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Thermal Diffusivity measurements of Hybrid Boron Nitride Nano Flakes, Nano Tubes and h-BN film on Copper substrate for LED thermal management

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Abstract

Increasing Power density due to miniaturization in modern electronic devices and growing higher flux light emitting diodes (LEDs) all lead to higher junction temperature requiring proper thermal management to meet the reliability and performance requirements. Minimizing the thermal resistance from the device junction to the external heat sink mounting bottom surface of packaging support substrate is critical to maintaining lower junction temperature. In order to achieve this objective, thin film electrically insulating, thermally conducting layer formed by drop cast drying process of a hybrid Boron Nitride (H-BN) solution, a combination of BN Nano Flakes (BNNF), Boron Nitride Nano Tube (BNNT) on Copper substrate with graphite layer and Copper substrate with a Monolayer h-BN (hBNmL) have been studied by measuring Thermal Diffusivity using Laser Flash Method. The Thermal diffusivity measurements show that the addition of 1 wt% Boron Nitride Nano Tubes to Boron Nitride Nano Flakes in Ethanol solution and drop cast as thin film on a Copper substrate with Graphite layer increases the Thermal diffusivity by 2.05% as compared to Boron Nitride Nano Flakes alone and on a Copper substrate with a monolayer h-BN in the place of the graphite layer increases the Thermal diffusivity by 23.96%. Further, to evaluate the influence of wt% of BNNT in the H-BN solution, different bare Copper substrate samples were coated with H-BN solution with BNNT weight percentage of 0.3%, 1.0%, 1.5% and 2% and the Thermal diffusivity was measured and the improvement in Thermal diffusivity with respect to bare copper were 5.5%, 9.3%, 14.6% and 16.1% respectively indicating impact of increasing wt% of BNNT in increasing the Thermal diffusivity. Thermal transient measurements of LED on FR4 board with this H-BN solution coating with 1wt% BNNT, reduced the thermal resistance by 4.22% as compared with FR4 Packaging substrate without coating. Thus, the H-BN coating has potential use as thermal heat spreading layer in all applications of LEDs including Li-Fi (Light Fidelity) communication LED packaging applications where fast On-Off switching of LEDs needs to happen.

Keywords:

LED;
Boron Nitride;
BNNT;
Thermal Management;
Thermal Diffusivity.

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1. Introduction

LEDs are ubiquitous now a days replacing fluorescent and incandescent lamps all over the world because of their energy efficiency. Use of LEDs has gone beyond just lighting to horticulture for indoor farming to medical disinfectants as UV-LEDs. Thermal management of LEDs at all levels from device to Packaging to luminaire level helps to increase External Quantum Efficiency (EQE) of the devices, reliability, life span and other aspects like color quality etc. So, it has become imperative to find and use suitable materials and processes at every step of the LED application to improve the overall performance. Another important area is use of light for communication as in the emerging LiFi to potentially supplement WiFi with anticipated growing device connectivity and faster communication requirements. Higher current increases light output of LED with an increase in the Internal Quantum Efficiency (IQE) of LED. However, increasing current density generates more heat which tends to decrease the IQE.Not all the power supplied to an LED is converted into light and a considerable portion of the power $\mathbf{P_{th}}$is dissipated in the form of heat. If V is the forward voltage of the LEDand I the current, IQE the Internal Quantum Efficiency and LEE the light extraction Efficiency then $\mathbf{P_{th}}$can beexpressed as :

$$\mathbf{P_{th}} = (1\text{-LEE x IQE})\ I \text{ x } V \qquad (1)$$

The more the heat dissipated the higher the LED junction temperature goes and lower the IQE becomes.

In an LED when the voltage increases to certain point, the LED starts functioning and after some point very small change in voltage will lead to large change in current increasing the current density resulting in more heat dissipation and higher temperature reducing the IQE. So, thermal management becomes critical to maximize the IQE.

