



## Studies on Thermal Characterization of Hybrid Filler Polymer Composites with Rice Husk and Aluminium Nitride

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**Abstract:** Polymers are widely used in electronic packaging due to their easy processing, lightweight, excellent insulation, and good mechanical properties. The growing demand for heat dissipation management materials was urgent. However, the continuous fabrication of thin highly thermally conductive polymer composites at a large scale remains challenging, especially requiring control of the filling content of fillers. In this, we reveal an effortless, effectual method to progress the thermal conductivity by using hybrid filler rice husk (RH) and aluminum nitride (AlN) with epoxy resin were made by hand layup technique and varied weight from 30 to 40 % with different ratios (1:1, 1:3, & 3:1 wt. %) are considered in the current research study. The thermal characteristics like thermal conductivity is determined by using Lee disc method. Co efficient of thermal expansion (CTE) and glass transition temperature (T<sub>g</sub>) are determined by using Thermal mechanical analyzer (TMA) by varying with temperature under nitrogen gas. Hybrid composite's molecular structure and peripheral morphology analyses as well as the interaction with epoxy resin are studied under the scanning electron microscope (SEM).

### 1. Introduction

Due to the demands of both commercial and domestic clientele, technology has substantially evolved over time (Lee et al., 2021). Electronic components have evolved into increasingly sophisticated gadgets (Kim et al., 2020). One of the significant advancements made in recent decades is the speedy processing of gadgets (Jian et al., 2018) like computers and smart phones. However, because of the increased heat generated throughout device operation, these developments have also led to a rise in the heat flow from electronic components (Wu et al., 2020; Blaschike et al., 2021; Sadashiva K. et al, 2023). It is widely established from numerous research and practical experience that the thermal environment they operate in directly affects the stability and lifespan of electronic components (Lee et al., 2021; Moradi et al., 2021, Sadashiva. K et al, 2023). In order to keep the working temperatures of devices at an acceptable level, it is crucial that the heat that has accumulated inside the device is dispersed as quickly as feasible. Current advances in heat dissipation include the invention of under fill materials and electronic encapsulation, which are now the most cutting-edge methods available. In order to protect the die bumps from thermo-mechanical stress created between the die and substrate or another die and to improve the link strength of electrical junctions, an ideal under fill material must not only have high thermal conductivity but also a low coefficient of thermal expansion. Failure to do so could result in equipment malfunction (Lee et al., 2017; Chang et al., 2017). Underfill

