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Investigation on Thermal Conductivity of CNF-Reinforced PLA Composites Fabricated Using FDM Technology

N. Hemanth Kumar[1,2*] and H. Adarsha[2]

[1]Department of Mechanical Engineering, JAIN (Deemed-to-be-University), Bangalore - 560079, baghavathhem@gmail.com

[2]Department of Mechanical Engineering, FET, JAIN (Deemed-to-be-University), Bangalore - 560069, Karnataka, India

Abstract

Additive Manufacturing (AM) methods, especially Fused Deposition Modelling (FDM), have been highly promising for fabricating polymer composites with improved thermal properties. The present study examines the influence of Carbon Nanofiber (CNF) reinforcement on the thermal conductivity of Polylactic Acid (PLA) composites with CNF loading from 1% to 3% by weight. Experimental findings reveal a notable improvement in thermal conductivity, with pure PLA showing a baseline value of 0.185 W/mK. Addition of 1% CNF enhanced thermal conductivity by 27% to 0.235 W/mK, whereas addition of 2% CNF resulted in an improvement of 32.4% to 0.245 W/mK. 3% CNF achieved maximum improvement at 54% to 0.285 W/mK. These enhancements can be related to the high thermal conductivity of CNF and to its function to decrease voids and create conductive paths inside the PLA matrix. The research points out the promising prospects of CNF-strengthened PLA composites for thermal management in electronics, aerospace, and automotive sectors.

Major Findings: *The study demonstrated that the addition of Carbon Nanofibers (CNFs) to Polylactic Acid (PLA) composites greatly enhanced thermal conductivity, with a maximum increase of 54% at 3% CNF loading to 0.285 W/mK, indicating the material's potential for heat transfer.*

Keywords: *Carbon Nanofiber (CNF), Fused Deposition Modelling (FDM), Polylactic Acid (PLA), Polymer Composites, Thermal Conductivity*

1.0 Introduction

AM is a revolutionary change from conventional subtractive manufacturing methods, allowing for accurate, layer-by-layer fabrication according to 3D CAD models. The last decade has witnessed immense research driving the growth of many AM methods, which have been utilised effectively in a wide range of industries such as digital art, digital architecture, aerospace, biomedical, and automotive engineering[1]. The quick growth of AM can be thanks to its built-in versatility, affordability, and unmatched speed in rapid prototyping and low-production levels. One of the biggest breakthroughs in AM is the creation of composite materials with added high-performance fillers like graphene, nanoparticles, carbon nanotubes, and fibres to gain enhanced mechanical, thermal, and electrical properties. Polymer reinforcement with fibres has significantly improved mechanical strength, durability, and thermal stability to make them capable of being used in a wide variety of

**Author for correspondence*

high-end applications[2-4]. Among such reinforcements, CNFs are a game-changer because they possess high mechanical rigidity, high specific strength, better thermal conductivity and excellent electrical properties. These outstanding properties have attracted significant attention, especially in aerospace, automotive, and high-performance electronics applications, where structural integrity and heat dissipation are of utmost importance. CNF-reinforced polymer composites have substantially enhanced thermal conductivity, providing better heat transfer, which is crucial for high-performance engineering applications[5-6].

Recent studies have been focused on utilizing various fillers to improve the thermal characteristics of 3D-printed polymer composites. Adarsha *et al.*,[7] investigated the improvement of thermal and electrical conductivity in ABS-based 3D-printed models through the addition of graphene through mechanical and chemical mixing. The work proved substantial improvement in conductivity, which is vital for electronics and functional engineering models. Su *et al.*,[8] researched the influence of Hydroxylated Boron Nitride (OH-BN) fillers in Thermoplastic Polyurethane (TPU) composites and showed improved isotropic and anisotropic thermal conductivity through chemical surface modification. Wang *et al.*,[9] investigated Graphene Nanoplatelet (GNP)-reinforced Isotactic Polypropylene (iPP) composites, wherein strategic filler orientation through fused deposition modelling improved substantially through-plane thermal conductivity, which renders them highly promising for energy and electronics applications. Agarwal *et al.*,[10] highlighted the significance of filler type, volume fraction, and alignment in polycarbonate-based nanocomposites. Their work validated that alignment of the fibre in the direction of heat transfer maximizes thermal conductivity, again emphasizing the importance of filler distribution and orientation.