2-D materials like Graphene and Boron Nitride have been finding applications both at device and packaging levels because of their high thermal conductivity. However, graphene being electrically conductive cannot be used in the exposed faces of electronics and Boron Nitride in its crystalline hexagonal form as Hexagonal Boron Nitride (h-BN) being electrically insulative lends itself suitable for thermal management with high thermal conductivity and chemically inert properties. Interest in monolayer hexagonal Boron Nitride h-BN started with Novoselov the Nobel Prize for Physics in 2010 pointing out the possibility of 2-D like h-BN in 2005, following his publication on Graphene the most popular 2-D materialin 2004[1]. As a structural analogue of graphite h-BN has alternating Boron and Nitrogen atoms within honeycomb lattice structure replacing carbon atoms. While Graphene is highly electrically conductive with equally high in-plane thermal conductivity in the range of 1800 to 5300 W/m K at room temperature as suspended Graphene [2] and its potential application in nano electronic circuits thermal management was pointed out by S. Ghosh et.al. [3], graphene cannot be used for thermal application in exposed areas of electronic applications where electrical insulation is required. Also, once the graphene layer is supported as is required in many applications, the thermal conductivity could drop to 600 W/mK as in the case of SiO2 support [4] but is still much more than copper. It is here that h-BN which is electrically insulative but has relatively high Thermal conductivity as compared to other insulating materials provides advantage. Thermal conductivity of 751 W/m K for single layer, 646 W/m K for a Two Layer and 602 W/m K for 3 Layer suspended single crystalline, atomically thin Born Nitride without polymer contamination was reported by Quiran Cai et. al. [5]. Experimental Thermal conductivity of few-layer BN were reported by Jo et al. [6] with a value of about 250 and 360 W/m K respectively for 5Layer and 11 Layer BN. Nano tubes of Boron Nitride (BNNT) are similar to Carbon Nano Tubes (CNT) with good thermal conductivity, electrically insulative property and resistant to neutron radiation with potential use in space application environment. Boron nitride has been studied in the form film for graphene devices [7], as nano sheet as AlNB nanocomposite as heat spreading Solid Thermal Interface Material (STIM) [8.], as Boron Nitride Nano Sheets along with graphene to improve epoxy composite thermal conductivity [9] or using graphene supported BNNT to improve thermal performance of polymer nano composites as in the study by Xu Li et.al. for graphene supported BNNT [10].Eventhough Chemical Vapor Deposition (CVD) produces high quality 2-D materials, their application in optoelectronic device like LED has not been widely adopted due to specialized processing, low yield and high cost hence solution processable 2D materials can prove to be alternative avenue as pointed out by Ricciardulli et al. specifically with respect to Graphene as Transparent Conducting Electrode (TCE) and also indicating that studies related to use of solution processed 2-D materials as Electron Transport Layer (ETL) still missing and BN was not mentioned as solution processable material in this paper[11]. So,in this paper, effect on the Thermal Diffusivity of a hybrid BNNF-BNNT in the form of H-BN solution drop cast and dried to form thin film electrically insulative coating on LED Packaging substrate is studied using graphite on Copper as the base material and h-BN on Copper as another base material. In addition, Transient thermal measurements of LED mounted on FR4 board with and without H-BN solution coating were done to compare the LED junction to Package substrate bottom thermal resistance.

2. Research Method

2.1 Materials used and Samples preparation

The Boron Nitride Flakes were used in the form of 30 wt% dispersion in Ethanol solution and was procured from US Research Nanomaterials with 99.8% purity BN Nano flakes of 80nm size. The BNNTs were from BNnano inc. in powder form. These are BN nano tubes that have nanocrystals of h-BN on the outer surface of the nano tubes also called NanoBarb^{TM}. The wall count is 1 to 3 with nominal 60 nm barb width. Graphene used is in the form of sheet from Wurth Electronik. Also, a CVD deposited monolayer h-BN film on Copper substrate in the form 20 micron film was procured from Grolltex for the study.

In this study, in the First set of samples, Five different sample types were prepared with four of them using 25mm diameter 0.60mm thick copper as base substrate material. The fifth sample used is 25 mm diameter, 20 micron thick copper with the CVD monolayer h-BN film on top as base substrate material. First sample which is the 25 mm diameter Bare copper (Cu) sample is used as a reference material for Thermal diffusivity measurements. The Second sample had only the Graphene sheet layer on top of copper (Cu+GR). Graphene was chosen because of its structural compatibility with hexagonal Boron Nitride. The Third sample (Cu+GR+BNNF) had copper, Graphene and on top the BNNF solution drop cast and dried to form a thin film of h-BN. Fourth sample (Cu+GR+BNNF+BNNT) is basically like the Third sample except that the BNNF solution now had 1 wt% BNNNTs added to it. The Fifth sample (Cu+ hBNmL+ BNNF+BNNT) does not have Graphene layer and has only copper with monolayer h-BN film with BNNF solution containing 1 wt% BNNTs drop cast in the form of film. Thermal diffusivity of the samples was measured as described in section 2.2.

