)	- 50% filler loading showed the lowest thermal properties - 60:40 of powder-to-solvent percentage of the paste	(Wen, Subramani, Devarajan, & Sulaiman, 2017a)
ZnO thin films	Chemical vapor deposition	59.40 (700mA)	151.01 (700mA)	- ZnO thin film annealed at 400 ℃ had the lowest thermal resistance	(Jassriatul Aida binti Jamaludin, Subramani, & Devarajan, 2018)
ZnO thin films	RF Sputtering	44.94 (700mA)	104.50 (700mA)	- ZnO thin film with thickness 800nm had good thermal performan ce	(S. Shanmugan, Zeng Yin, Anithambig ai, & Mutharasu, 2016)
ZnO thin films	Chemical vapor deposition	33.92 (700mA)	74.7 (700mA)	- ZnO thin film prepared at 5 sccm flow rate of O2 had the lowest thermal resistance	(Jassriatul Aida binti Jamaludin & Subramani, 2018)
ZnO thin films	RF sputtering	49.4 (700nm)	-	- ZnO thin film with thickness 200nm had low thermal resistance	(., 2015)

				and surface roughness	
ZnO thin films	Spin coating method	33.88 (700mA)	74.5 (700mA)	- ZnO thin film annealed at 400 ℃ had the lowest thermal resistance and junction temperature	(S Shanmugan & Mutharasu, 2015)
ZnO thin films	RF sputtering	32.91 (700mA)	109.08 (700mA)	- ZnO thin film with thickness 200nm had low thermal resistance and surface roughness	(Subramani & Devarajan, 2016)
ZnO filled epoxy	Co-precipitation method	34.20 (700ma)	81.45 (700mA)	- As grown ZnO filled epoxy had low thermal resistance compared to ZnO filled epoxy annealed at 500 ℃ and 700℃	(Optoelectronics, 2016)

1.3 Problem statement

LED technology has monopoly the worldwide lighting industry by making design of luminaires that are more energy-efficient, longer-lasting, more durable, and more environmentally friendly than existing lighting technologies. However, it has a maximum operating temperature in which light output and LED lifetime may degrade after used several times (Padmasali & Kini, 2017). Miniaturization of high power LEDs package and increasing operating parameters could greatly affect the light output. LEDs are designed to dissipate heat out through the bottom while light is projected up. If the heat dissipation is ineffective, it will cause accumulation of heat flux inside the dies which eventually caused excessive rise in junction temperature. Junction temperature (T_j) and thermal resistance (R_{th}) are the crucial parameters that will affect the reliability of LEDs thus, both need to be minimized as possible. It has been predicted that the lifetime of the device reduces exponentially when heat flux increases. High heat flux required excellent heat spreader such as thermal interface material to transport massive heat loads to the ambient (Razeeb et al., 2018). Hence, a new TIM with thermally conductive material need to be developed for faster heat absorption. As a result, the junction temperature and thermal resistance of LEDs package can be reduced.

Thermal interface material has been recommend to be one of the effective ways to enhance the thermal management of LEDs. TIM can be categorized as thermal pad, thermal paste, polymer composite and thin film. Selections of material are very important as material with high thermal conductivity and good adhesive strength are more favourable in order to transport heat very well. Thermal paste is the most popular TIM used in optoelectronics field as it has high thermal conductivity from metal filler. However, thermal paste could dry out after a certain period of time which make it less efficient plus, pump-out and grease dry-out during power cycling, resulting in increased thermal resistance (Due & Robinson, 2013). In this project, thin film coating was recommended as thermal interface for LEDs as it had the capability to achieve thin layer from the various method as uniform coating playing important role to reduce thermal resistance. There were numerous works on thin film as TIM and the incorporation of ZnO as the coating material. Shanmugam et. al. conducted a research on ZnO thin films with copper as the substrate and then grew the ZnO thin films by sputtering method. The thermal resistance was 44.94 K/W when tested with 700mA driving current (S. Shanmugan et al., 2016). Meanwhile, Jassriatul et. al. claimed that ZnO thin film prepared at 5 sccm flow rate of O_2 had low thermal resistance (33.92 K/W) and junction temperature (74.7 ℃) compared to thin films prepared at 10 sccm, 20 sccm and 30 sccm flow rate (Jassriatul Aida binti Jamaludin & Subramani, 2018). The ZnO thin films were deposited by chemical vapor deposition. Next, Mutharasu et. al. reported that ZnO thin film grew by RF sputtering had low thermal resistance compared to commercial paste which is 53.31 K.W and 50.19 K/W respectively (Mutharasu, Shanmugan, Anithambigai, & Yin, 2013).