materials are typically polymer composites filled with fillers that have high thermal conductivity, such as epoxy matrix mixed with silica particles. Despite the remarkable performance of polymer composites, Underfill encapsulated materials with improved mechanical characteristics and better TC are commonly needed for future-generation chip connectors (Ren et al., 2014; Dittanet et al., 2017). Very thermally conductive fillers have frequently been added to epoxy matrices to produce composites with thermal conductivity higher than the neat matrices' typically quite low (0.1-0.3) W/(m\*K) values. (Bai et al., 2021; Salunke et al., 2021). Metals and ceramics like silver, aluminium, copper, AlN and BN are examples of typical filler materials to enhance thermal conduction (Ouyang et al., 2018; Sun et al., 2018). Yet with the quick development of materials and technology, numerous novel materials have been used, including carbon-based materials to enhance the polymer composite thermal conductivity (Soong et al., 2021). The widespread use of such novel materials in electronic packaging is nonetheless constrained by the high electrical conductivity of carbon nano-materials (Samsudin et al., 2022; He et al., 2019). The inherent characteristics of flake particles, however, have proven in several experiments that it is difficult to reach the specified thermal conductivity by improving dispersion and obtaining high boron nitride content (Moradi et al., 2019; Isarn et al., 2021; Nayak et al., 2019). Thermal conduction channels the connections between filler particles and matrices are crucial to thermal conductivity. Filler loading, filler distribution, filler inherent qualities (i.e., size, shape, and form), filler high specific surface area, filler orientation, filler-matrix interface suitability, and process parameters are just a few of the variables that influence the creation of these conductive pathways (Kimet al., 2015; Bae et al., 2016; Dang et al., 2019; Chiu et al., 2011; Lee et al., 2022; Zhang et al., 2021; K. Sadashiva et al., 2022). Creating continuous thermally conductive pathways in the polymer matrix by fusing different material characteristics and building a connected heat-conductive structure is also a good way to increase heat conduction, decrease the thermal resistivity at the matrix and filler interface, and maximize filler consumption. Using hybrid fillers in composites with various proportions has been associated with enhance heat conductivity, according to numerous research. With superior filler distribution of BN and graphene oxide at different proportions in polymer matrices, (Huang et al., 2017) achieved improved thermal conduction than neat polymer resin. HDPE was made by (Feng et al., 2022) using 3 wt% MWCN and 30 wt% BN as fillers. Compared to the binary and ternary composites, the ternary composites displayed a higher thermal conductivity 1.54 W/m\* (Ramu et al., 2022) investigated the thermal conductivity of polymer composites containing a hybrid filler of rice husk and MWCNT and discovered that thermal conductivity increases with MWCNT concentration rather than rice husk presence. Thermo-gravimetric analysis performed by (Tejsingh et al., 2019) revealed that increasing the rice husk and walnut shell particles improved the thermal stability of the hybrid composites. (Dayanidhi Jena et al., 2022) compared the thermal conductivity of hybrid polymer composites with rice husk and carbon fiber reinforcement of experimental value with mathematical model and discovered that experimental values are close to analysis value. (Balaji et al., 2022; Balaji et al., 2022; Ngo et al., 2019) investigated the thermal conductivity of hybrid filler composites with randomly distributed fillers numerically and experimentally, and discovered that numerical values are very close to experimental values, allowing the cost of composite fabrication to be significantly reduced. (Naizi et al., 2014) performed heat conduction and coefficient of thermal expansion simulations by creating RVE models in Digimat and importing them into Cosmol. The results were not closer to the experimental values.

In this study simple approach was utilized for fabrication of hybrid filler composite by simple hand layup method. Thermal characterization of prepared specimens with varying ratios and weights of rice husk (RH) and aluminium nitride (AlN), as well as the impact of filler on polymer. SEM was used to

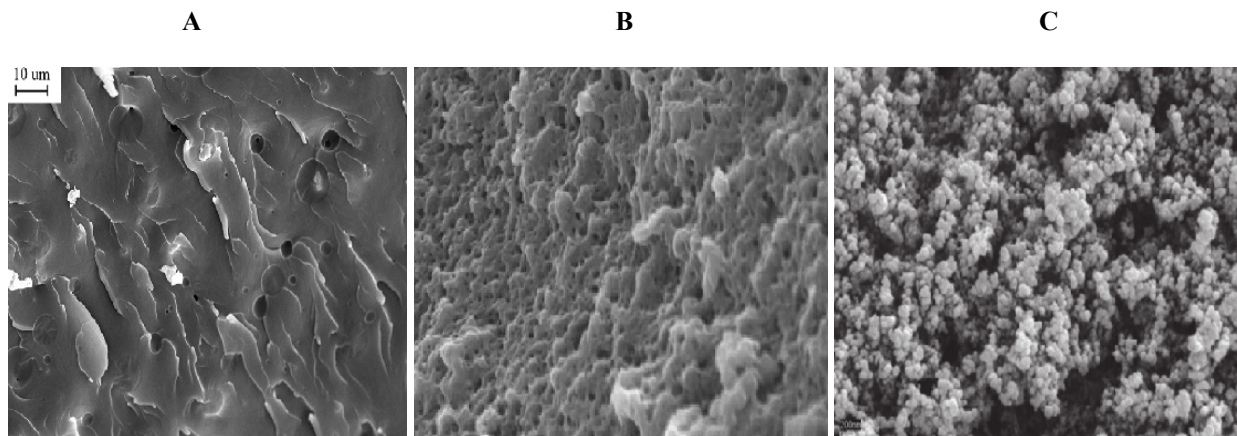
perform morphological analysis of the hybrid filler interaction with neat epoxy. Ansys software was used to simulate the effective thermal conduction, which was then compared to experimental values.