In a second set of measurements, the influence of wt% of BNNTs in the Hybrid BNNF+BNNT (H-BN) solution on thermal diffusivity was studied using 5 samples. Again, taking the first sample Bare copper as reference, Four other 25mm diameter, 0.60 mm thick Copper samples were coated with thin H-BN solution containing 0.3 wt%, 1.0 wt%, 1.5 wt% and 2.0 wt% of BBNT respectively to form thin film. Thermal diffusivity of these samples was also measured in the same way as the first set of samples.

2.2 Thermal Diffusivity Measurement by Laser Flash Method

Laser flash method has been established as one of the versatile tools to measure thermal diffusivity of different materials [12]. In this study, thermal diffusivity of the 25mm circular samples were measured at room temperature using Netzsch LFA 467 system. The sample is placed in a holder and the bottom surface is heated using a laser beam and an IR detector measures the temperature increase on the upper sample surface. The laser flash system was continuously purged with nitrogen gas at 40ml/min rate. The temperature rise on the back side of the sample can be solved as a onedimensional thermal conduction equation under ideal conditions and the temperature rise can be expressed as [13]:

$$\Delta T = \Delta T_m \left[1 + 2 \sum_{n=1}^{\infty} -1^n \exp(-n^2 \Pi^2 \alpha t / L^2) \right] \qquad (2)$$

Where α is the thermal diffusivity, ΔT is the temperature rise in the sample, $\Delta T_m$is the maximum rise in temperature, t is the time after pulse heating and L is the sample thickness.

Setting the time when the rise in temperature reaches half the maximum value ($\Delta T / \Delta T_{m)} = 1/2$ as $t_{1/2,}$ thermal diffusivity can be calculated as:

$$\alpha = (0.1388 L^2 / t_{1/2}) \qquad (3)$$

From the measured thermal diffusivity values, thermal conductivity can be calculated using the relationship

$$k = \alpha \rho C_p \qquad (4)$$

where, k is the thermal conductivity, α is the measured diffusivity, ρ is the bulk density and C_p is specific heat capacity of the material.

2.3. SEM and Raman Spectrum Analysis

SEM analysis was done on Sample 4 which is described in Table 1. Sample 4 has Graphene film on Copper as the base substrate with the coating of hybrid solution H-BN containing BN Nano Flakes and 1wt% BN Nanotubes. SEM pictures of the Top H-BN coated Sample 4 is shown in Figure 1. The Left picture in Figure 1., shows the BN Nano flakes mainly and some BN Nanotubes and the Right picture with a close up view shows the BN Nano Tube clusters in the midst of BN Nano Flakes.

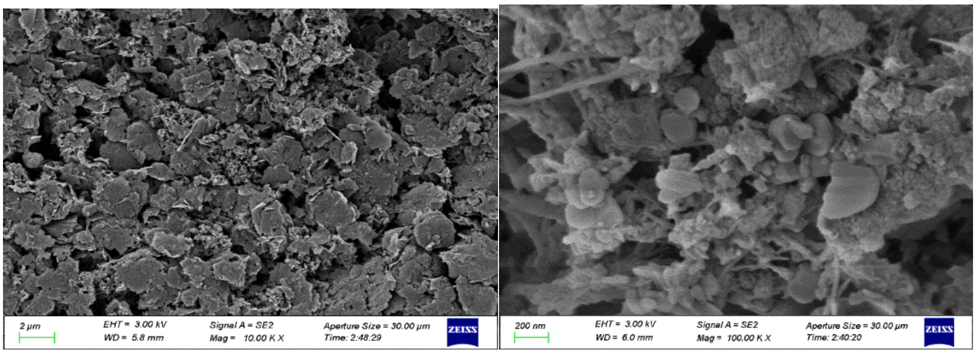

Figure 1. SEM Pictures of Sample 4 showing the Top H-BN coated surface with BNNF and BNNT.

For comparison, SEM was done on Sample 2 which has only Graphene film on copper without any H-BN coating. The SEM pictures of the Graphene Film layer are shown in Figure 2. in which the first two pictures from the Left are for Graphene and the Third picture is that of H-BN for comparison of relative colors. From the pictures it can be seen that the particles in the Graphene layer is relatively dark compared to the H-BN which is seen whiter. Hexagonal Boron Nitride is also called as the "White Graphene because of its structural similarity with Graphene.