In order to produce thin film, sol-gel spin coating technique was used in this research project. It has been proposed in this research project due to its low cost and easy control of stoichiometry to achieve desire TIM. Besides that, only Shanmugam et. al reported on thermal analysis of ZnO thin films deposited by spin coating method (S Shanmugan & Mutharasu, 2015). The selection of aluminium as a substrate is due to its high thermal conductivity that can improve heat dissipation to ambient despite their expensive material cost. Thus, spin-coating has to be established for metal oxide thin film coating on the metal substrate.

1.4 Objectives

There are three objectives need to be achieve in this study:

- To fabricate zinc oxide and metal impregnated zinc oxide thin film on Al substrate by using sol-gel spin coating method
- To analyse the influence of metal impregnation on structural, surface and thermal characterization of ZnO
- To evaluate the thermal performance of thin film as thermal interface material in LEDs

1.5 Thesis outlines

This thesis consists of 6 major topics which includes introduction, literature review, theoretical background, methodology, results and discussion and finally conclusion and recommendations

i. Chapter 1: Introduction

This first chapter highlight the introduction to light emitting diodes and current issues face by the LEDs. Objectives of the research also has been included

ii. Chapter 2: Literature review

The second chapter focuses on thermal management of LEDs followed by benefits of using thermal interface material in LEDs. Apart from that, literature review of previous work on uses thin film as TIM and synthesis of ZnO thin film also briefly explain in this chapter.

iii. Chapter 3: Theoretical background

In third chapter, general introduction of zinc oxide, copper doped zinc oxide, silver doped zinc oxide with their properties are discussed here. Besides, the theory and principle of sol-gel method and spin coating deposition technique are explained detail in this subchapter. A brief description on thermal transient measurement as well as thermal resistance and junction temperature are also included.

iv. Chapter 4: Methodology

This chapter describes in detail the methodology used in this project. This involves the synthesis of the thin films and characterization done on coated substrates.

v. Chapter 5 : Results and Discussion

This chapter presents the results of the data collected from the thin films analyses and LED thermal characterization. The results are presented in a graphical and figure formats for every analysis equipment

vi. **Chapter 6: Conclusion and Recommendations**

In this last chapter, the results of the work are summarized, and conclusion of the project is presented. Future suggestions and recommendations to enhance this research are also included in this chapter.

CHAPTER 2

LITERATURE REVIEW AND THEORITICAL BACKGROUND

2.1 Overview

In this chapter, it contains scientific literatures information on the concept of thermal management in LEDs and the importance of its in LED application. Apart from that, the benefits of using TIM on enhancement heat dissipation in thermal package design are also included. Lastly, this chapter also presented the literatures on deposition techniques used in synthesis of undoped ZnO and Metal (Cu, Ag) doped ZnO thin film. Therefore, this chapter is focusing on:

- Thermal management in LEDs
- Impact of Thermal Interface Material in LEDs
- Thin films as Thermal Interface Material in LEDs
- ZnO Thin Film deposition methods

2.2 Thermal management in LEDs

Light emitting diodes (LED) development has become very active research field nowadays where it is primarily focus in markets such as car headlamps, mobile phones and other electronic applications. LED has being develop to improve the solid state lighting technology due to its outstanding performance such as long lifetime, high efficiency and controllable of optical output (P. Huang, Pan, Wang, & Chen, 2012). Previous generations of incandescent lighting converted less than 5% of electrical power input into light while the remaining power being dissipated as excess heat. After a few years, incandescent fixtures has slightly improved but new lighting technologies, including LED systems has made better efficiency. Therefore, a proper thermal management is required in order to achieve reliability and better performance of LED lighting. Improper thermal management and heat could becoming the greatest obstacle in LED technologies. These relate with the continuous amount of heat that could degrade the performance of the LED and rise the mechanical failure in the system (“Efficient thermal management solutions for LED systems - ElectronicsB2B,” n.d.).