Of the several AM methods, FDM is especially adapted to the exact addition of nanofibers into polymer matrices, allowing for complex geometries and designs. This technology, characterized by sustainability and cost-effectiveness, has found extensive application in biomedical, aerospace, and automotive industries for manufacturing lightweight, high-performance parts. Nevertheless, fused deposition modelling's reliance on thermoplastic filaments is limited to materials, and thus only a narrow set of polymers is acceptable for high-thermal-performance applications[11-13]. As much as AM research has been carried out, there remains a vast lack of literature concerning the thermal conductivity behaviour of 3D-printed polymer nanocomposites, i.e., PLA composites filled with CNFs. Closing this knowledge gap, the current study examines how different CNF weight percentages (0% to 3%) affect the thermal conductivity of PLA composites. The research findings are of far-reaching significance to industries demanding effective thermal control, such as next-generation electronics cooling systems, aerospace thermal protection, and heat exchanger applications.

This study specifically contributes to the development of AM-based nanocomposites by reporting the first systematic thermal conductivity assessment of 3D-printed CNF-reinforced PLA composites, bridging an essential knowledge gap. It illustrates a remarkable thermal conductivity improvement of as much as 54% using just 3% CNF addition, exceeding enhancements in earlier studies. The work sets fused deposition modelling as a promising fabrication pathway to uniformly disperse CNFs within PLA, enhance interfacial adhesion, and reduce phonon scattering to achieve better heat transfer. It also provides a new reference for polymer-based thermal management materials that can be translated directly to aerospace, automotive, and future-generation electronics demanding cutting-edge heat dissipation solutions. Not only does this research broaden the foundational knowledge of CNF-PLA thermal properties, but it also lays a solid foundation for future developments in AM-driven composite engineering.

2.0 Experimental Part

2.1 Materials and Composite Development

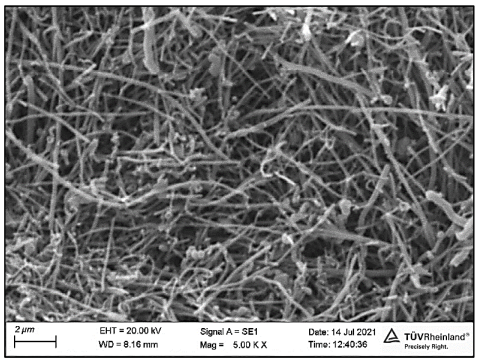
PLA pellets were procured from M/s Tespo International Pvt. Ltd. in Bangalore, India, and were used in the matrix. One of the essential steps in processing was a drying period (3 h, 140°C) of the pellets to eliminate moisture and guarantee that there were no principal challenges during each further process. For this assessment, carbon nanofibers had been added to the dried pellets in fixed percentages of one, two, and three weight per cent. Beyond three weight per cent, achieving high-quality filaments with uniform dispersion was challenging due to poor extrusion quality. The filament manufacturing and

subsequent printing of fused deposition modelling parts were unsuccessful due to the poor ductility of the material and the agglomeration of carbon nanofibers. Filaments were subsequently made from these reinforced pellets. Figure 1 shows the carbon nanofibers Scanning Electron Microscopy (SEM) picture together with the Energy-Dispersive X-ray Spectroscopy (EDAX) technique utilised in this study. Following this, the filaments underwent the process of pelletisation and were exposed to a 100°C drying temperature, after which they were introduced into an extrusion machine. The twin-screw extrusion technique employed in the fabrication of specimens uses short filament lengths. A single-screw double-rod extruder and a compounding machine of laboratory grade were used to extrude filaments with a diameter of 1.75mm.