## 2. Experiments

The main components of the composite were epoxy, aluminium nitride, and rice husks. From vruksha composites in Telangana, rice husks and micro-sized aluminium nitride with a particle size of 25 microns were purchased. Epoxy (LY 556), also known as Bisphenol-A-Diglycidyl-Ether, is used as the matrix material (BADGE or DGEBA). When epoxy is combined with the hardener tri-ethylene-tetramine (TETA), an aliphatic essential amine with the trade name HY 951, it produces a solvent-free room temperature curing system. Nano Technologies Bangalore supplied the hardener HY-951 and the epoxy resin LY 556. Table 1 Details of materials.

**Table 1.** Details of materials

<i>Material description</i>	<i>Particle size in <math>\mu\text{m}</math></i>	<i>TC (W/m K)</i>	<i>Density (g/cc)</i>
<i>Epoxy</i>		0.310	1.1
<i>Rice husk</i>	100	0.03	0.5
<i>Aluminium nitride</i>	25	200	3.26



**Figure 1.** A Epoxy

B Rice husk

C Aluminium nitride

### 2.1 Preparation of epoxy with hybrid filler

The traditional hand lay-up process is used to make the epoxy-based hybrid composites. For this investigation, two different types of fillers were used; one was synthetic AlN and the other was organic RH, and both were distributed at random. fabrication of specimens with various weight proportions, such as 30 and 40 weight percent, while maintaining weight ratios of 1:1, 1:3, and 3:1 between aluminium nitride (AlN) and rice husk (RH). The positioning and naming of several composite specimens created utilising epoxy as the basis matrices are shown in Table 1 below, accordingly. Epoxy LY556 and hardener HY951 are combined in this composite matrix at a ratio of 10:1 by weight, as recommended. Sensibly increased amounts of AlN and RH were added to the Epoxy matrix as needed to ensure uniform particle dispersion and prevent large molecule aggregation. The mixtures were combined into a single batch and stirred mechanically for 30 minutes at room temperature. The mixture was then degassed after being heated to 50° C. The samples were created using a traditional casting process in a wooden mould. The mould has been treated with a releasing agent to make it simple to remove the composite material. The specimens were created using a wood mould and a traditional casting technique. The mould has been treated with a releasing agent to make

it simple to remove the composite material. The uniformly combined solution is slowly poured into the 30x30x0.3 centimeter wood mould. The castings were allowed to cure for 24 hours at room temperature. The curing took place in an oven for 1 hour at 80°C, followed by 2 hours at 145°C, and then it was cooled at room temperature to obtain the AlN/RH filled epoxy composite. The composite was then removed from the mould and cut using a water jet in accordance with ASTM requirements to conduct the thermal properties.

**Table 2** Composite composition with varying weight and ratios

<i>Slno</i>	<b>Ratio</b>	<b>Composite</b>	<b>composition</b>
1	1:1	RAC7	Ep (70 wt.% ) + RH (15 wt.% ) + AlN (15 wt.% )
2		RAC8	Ep (60 wt.% ) + RH (20 wt.% ) + AlN (20 wt.% )
3	1:3	RAC9	Ep (70 wt.% ) + RH (22.5 wt.% ) + AlN (7.5 wt.% )
4		RAC10	Ep (60 wt.% ) + RH (30 wt.% ) + AlN (10 wt.% )
5	3:1	RAC11	Ep (70 wt.% ) + RH (7.5 wt.% ) + AlN (22.5 wt.% )
6		RAC12	Ep (60 wt.% ) + RH (10 wt.% ) + AlN (30 wt.% )