Thermal management can be divided into internal thermal management and external thermal management. Internal thermal management can be classified as the transfer of heat from the p-n junction to the heat sink or luminaire body (Vossloh-Schwabe Deutschland GmbH, n.d.). This heat transfer can be relate to thermal conduction mechanism where the efficiency is reliant on materials used and the geometry of LEDs. The material used should have high thermal conductivity such as aluminium and copper for optimum heat transfer along LED module to luminaire body. Apart from that, air gaps between the layer paths need to minimize at any cost by applying suitable thermal interface material like thermally conductive pad or pastes. External thermal management is referring to heat dissipation from heat sink or luminaire body to the ambient air (Müller & Müller Gmbh, 2014). Convection and heat radiation are main processes involve in this dissipation where LED are solely dependent on them as can be seen in Figure 2.1 (“Basics of Heat Transfer: LED Fundamentals,” n.d.). Excellent convection can be achieved by having wide surface area with continuous air circulation. Normally, heat sink has been used to improve the convection heat transfer. In terms of heat radiation, the temperature and surface area of the Led are the main aspect.

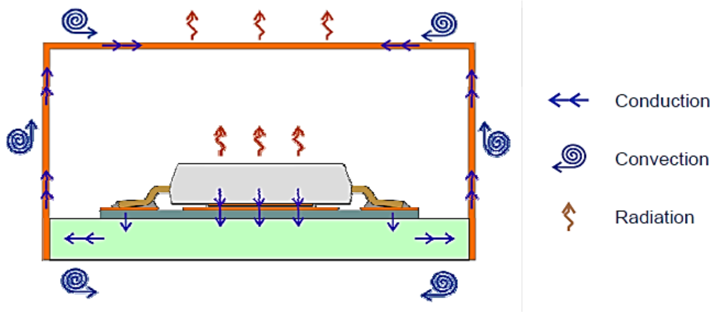

Figure 2.1 Heat transfer from the die source to the surrounding [5]

The purpose of the LED lighting is to minimize the maintenance cost and operating in the luminaires for building long lasting light sources. The main source of the heat generation in LED are usually happens in the junction between the p-type and n-type semiconductor material (Steven Keeping, 2015) . The thermal dissipation path of LED system shown in Figure 2.2 was illustrated with simple resistor network of an LED on a printed screen board (PCB) mounted to a heat sink in ambient air. From this thermal path, it can be summarized that heat conducted from heat source (Q) through the LED package to PCB which then flow through the thermal interface material (TIM) to the heat sink and finally convected to ambient air. By maintaining the low value of critical parameters such as junction temperature, T_j and thermal resistance, R_{th} shown in Figure 2.3 this will further improve the performance and reliability of the LEDs (LED ACADEMY, n.d.). Thus, the thermal conduction path need to manage well in order to improve the performance of heat removal (Ben Abdelmlek et al., 2016) .

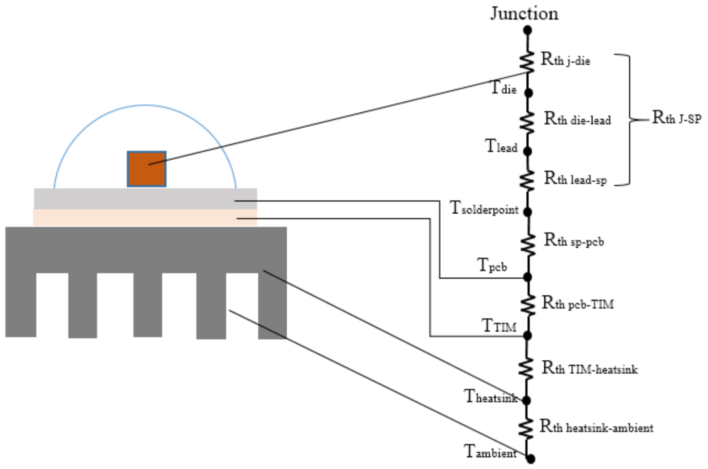

Figure 2.2 Thermal resistor model of a single LED array mounted on heatsink

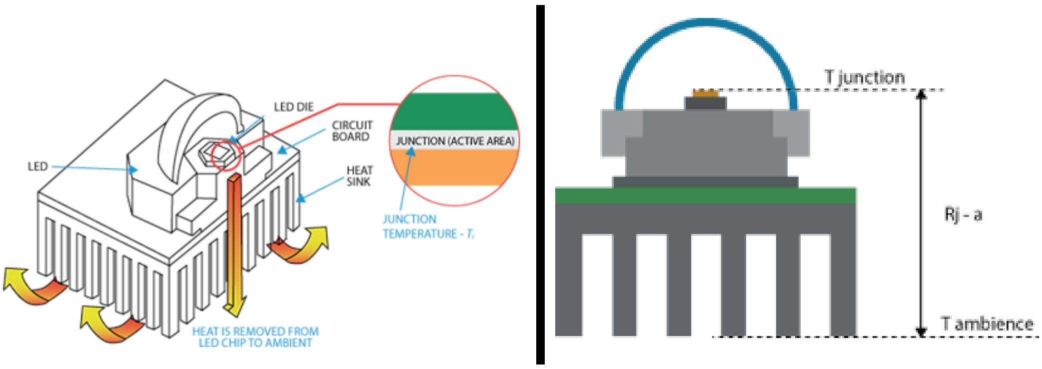

Figure 2.3 Basic parameters to evaluate LED performance [7]