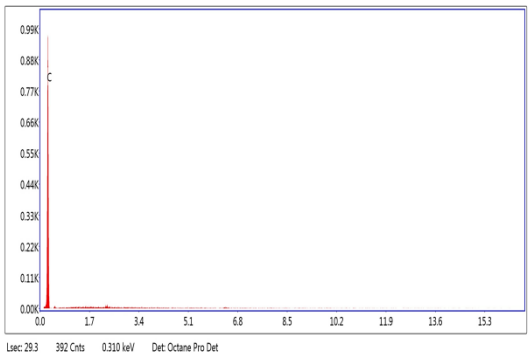
One popular additive manufacturing method is FDM, which builds three-dimensional structures by sequentially depositing layers. For thermal conductivity specimens, a Global 3D Labs Pramaan printer with an enclosed chamber of a space capacity as high as 400mm^3 was used to manufacture them. To print the models, we used optimal parameters, which include the shell thickness of 0.4mm and an infill density of 100 per cent, while the layer/thicknesses of the top and bottom were set at 1.2mm in total height. The print was oriented at 45°, printed at 5 mm/sec. The temperature of the nozzle had been configured to 240°C, and at 80°C, the bed was configured. Component assembly along the Y-axis took place on the print bed after the filament was ejected from the nozzle[14-16].

2.2 Thermal Conductivity Test

To quantify the PLA polymer composites that are 3D printed and reinforced with carbon nanofibers in terms of thermal conductivity, the following procedure was adopted, adhering to the American Society for Testing and Materials (ASTM) E-1530 standard. The test specimens measured 25mm in diameter and had a thickness of 3mm. Specimens have been made from 3D-printed PLA polymer composites strengthened with CNF. The specimens' ability to conduct heat was assessed using the Unitherm™ 2022 Thermal Conductivity Measuring System at the Central Institute of Plastics Engineering and Technology, Chennai, India. In the test procedure, the prepared specimens were placed between two plates within the measurement system. One plate was heated to a temperature of 100°C, while the other plate was cooled simultaneously to create a temperature gradient across the specimen. The system was allowed to reach a steady state, where the temperature difference across the specimen remained constant. At steady state, the following data were recorded: the temperature difference between the two plates, the specimen thickness, and the heat input into the system. The composite specimens' thermal conductivity (k) was determined using the Fourier equation for heat conduction in a linear form. To ensure the accuracy and repeatability of the measurements, multiple specimens have been examined, and the stated mean thermal conductivity values. Any deviations in the measurements were analysed and corrected as per standard practices. This experimental setup and procedure ensured reliable and reproducible measurement of thermal conductivity



(a) SEM of CNF nano fibres



(b) EDS of CNF nano fibres

Figure 1. (a-b). Scanning electron microscopy (SEM) images and energy dispersive spectroscopy (EDS) pattern of the as-received carbon nanofiber (CNF) powder.

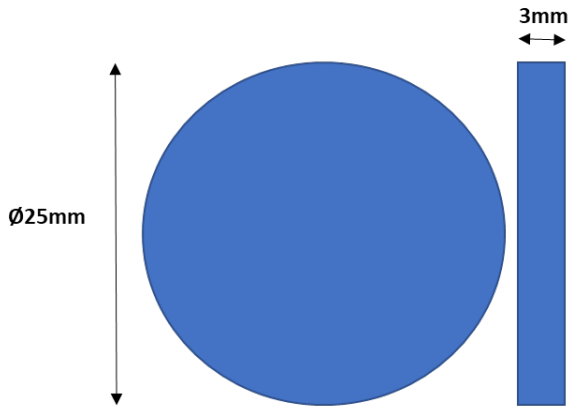


Figure 2. Schematic representation of the thermal conductivity test specimens, detailing their precise dimensions.



Figure 2(b). High-resolution photograph of the 3D-printed composite specimens fabricated using fused deposition modelling (FDM).