## 2.2 Investigation Technique

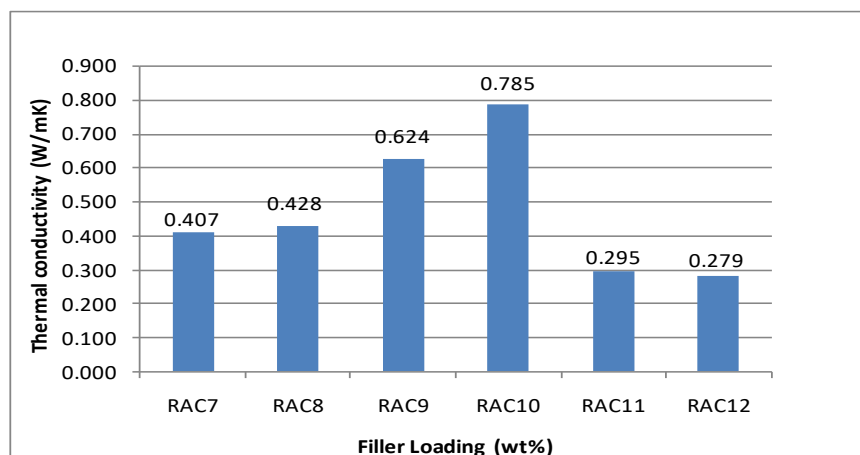
Thermal characteristics of the hybrid polymer composites specimen was determined as per ASTM standards. The dimension of the specimen should be 10 cm diameter with thickness 3 mm for testing thermal conductivity by using Lee hot disc method, the specimen was placed between top hot plate and bottom cold plate and studied the heat conduction between plates through the hybrid polymer sample. Co-efficient of thermal expansion is most important property of the material in which the material could expand or contract due to rise in the temperature. The linear co efficient of thermal expansion of polymer composite was measured by using Thermal mechanical analyzer (TMA). The glass transition temperature is an ideal property of the material and it is determined by using TMA. The samples were cut to required dimension 8 X 3X 3 mm and heated in rig at a rate of temperature 5° C/ min to 130 ° C under the atmosphere of nitrogen. Samples for scanning electron microscopy (SEM) were prepared by shattering a rectangular sample using diagonal cutting pliers and coating the sample surface with platinum. The morphology of all composites was characterized using a field emission scanning electron microscope (JEOL JSM-6480LV).

## 3. Results and Discussion

### 3.1 Thermal conductivity

Figure 2 shows that the changes in the thermal conduction of the distinct polymer composites with different weight and ratios of fillers loading. Resin (epoxy) exhibits low thermal conductivity of 0.310 W/m K. Overall, thermal conductivity of epoxy composite increases and decreases with filler concentration in all above cases. More aluminium nitride filler interacts with rice husk and epoxy establishing efficient thermal conductive path and improving thermal conductivity of composite. It was noticed that the thermal conductivity of sample RAC10 is 0.785 W/mK significantly higher than rest of the samples, due to presence of higher percentage of AlN forms the heat transfer channels. On the other hand, RAC12 exhibits the low thermal conductivity 0.279 W/mK because of higher percentage

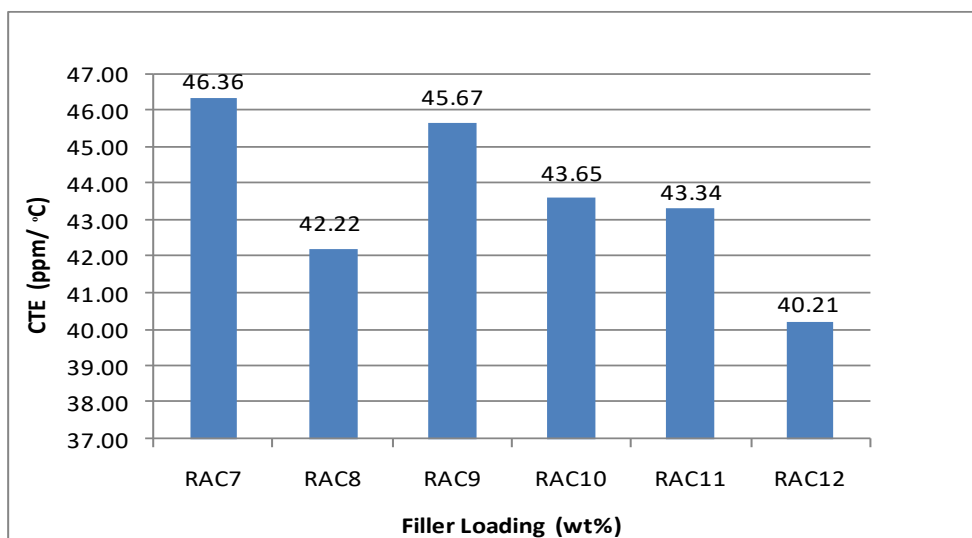
of rice husk which blocks the heat conductive path. This result was attributed to the incorporation of a small amount of AlN filler. The thermal conductance of sample RAC8 is 0.428 W/mK comes between the above two samples because the AlN and RH are present in equal proportions, heat transfer channel as well as the heat restricting path were in equal phase.