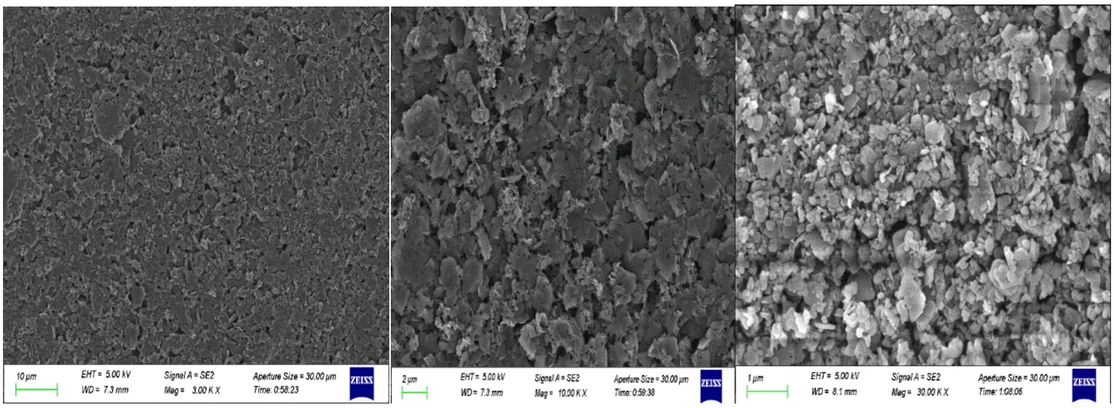

Figure 2. SEM pictures of Sample 2. The First Two pictures from the Left are that of Graphene layer and the Third picture shows BN for comparison of color.

Next, Raman spectrum analysis of Sample 4 with H-BN coating containing BNNF and BNNTwith Graphene on Copper as the base substrate was done to capture the Raman signatures of BN. Figure 3., shows the Raman spectra of the Sample 4.

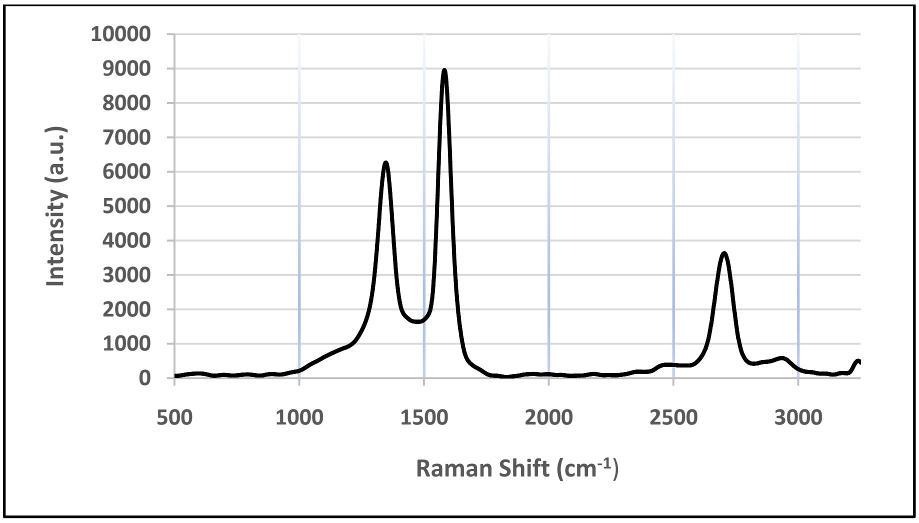

Figure. 3. Raman Spectra of Sample 4 with H-BN coating with underlying Graphene layer.

Three Peaks are seen at 1347.1, 1582, 2710.32 cm^{-1} and a small bump like peak at 2934 cm^{-1}. This is similar to the Raman spectrum of G/hBN with peaks at 1348,1576, 2664 and 2917 cm^{-1} as in reference [14]. The h-BN/ /Graphitic system in this study shows similar broader E2g peak for h-BN at 1347 cm^{-1} as compared to 1348 cm^{-1} given in the reference, both showing a red shift.

3. Results andDiscussion

3.1Thermal diffusivity of First Set of Five samples:

Measured Thermal diffusivity of the First set of Five samples are shown in Table 1. The measured thermal diffusivity comparison chart is shown in Figure 4.