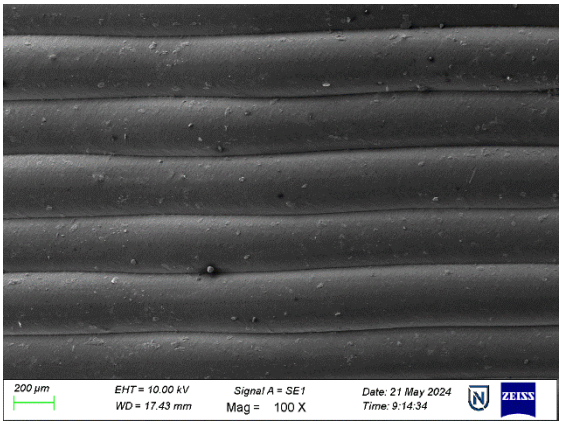
for CNF-reinforced polymer composites. Figure 2a shows the dimensions of the thermal conductivity test specimens. Figure 2b presents the photograph of the 3D-printed test specimens.

3.0 Results and Discussion

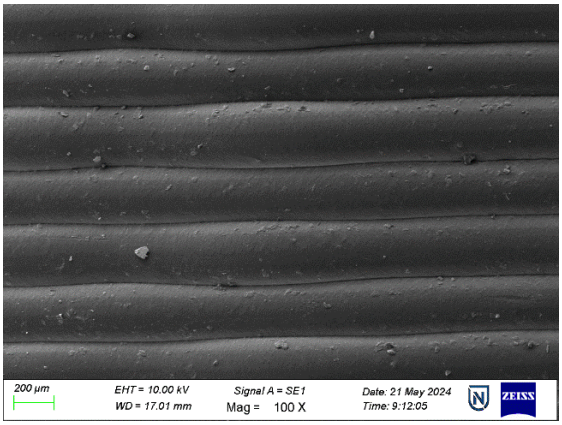
3.1 Scanning Electron Microscopy

The SEM micrograph in Figure 3 displays the unreinforced PLA, revealing a high-quality fusion between the rasters. Bonds involving the two rasters are created as a result of interactions between the deposited raster molecules and the raster molecules being deposited. Another sign of robust adhesion between the succeeding rasters is the absence of voids or microcracks. A small amount of partially filled molten PLA was found at a few spots, but not enough to be significant. The extrusion temperature selected in the present scenario is adequate to cause full PLA liquefaction, leading to homogeneous raster deposition and little void presence. The overall assessment indicates that the adhesion between each layer of the raster is satisfactory. However, the composite of PLA containing two and three per cent CNFs SEM pictures, displayed in Figures 3a and 3b, reveal a strong fusion between rasters. The printing of raster patterns was consistent and free from any irregularities, showing that the composite filament had completely melted and extruded at the optimal temperature. Strong adhesion between old and fresh rasters implies that prior to cooling below the temperature at which glass transitions, fusion took place between them.

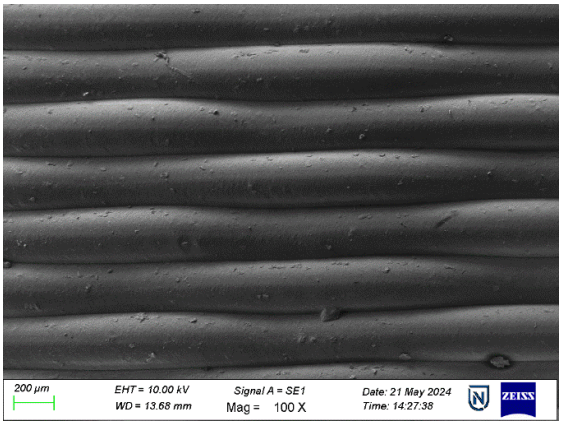
Three phases usually characterise the adhesion between rasters: molecular diffusion, contact of surfaces, and neck initiation and development. Wetness of surface, rearrangement of surface, randomisation, surface approach, and diffusion are the five steps that are commonly used to characterise the diffusion bonding process, also known as chain motion. Nonetheless, the bond generation mechanism is examined in three phases earlier. Both the previously deposited raster and the newly placed raster make contact with the surface when they are positioned next to each other. The merger of the two rasters due to viscous sintering and diffusion is known as the neck formation, which occurs next. Before continuing, a quick overview of viscous sintering will be given to enhance comprehension. Surface tension facilitates a process known as viscous sintering, which is the coalescence of particles adhering to one another. Viscous sintering, which was previously found solely in polymers, has also been observed in metals and ceramics. It has emerged as the process for bond creation in additively manufactured materials that has been studied the most. As mentioned earlier, the neck creation at surfaces in contact is the first step in the process. The capillary forces decrease the surface area, whereas the PLA flow's driving force is determined by surface tension. Viscous sintering occurs by minimising the free energy of the particles, and it includes minimising the surface area and releasing the energy that has been held in check by viscous motion. Surface contact of two raster materials leads to wetting, which promotes intermolecular diffusion[17,18]. The neck development and subsequent fusion between cells occur