**Figure 2.** Thermal conductivity v/s Filler loading

### 3.2 Co-efficient of thermal expansion

Coefficient of thermal expansion is most important property of composite to maintain the dimensional stability. Figure 3 depicts variation of CTE with respect to the filler loading, CTE of the neat epoxy exhibit higher than the AlN and RH, these fillers help in the maintaining stability in dimensions of the composite. CTE of the composites was depend on the filler inclusive and interaction with epoxy. CTE of the RH/AlN/EP composites (RAC7) at 30 wt% showing 46.36 ppm/°C high compare to the composites (RAC8) at 40 wt% (42.22 ppm/°C).



**Figure 3.** CTE (ppm/°C) v/s Filler loading (wt %)

The higher filler particle restricts the expansion of epoxy composites due to efficient interaction between epoxy and fillers. Similarly the sample RAC10 (43.65 ppm/°C) exhibits lower CTE than the RAC9 (45.67 ppm/°C), Synergic effect of two fillers results in more efficient filler and epoxy interaction and avoiding the expansion of epoxy composites. RAC11 and RAC12 sample exhibit CTE was 43.34 and 40.21 ppm/°C, RH restrict the expansion of the composites by significant interaction with AlN and epoxy which will be useful for electronic industry.

### 3.3 Glass Transition temperature

The glass transition temperature ( $T_g$ ) is defined as on continuous heating of materials leads to change its physical state from solid to rubber, this transformation due to the mobility of molecules at particular temperature that is considered as glass transition. Figure 4 plots  $T_g$  for different samples with respect to the filler loading for varying ratios and wt%. The RAC7 showing the 110 °C as compare to the RAC8 is 112 °C, this due to the RH restricts the movement of molecules of epoxy and AlN by efficient interaction. Similarly, the RAC9 and RAC10 showing 100 °C and 97 °C, thereby the larger content of AlN does not have efficient interaction with epoxy and RH, allows the molecule mobility. The  $T_g$  of the sample RAC12 is higher than the rest of the samples as shown in the figure 4, here with higher concentration of RH have significant interaction with both AlN and epoxy, restrict the mobility of molecules. Overall increase the  $T_g$  of composites involves the significant interaction of filler / epoxy can restrict the mobility of polymer chain and interaction of composites can be enhanced by increasing the filler content and distribution.

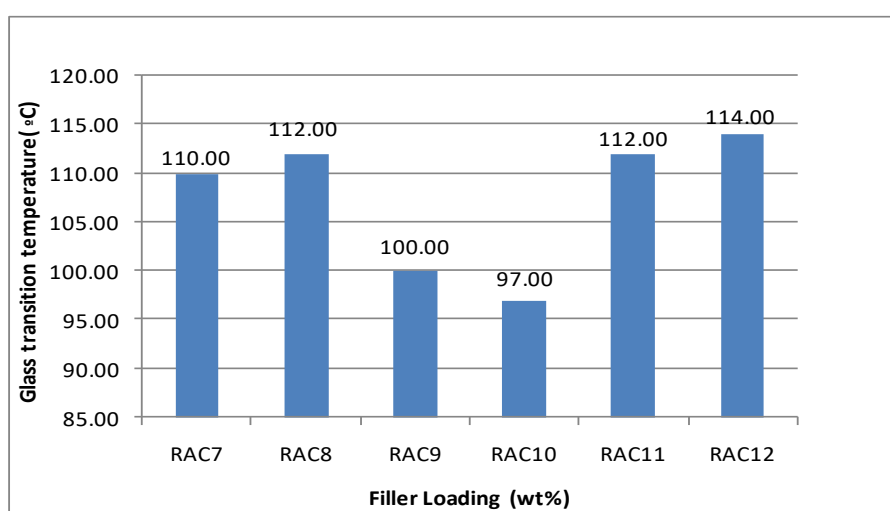
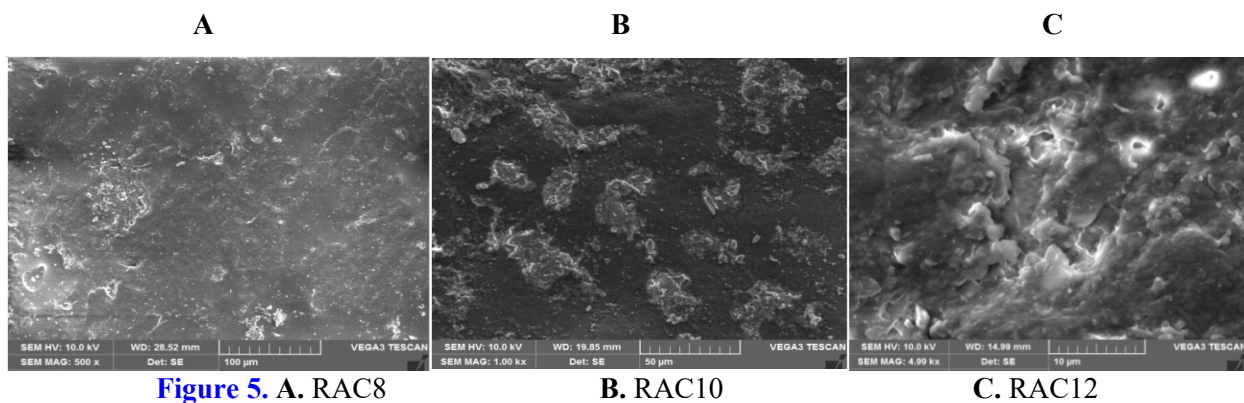