Table 1. Measured Thermal Diffusivity of First Set of samples

Sample No. and Description	Measured Thermal Diffusivity (mm^2/s)	% Increase over Bare Cu	% Increase over Cu+GR+BNNF. Comparing BN coatings
1. Cu	116.13	Reference	N/A
2. Cu+GR	146.09	25.8	N/A
3. Cu+GR+BNNF	119.49	2.89	Reference
4. Cu+GR+BNNF+BNNT	121.94	5	2.05
5. Cu+hBNmL+BNNF+BNNT	148.12	27.55	23.96

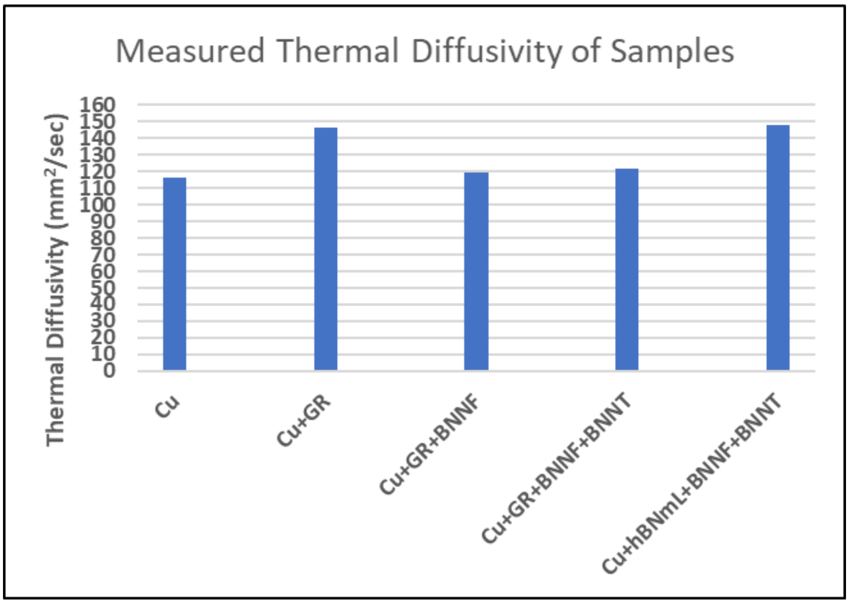

Figure 4. Bar chart comparing the Thermal diffusivity of the First set of 5 samples

Description of the samples are given below:
Cu = Bare Copper
Cu+GR = Copper with Graphene Film Top Layer
Cu+GR+BNNF = Copper with Graphene and BN Nano Flakes
Cu+GR+BNNF+BNNT = Copper with Graphene, BN Nano Flakes and BN Nano Tubes
Cu+hBNmL+BNNF+BNNT= Copper with Mono Layer h-BN, BN Nano Flakes and BN Nano Tubes

Sample1 is bare copper that is taken as reference to compare the thermal diffusivity of the other four samples against. Also, thermal properties of bare copper like thermal diffusivity, thermal conductivity, specific heat capacity and volume density are well documented in literature. The measured value116.13 mm^2/s compares well with value of 115 mm^2/s [12]. Also, using the measured Thermal diffusivity value116.13 $mm^2/s$and knowing the density 8.83(gm/$cm^{3)}$ and specific heat value of 389 (J/kg K), the thermal conductivity of copper is calculated using equation (4) to be 398.89 (W/mK) which matches well with the conductivity range 380 to 400 (W/m K) of copper reported in literature. So, this reinforces the validity of the Laser Flash method used in the measurement of the samples. The third column in Table 1, compares the relative Thermal diffusivity performance of different samples with respect to bare copper. Even though the Sample 2 with Graphene film layer on top of Copper provides good improvement, it cannot be used as it is since it is electrically conductive and needs an insulating layer like BN layer above it. However, Graphene because of its structural similarity and a very low lattice mismatch can be used as underlying layer for BN based films. Also, Graphene can be grown by CVD process on copper which is commonly reported in literature. Since the requirement of heat spreading in exposed electronic parts in order to reduce the device junction temperature is to have an electrically insulative but thermally highly conductive layer, samples 3 to 5 are of interest. From Column 3 of the Table. 1, it is seen that sample 5 with Monolayer h-BN over copper as the substrate with hybrid BN based film gives the best performance with an improvement of 27.55% over the bare copper substrate. Column 4 of Table 1, compares BNNF only based coating against (BNNF+BNNT) and (hBNmL+BNNF+BNNT). Here again the sample 5 provides an improvement of 23.96% over sample 3 and sample 4. The reason for Sample 5 to exhibit higher thermal diffusivity could be due to the fact that it has only BN layers and no Graphene underlying layer like other samples and it is known that phonon scattering at interface of dissimilar materials always tends to lower the thermal transport phenomenon and reduce the thermal conductivity[15]. The wt% of BNNT used in the H-BN solution in the above samples is 1%.