It is widely known that the luminaires of LEDs are strongly depend on the operating conditions such as driving current and forward voltage. As the current is supplied in forward direction, a part of electrical energy will change into incoherent narrow spectrum of light. In order to allow the current to flow across the LED a certain voltage is needed which only light can be emitted (King, n.d.). As the forward current increase, more light will be generated from LED. However apart of light, heat energy also produce at the die junction area which must be dissipated to ambient to avoid self-heating issue. This thermal heat can be related to current crowding phenomenon which is non-uniform lateral temperature distribution in the chip. Current crowding generates localization of heat and causes increases in active region temperature. It has been reported by Yursaven that the luminous efficiency has significantly drops with the increment of driving current as the value of ambient temperature is fixed (Yurtseven, Mete, & Onaygil, 2016). It can be said the luminous output is expected to degrade over time because of junction heating from high forward current. In general, LED only has 20 to 30% photoelectric conversion efficiency while the rest of 70% of the total energy is converted into heat (J. Chen et al., 2013). Due this reason, the higher the temperature of the LEDs operate, the less efficient they become. Thus, improvement in thermal management is essential for optimal performance.

2.3 Impact of Thermal Interface Material in LEDs

Heat removal has becoming the main challenge for high power LEDs to maintain the lifetime and optical performance. Before heat can be transferred to ambient, it has to pass many junctions which cause heat to accumulate inside. By using thermal interface material (TIM), mating surfaces between two contacts can be improved to increase thermal transfer. TIM are thermally conductive materials which design to fill in any air gaps and thus reduce the thermal resistance (Singhal, Siegmund, & Garimella, 2004). A variety of thermal interface material have been establish in response to the needed of the LEDs applications. TIM is one of important media in dissipating the heat out from the LED package to the board or heat sink. By properly applying TIM this can help eliminating the air gaps while improving the thermal dissipation. Ling reported an air gap between two irregular solid surfaces can interrupt the heat transfer therefore TIMs will use to conform microscopic surface contours of the adjacent solid surfaces in order to improve surface contact between the LED heat source and PCB/FR4 PCB(Luger, 2007) . As a result, it is able to reduce the temperature drops across this contact.

In any interface material, thermal conductivity and thermal contact resistance are the main aspects that need to be consider. Chung et. al. reported that if a great conductor material is chosen, the effectiveness of thermal interface material in LEDs will be boost due to its high thermal conductivity and low contact resistance (Chung, 2001). The material will maximize the thermal conductivity and reduce junction temperature of the LEDs by filling in the voids with highly conductive substances such as metal oxide. Distortion of surfaces is the main cause of thermal contact resistance as the gaps filled with low thermal conductivity air (Sarvar, Whalley, & Conway, 2006) . Eventhough PCBs and heat sinks are properly design, minor surface imperfections will still exist which disrupt the efficiency of thermal transfer due the surfaces of the two materials are partially separated by air pockets. Therefore, TIM are really recommended as it is capable to filling in the air gaps as shown in Figure 2.4. As long as the thermal interface material is more thermally conductive than air, its presence can improve the thermal contact. On the other hand, if the thermal interface material is relatively large in thickness, the thermal conductivity will be more important (Leong & Chung, 2004).

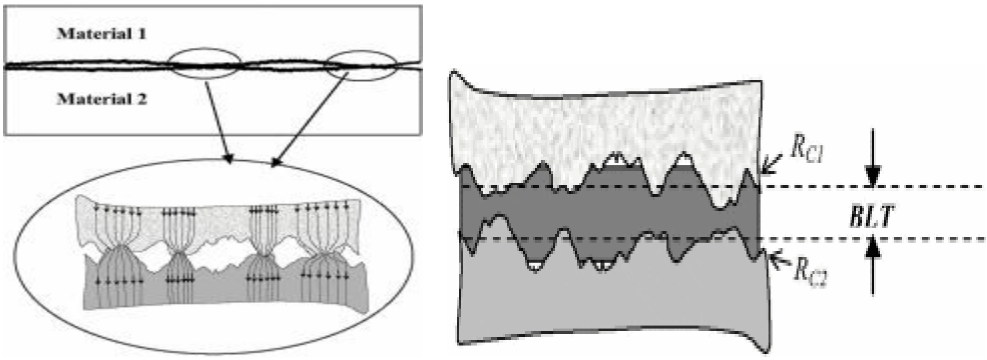

Figure 2.4 Thermal interface material filling in the gap between two surfaces (Sarvar et al., 2006)