Figure 4. Glass transition temp<sub>t</sub> (°C) v/s Filler loading (wt%)

### 3.4 Morphological study of polymer composite

Figure displays cross-sectional scanned pictures of RH/AlN/Ep with different weight and ratios. There is no agglomeration, as shown in Figure 5 A, despite the fact that the RH and AlN filler is very evenly dispersed throughout the polymer matrix. Due to the fact that these hybrid fillers have the least contact area, they do not generate a conductive channel even when AlN particles are loaded at a 20 wt % concentration. Minute holes are identified due to combustion reaction between hybrid fillers and polymer resin. Furthermore, the AlN particles in Figure 5 B may be seen to be evenly spread and to create thermally conductive channels, but there is also some agglomeration, which can be attributed to one of cause. The larger surface area and inherent shape of AlN particles allow for mutual attachment of the particles. The AlN particles have a larger specific surface area than the RH particles due to their smaller size, which intensifies the intermolecular interactions that encourage agglomeration. However, as seen in Figure 5 C, the RH/AlN/EP composite has a continuous connection with negligible agglomeration, indicating that the RH/AlN hybrid filler creates a more powerful interfacial coupling. Compared to RH particles, AlN particles have a lower particle size. As a result, during mixing, shear motion allows the AlN particles to fill the spaces between the RH particles. As a result, aggregation is reduced and a non-heat conduction path is created. In the RH/AlN/EP composites, no additional functional heat routes were thereby produced.



**Figure 5. A. RAC8**

**B. RAC10**

**C. RAC12**

## Conclusion

To establish the influence of RH/AlN particle on thermal characteristics of polymer composites through experimental study has been conceded. The specimens are fabricated by varying the concentration of fillers with respect wt% and ratios. The RH/AlN polymer composite that was produced had a thermal conductivity of 0.785 W/mK at a filler loading ratio of 1:3, which is nearly twice as high as neat epoxy. The conduction path that was created is what increased the polymer composites' exceptional capacity for thermal management and thermal conductivity. Because of the filler interaction and restriction of molecule mobility to generate space, these composites exhibit low coefficient of thermal expansion (40.21 ppm/°C) and thermal stability compared to the other composites, which is suitable as encapsulation for electronic applications. Glass transition temperature of polymer composites is 114 °C which is higher than neat epoxy, where RH upholds the AlN and epoxy by restriction of mobility of atoms so that the maintain the stability of the composites.

**Disclosure statement:** *Conflict of Interest:* The authors declare that there are no conflicts of interest.

*Compliance with Ethical Standards:* This article does not contain any studies involving human or animal subjects.

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