3.2 Thermal diffusivity of Second Set of Five samples to study the impact of wt% of BNNT

Next, to study the influence of wt% of BNNT in the H-BN solution used to form the insulative film, a Second set of five samples with BNNT wt% of 0.0% (Bare copper with no coating) ,0.30%, 1.0%, 1.5% and 2.0% of BNNT were prepared with copper as the substrate. Here again, Bare copper substrate Sample 1 was used as reference. Thermal diffusivity of the samples was measured using the same Laser Flash equipment Netzsch LFA 467 and the results are given in Table 2 and the corresponding values are plotted as chart in Fig. 5, showing the increase in Thermal Diffusivity with increasing wt% content of BNNT. Even though increasing wt% of the BNNT powder increases the thermal diffusivity, the maximum wt% will be limited by the practical limitations of solution dispensability on the substrate.

Table 2. Influence of wt% of BNNT on Thermal Diffusivity. Second set of samples

BNNT (wt%) in H-BN solution	Thermal Diffusivity (mm^2/s)	% Improvement
Sample 1. 0 %	108.72	Reference
Sample 2. 0.3%	114.70	5.50%
Sample 3. 1.0%	118.84	9.30%
Sample 4. 1.5%	124.62	14.62%
Sample 5. 2.0%	126.26	16.13%

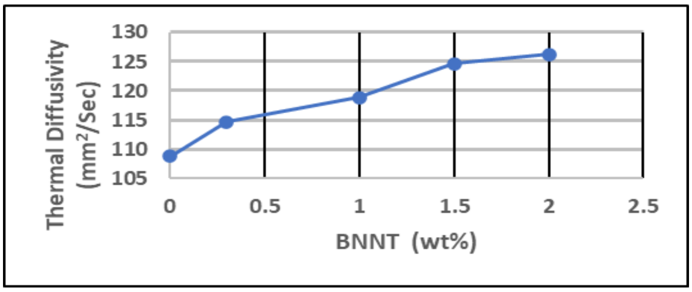

Figure 5. Increasing Thermal Diffusivity with increasing wt% of BNNT

3.3 Transient Thermal Measurements of LED on FR4 board with and without H-BN coating

Two FR4 substrate boards with LUXEON Rebel Cool White LXML-PWCI-0120 LED mounted on them were made. One of the FR4 substrate was coated on the LED mounted Top surface of the board with 1 wt% BNNT H-BN solution and dried to form a thin film coating of H-BN. The two LED mounted FR4 board samples were tested using Anatech Thermal Analyzer under a drive current of 700mA and at a power of 2 Watts. The bottom side of the FR4 boards were kept in contact with liquid cooled heat sink plate to maintain constant temperature. This is referred to as the Case Temperature and denoted by $\mathbf{T}_{C.}$ The Case Temperature was monitored constantly using thermocouple in contact with the bottom side of the board. The Transient thermal response due to the applied constant current of 700mA was recorded and the structure function using the Anatech internal software were obtained for the two samples and the plots for the FR4 board without coating and FR4 with coating (FR4 H-BN coating) are shown in Figure 6. and Figure 7. respectively. The Structure function shows the Transient thermal response of the different material layers from the LED device internally to the solder attachment to the FR4 substrate board. The X-Axis shows the Thermal Resistance and the Y-axis the Thermal capacitance. These two plots are then combined into one using the raw data to compare the performance as in Figure 8.