TIM can be categorized as epoxies compound, thermal greases, gap filling pads, sintered metallic TIMs and even thin film (Im & Kim, 2012; Subramani Shanmugan, Mutharasu, & Hassan Haslan, 2012; Y. Tang, Liu, Yang, & Yang, 2016). Various research study have showed the benefits of using thermal interface material for better thermal management. Yunxia reported gallium based thermal interface showed decrement in thermal contact resistance and thermal conductive resistance at increase pressure. The gallium thermal paste obtained thermal interface resistance 2.6 $mm^2\ kW^{-1}$ which is much lower than commercialized thermal greases (Gao & Liu, 2012). Ceramic-epoxy composites which used Aluminium Nitride as filler has improved the thermal performance of LED. This epoxy composites showed reducing in junction temperature and total thermal resistance when amount of filler loading increased (Anithambigai, Mutharasu, Huong, Zahner, & Lacey, 2014). Mah et al reported that applying silver doped ZnO thick film between LED and Al substrates could give low junction temperature even at high driving current (Wen, Subramani, Devarajan, & Sulaiman, 2017b). Meanwhile, carbon nanotube TIM tested on high brightness LED was achieve total thermal resistance 7 $mm^2\ K\ W^{-1}$ compare to ‘air’ that had 107 mm2 $K\ W^{-1}$ (Zhang, Chai, Yuen, Xiao, & Chan, 2008). Hence, a suitable thermal interface material is essential for maximising the heat transfer between two surfaces.

2.4 Thin films as Thermal Interface Material in LEDs

The existing of air gaps in irregular surfaces can lead to thermal contact resistance which will affect the performance of LEDs. Therefore, this thermal problem can be avoided by filling in the air gaps with thermal interface material in order to achieve good thermal contact. By applying TIM, this could reduce the heat accumulation inside LED package which give benefits in longer lifetime of LEDs. Thin film have been used extensively in current research as thermal interface material due to its uniform coating and good adhesion to various substrates. Thin film can be classified as a layer of material on certain substrates made by intensifying and one-by-one from any ionic or molecular types of matter (Wasa, Kitabatake, & Hideaki, 2004). Apart from that, thin film is selected because the bond layer thickness can be control and minimize in any deposition.

Several researcher had reported on thin films based thermal interface material which showed significant improvement in thermal properties. ZnO thin films synthesized on Al substrates by Shanmugam et. al. were used as TIM and tested with

3W green LED package. Thermal behaviour of tested films were compared with paste and bare Al at different driving current. Transients graph in Figure 2.5 revealed that ZnO coated substrates had lowest total thermal resistance than other samples (Subramani Shanmugan, Mutharasu, & Hassan, n.d.). Besides that, Lim et. al. had studied the thermal resistance of Cu-Al_2O_3 thin films as TIM in LEDs. The deposited films had undergone different annealing temperature and were compared with bare Al. Detailed results in Table 2.1 showed Cu-Al2O3 thin films annealed at 300ºC obtain significant difference of ΔTj= 12.3º and ΔRth= 5.28K/W compared to bare Al substrates (Qiang & Shanmugan, n.d.). It was reported by O.Z. Yin et. al. that Boron doped AlN(B-AlN) thin film based interface material can conduct heat efficiently to ambient. B-AlN deposit at high temperature with low thickness showed lowest thermal value when tested at 700mA (Ong, Shanmugan, & Mutharasu, 2015). Later work by Shanmugam et. al. also proved that thin film is capable enough to reduce heat in LEDs. Zn thin film as TIM was coated at different thickness and annealing temperature to evaluate the thermal performance. The tabulated results in Table 2.2 indicated high thickness and annealing temperature on Zn thin films delivered low junction temperature which is good in delivering heat (S & Mutharasu D, 2016). Strong evidence from the literatures above showed that thin film is suitable for the thermal interface material in order to enhance heat dissipation from LEDs package to surrounding environment.

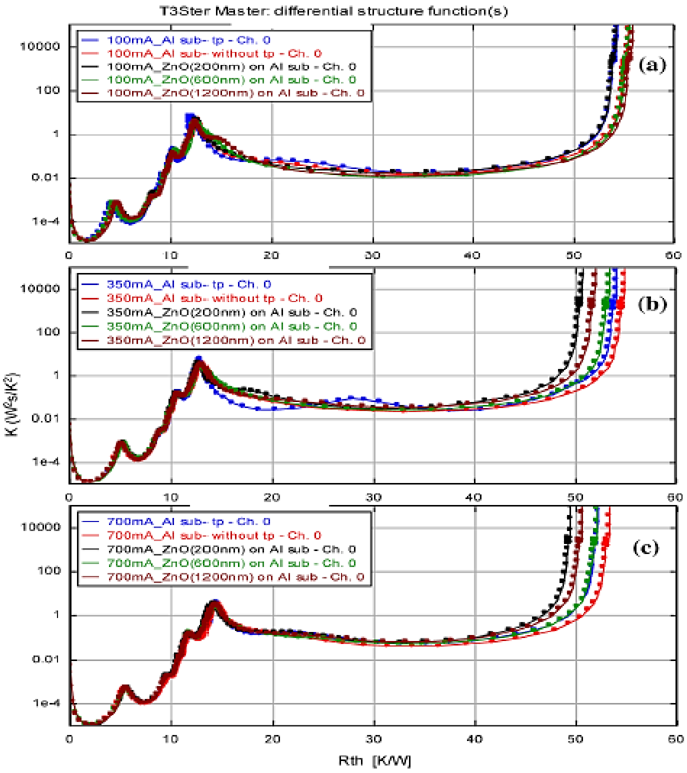

Figure 2.5 Differential structure function of tested samples at different driving current (Subramani Shanmugan et al., n.d.)

Table 2.1 Thermal resistance and junction temperature of bare Al and Cu-Al_2O_3 thin films (Qiang & Shanmugan, n.d.)

Driving current (mA)	Bare Al	Cu-Al_2O_3 un-annealed	Cu-Al_2O_3 annealed (200°C)	Cu-Al_2O_3 annealed (300°C)	Cu-Al_2O_3 annealed (400°C)	Cu-Al_2O_3 annealed (500°C)
Total thermal resistance, R_{th}(K/W)						
100	35.97	37.53	39.74	30.27	35.27	37.02
300	35.82	37.42	36.90	31.20	35.98	36.14
500	36.63	38.45	37.57	32.23	36.76	36.96
700	37.98	39.91	38.91	32.70	37.85	38.29
Substrate to ambient thermal resistance, $R_{th\ s\text{-}a}$ (K/W)						
100	23.65	26.04	22.37	19.23	24.30	26.49
300	23.34	25.02	24.61	19.45	23.55	24.24
500	23.27	25.41	24.38	19.95	24.06	24.44
700	23.25	25.74	24.11	19.58	23.89	24.36
Junction temperature, T_j(°C)						
100	10.27	10.70	11.32	8.67	10.09	10.63
300	34.24	35.83	35.15	30.12	34.41	34.59
500	62.06	65.12	63.92	55.34	62.35	62.82
700	95.51	100.14	97.83	83.23	95.44	96.31

Table 2.2 Junction temperature of Zn thin films at different thickness and annealing temperature (S & Mutharasu D, 2016)

Annealing Temperature	300nm Zn/Cu	400nm Zn/Cu	500nm Zn/Cu	800nm Zn/Cu
		300mA		
150°C	45.3631	46.0265	47.5839	46.5815
350 °C	47.1338	48.2196	47.3963	47.7493
		500 mA		
150°C	88.9892	89.2528	93.0877	91.6713
350 °C	93.1049	90.5062	91.3372	93.7076
		700 mA		
150°C	146.827	148.018	152.364	151.158
350 °C	155.083	149.054	149.931	154.247

Recently, zinc oxide material have been widely used in various field especially optoelectronics devices. ZnO is preferred than GaN because the raw materials are costly for nitride and it uses NH_3 to produce nitride thin film which is quite hazardous. ZnO can be classified as excellent alternative to this expensive material. Besides that, high temperatures process of GaN will limit the usage of substrate material in this project. ZnO can be classified as promising photonic material in ultraviolet region because it is a direct bandgap and have low threshold voltage. The excellent properties such as good thermal conductivity (1-1.2W/mK) and low thermal expansion coefficient of ZnO thin film meet the requirement of good quality thermal interface material (Pearton, Norton, Ip, Heo, & Steiner, 2005).