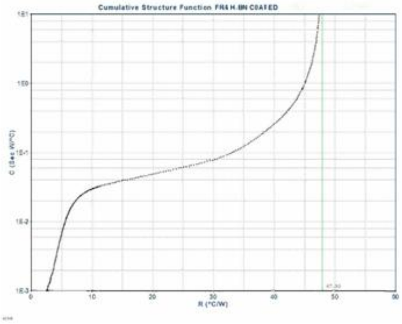

Figure 6. FR4 Board without coating

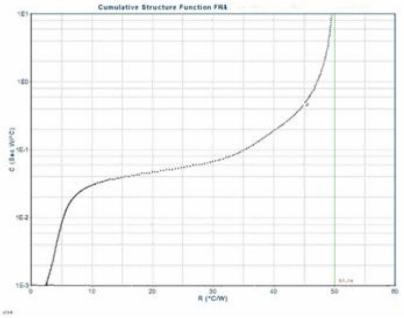

Figure 7.FR4 board with H-BN coating

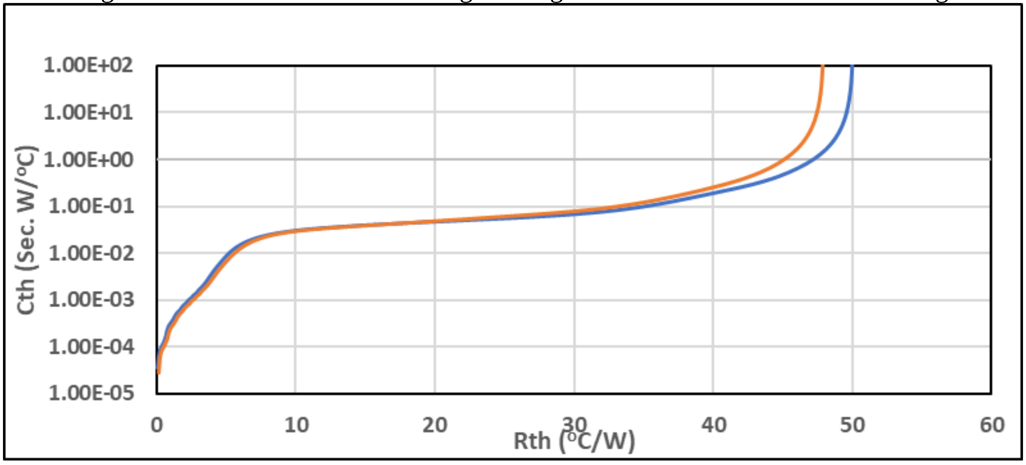

Figure 8. Combined Structure Function plots for FR4 No coating (Blue Line) and FR4 with H-BN Coating (Red Line). Rth for Blue Line FR4 is 50.04 C/W and Red Line FR4 with H-BN coating is 47.93C/W

The X-Axis Rth Thermal resistance values plotted represent the Thermal Resistance from the LED device Junction to the bottom side of the FR4 board. Referring the LED Junction temperature as $\mathbf{T_J}$ and the Bottom side of the FR4 packaging Substrate temperature as Case temperature $\mathbf{T_C}$, the LED Junction to the FR4 board bottom Case Thermal resistance referred to in the industry as Junction To Case Resistance $\mathbf{R_{JC}}$ and defined as:

$$\mathbf{R_{JC} = (T_J - T_C)/ P} \tag{5}$$

Here**,** $\mathbf{T_J}$ is the LED Junction Temperatureand $\mathbf{T_C}$ is the Case temperature of the bottom of the FR4 packaging substrate monitored by the thermocouple and **P** is the applied power to the LED using constant driving current.Thus,the plotted **Rth** is the same as $\mathbf{R_{JC}}$.

From the plotted values we can see that the H-BN coated FR4 substrate has less Thermal resistance of 47.93 C/W compared to the FR4 board without coating having 50.04 C/W resistance. When the LED is driven at 2W power this translates into 4.22 C difference in the Junction Temperature. Thus, the H-BN coating on the Top surface of the FR4 packaging substrate helps to reduce the LED Junction temperature which has impact on long term reliability and light output degradation.

3.4 Finite Element Analysis of the tested LED on the FR4 board

Finite Element Thermal Analysis of the tested LED on FR4 board was carried out using the commercial software COMSOL. A picture of the sample with LED mounted on the FR4 substrate and the geometric model are shown in Figure. 9.

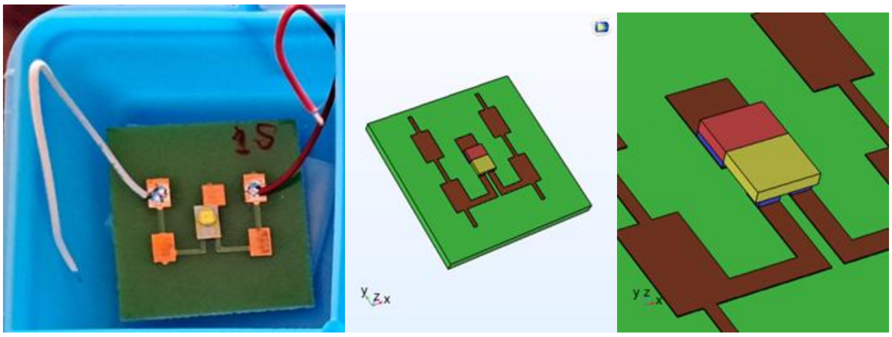

Figure 9. From Left toRight : First picture is the Actual FR4 board with the LED, The second and third pictures are Finite Element Geometric model of the Sample and the close up of LED mounted area.