There are several works on applying ZnO thin film as TIM that have been reported by researchers. The ZnO thin films synthesized by Jassriatul et. al. showed good thermal results when tested with LEDs. The ZnO coated Al substrates had been deposited with various flow rate and had been compared with non-coated Al substrate. Thermal transients results in Table 2.3 showed all the ZnO coated substrates had low value junction temperature and thermal resistance than non-coated substrate (Jassriatul

Aida binti Jamaludin & Subramani, 2018). Meanwhile, Mutharasu et. al. conducted a study of ZnO thin films on Al substrate as TIM prepared by using RF sputtering. The coated substrates was then compared with bare Al substrate and thermal paste coated substrate. Thermal transient analysis in Figure 2.6 revealed that ZnO coated substrate had the lowest total thermal resistance compared to other tested samples (Mutharasu et al., 2013). Shanmugam et.al. also deposited ZnO thin film on Cu substrate with different thickness (400nm and 800nm) where the thermal properties of LEDs were studied for different TIM. Figure 2.7 showed the variation junction temperature of the samples where 800nm of ZnO thin film possessed lowest Tj even at high driving current. The huge difference of Tj between 800nm (ZnO) sample and bare Cu sample give value $\Delta Tj = 11.83°C$ which indicate ZnO thin film increase the contact between mating surfaces and improve the heat transfer (S. Shanmugan et al., 2016). Based on the explanation above, Table 2.4 summarized the lowest junction temperature and thermal resistance tested at 700mA driving current from various thin films. Therefore, it can be concluded that ZnO thin film is recommend to be apply as thermal interface material in LED as it capability to improve heat dissipation. The thin film will be synthesized by using sol-gel spin coating method.

Table 2.3 Summarize of Rth and Tj of ZnO thin films coated on Al substrates and bare Al substrates (Jassriatul Aida binti Jamaludin & Subramani, 2018)

	Bare Al	ZnO 5 sccm	ZnO 10 sccm	ZnO 20 sccm	ZnO 30 sccm	ZnO 40 sccm
Junction to ambient Thermal Resistance, $R_{th\text{-}tot}$ (K/W)						
100 mA	34.73	32.20	32.48	32.48	32.33	34.85
350 mA	35.21	32.78	33.69	34.19	33.42	34.50
700 mA	36.06	33.92	34.72	35.51	35.11	35.68
Junction Temperature, T_j (°C)						
100 mA	9.5	8.9	9	9	9	9.6
350 mA	36.6	34.1	35.1	35.6	35.1	36.6
700 mA	79.5	74.7	76.6	78.2	77.3	79.9

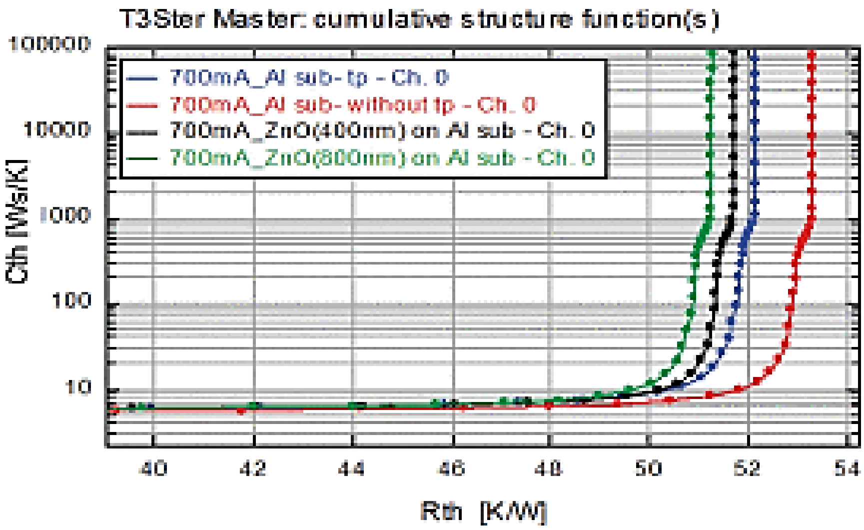

Figure 2.6 Cumulative structure function curve of 3W LED fixed on ZnO thin film coated Al substrates recorded at 700 mA (Mutharasu et al., 2013)

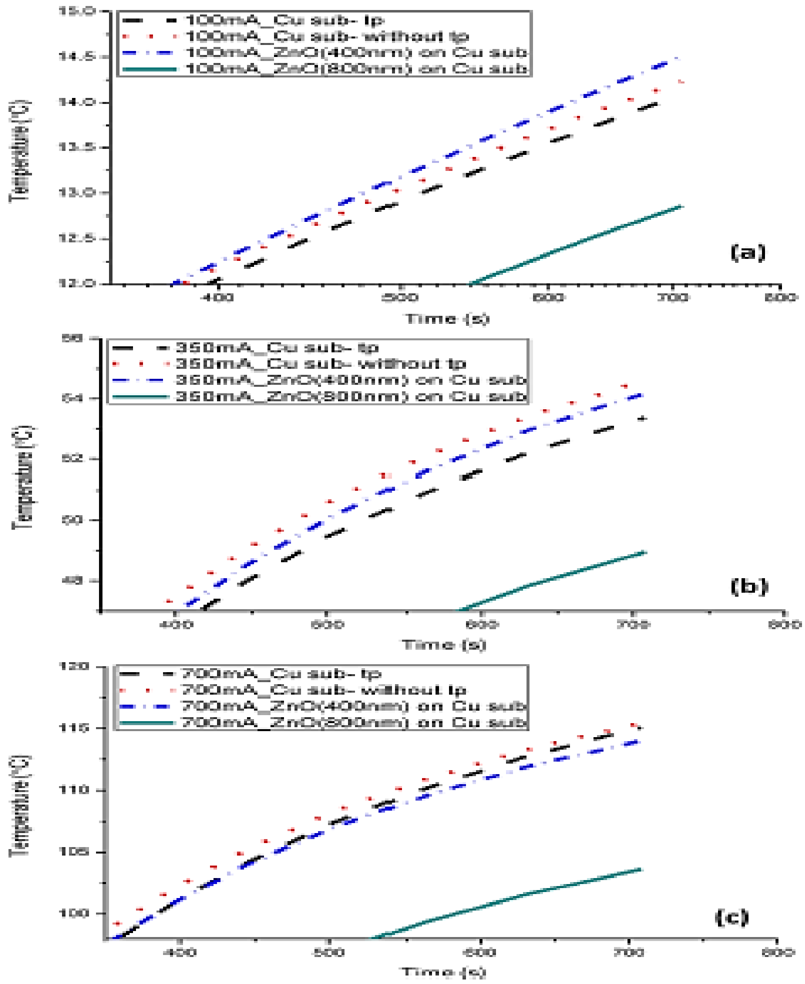

Figure 2.7 Junction temperature rise of different sample at (a) 300mA, (b) 500ma, (c) 700mA driving current (S. Shanmugan et al., 2016)

Table 2.4 Thermal resistance and junction temperature of various thin films prepared at different coating technique

TIM Material	Coating Technique	Thermal resistance, R_{th} (K/W)	Junction temperature, T_j (°C)	Samples	Ref
ZnO thin film	RF sputtering	49.46 (700 mA)	-	- ZnO thin film with thickness 1200nm had the lowest Rth compared to bare Al substrates and commercial paste	(Subramani Shanmugan et al., n.d.)
Cu-Al_2O_3 thin film	RF sputtering	32.70 (700mA)	83.23 (700mA)	- Cu-Al2O3 thin film annealed at 300°C achieved low Rth and Tj than other annealed samples and non-annealed samples	(Qiang & Shanmugan, n.d.)
AlN(B-AlN)	RF sputtering	53.27 (700mA)	120.98 (700m)	- B-AlN thin film annealed with temperature 200 °C had low Rth and Tj compared to thin film annealed at room temperature	(Ong et al., 2015)
Zn thin films	DC sputtering	58.85 (700mA	149.054 (700mA)	Zn thin film annealed at 350 °C had low Rth and Tj compared to annealed	(S & Mutharasu D, 2016)

				sample at 150℃	
ZnO thin film	Chemical vapor deposition	33.92 (700mA)	74.7 (700mA)	ZnO thin film prepared at 5 sccm flow rate of O_2 gas had the lowest Rth and T compared to thin films prepared at flow rate 10 sscm, 20 sccm and 30 sccm	
ZnO thin film	RF sputtering	51.19 (700mA)	117.46 (700mA)	ZnO thin film with thickness 800nm had low Rth and T compared to bare Al substrate and commercial thermal paste	(Mutharasu et al., 2013)