A First cut Finite Element Thermal analysis was done using Thermal Conductivity of 0.90 W/m K for the FR4 material, 380 W/m K for the copper traces, 13 W/ m K for the LED device mounted area of the LED package, 27 W/m K for the rest of the LED package portion and 35 W/m K for the solder that was used to attach the LED package to the FR4 board. With the boundary condition of the bottom surface of the FR4 board kept at 20 C as mounted on the liquid cooled heat sink and total power of 2W in the LED device volume, steady state thermal analysis was carried out. The results showed a LED Junction temperature $\mathbf{T_J}$ of 124 Cand theresulting Thermal Resistance $\mathbf{R_{JC}}$ of 52 C/W using equation (5). This Finite Element model thermal resistance value of 52 C/W is very close to the measured resistance of 50.04 C/W of the FR4 board without coating to within 3.92% accuracy. So, this base model was further analyzed by only changing the FR4 Thermal conductivity to 1.0 W/m K keeping all other material properties the same and the thermal resistance based on the Maximum Junction temperature of 117.08 C was calculated to be 48.54 C/W which is close to the measured Thermal resistance of the H-BN coated FR4 board of 47.93 C/W to within 1.27%. Further refining the analysis with FR4 thermal conductivity of 0.955 W/m K gave Thermal resistance of 50.04 C/W with Maximum Junction temperature of 120.07C matching the FR4 without coating measured data. In the same way, an analysis with FR4 thermal conductivity value 1.02 W/mK resulted in a Thermal resistance of 47.91 C/W matching the measured value of 47.93 C/W for the FR4 with H-BN coating with a Maximum Junction temperature of 115.82 C. So, the ratio of the FR4 thermal conductivities that match the measured Thermal resistance values show an improvement of 6.8% in Thermal conductivity of the H-BN coating on FR4 board. The results of the COMSOL Finite Element Analysis showing Temperature distribution for various FR4 thermal conductivity values are shown in Figure 10.

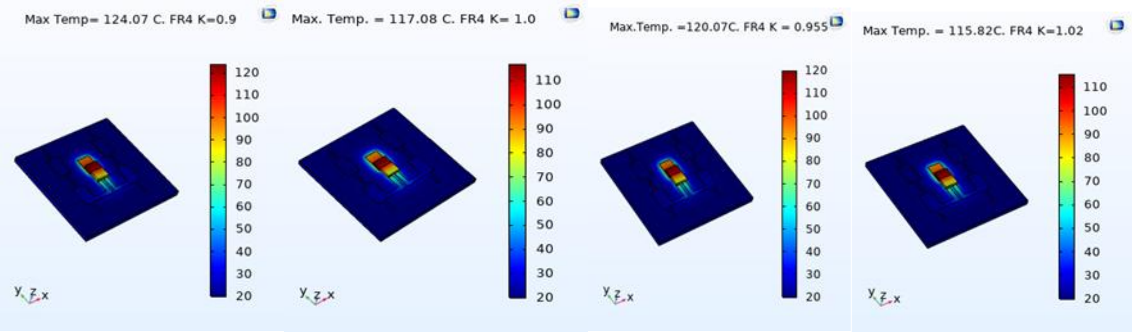

Figure 10. Temperature plots of the 4 different models with different Thermal conductivities of FR4 board

4. Conclusion

In summary, a Hybrid approach to mixing BNNT powder in BNNF solution with varying wt% of BNNT referred to as H-BN solution in this study and using it on bare copper, Graphene film on copper, mono layer h-BN on copper as underlying Packaging substrates to form electrically insulating but highly thermally conductive layer helps to reduce the Temperature ofLEDs by spreading the heat quickly and effectively. In the first study, thermal diffusivity which is a measure of how fast the material can transfer the heat and reduce hot spot temperature was studied with different combinations of underlying substrate and the BN coating. The results show that a copper substrate with a Mono layer h-BN with film formed using the H-BN solution with 1wt% BNNT gives 27.55% higher performance compared to bare copper and 23.96% improvement over other coated samples. A second set of samples focusing on influence of wt% of BNNT in the H-BN solution showed Thermal diffusivity increasing with wt% of BNNT.

Increasing wt% of BNNT from 0.3% to 2% improved Thermal diffusivity by 10.08% according to the measured data. Also, based on Transient thermal testing, H-BN coating with 1wt% BNNT on the top surface of FR4 package substrate helped reduce thermal resistance by 4.22% as compared against FR4 board without H-BN coating. So, while CVD processed hBN may be useful at device level as underlying insulative layer for Graphene based devices which are usually very small, for large area applications like Packaging substrates for LEDs, a sprayable, dispensable form of solution like our H-BN solution will be useful in improving thermal performance.

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