(a) PLA



(b) PLA+0.5CNF



(C) PLA+0.75CNF

Figure 3. (a-c). Scanning Electron Microscopy (SEM) images of 3D-printed pure polylactic acid (PLA) and PLA reinforced with carbon nanofibers (PLA+CNF) nanocomposites.

primarily through two major mechanisms. From a reinforcing perspective, effective bonding between the interlayer and raster is facilitated in large part by the CNF.

The CNF is widely recognised for its high thermal conductivity, ranging from around 449 W/mK to 2000 W/mK when taking into account thermal contact resistance[19]. According to the source, CNF has a thermal conductivity of about 2000 W/mK. Whenever a fresh raster is being placed, the CNF's ability to conduct heat makes it easier for the earlier deposited raster to re-soften[20]. This encourages improved raster adherence and also makes it easier for rasters to be filled in efficiently, which minimises voids. Furthermore, the strong adhesion and notable alignment of the fibres show that adding carbon nanofibers (CNFs) did not cause the composite's melt viscosity to increase. The CNF's weight percentage falls between one per cent and three per cent, indicating a relatively low CNF concentration in PLA. The mobility of PLA molecules may decrease at higher concentrations[21]. The addition of a small amount of CNF greatly increased PLA molecule mobility, which led to the creation of robust connections between the raster's constituent parts.

3.2 Thermal Conductivity

Thermal properties, like the ability of heat-conductive polymer composites, are of great importance for many thermal management applications. The reinforcing phase and type of processing route employed for the fabrication of composites serve a crucial part in dictating the thermal qualities. The type of reinforcing phase, or in this case, CNF, its content, morphology, dispersion, and bonding with the PLA matrix are important factors that have a say in the thermal conductivity. The processing routes also influence the properties, and in the case of the fused deposition modelling technique, the macro- or microstructure of composites is entirely different from the microstructure fabricated using conventional techniques. The influence of varying CNF content on the thermal conductivity of PLA composites was investigated, and Figure 4 shows the results. The pure PLA has a value of 0.185 W/mK, as can be seen in the figure. This value recorded is almost similar (or marginally on the lower side) to that reported in the literature. Between 0.16 and 0.287 W/mK is the range of PLA's thermal conductivity value, and one should understand that the difference is mainly due to different processing routes adopted to

fabricate PLA[22-24]. Take, for instance, Blanco *et al.*[22], who reported a thermal conductivity of 0.16 W/mK for neat PLA when developed using fused filament fabrication technology. Despite some porosity at higher filler levels, better fibre distribution showed significant conductivity gains, enhancing thermal transport networks for improved heat dissipation. In another case, Lule and Kim[23] reported the development of PLA by the solution cast process and measured its thermal conductivity. The 0.287 W/mK had been obtained for neat PLA when measured using a laser flash analyser. Literature demonstrates that thermal conductivity tends to vary with reference to the processing route. In the present case, the value obtained is between the values reported in the literature for neat PLA, which is quite good.

When 1% CNF was added to PLA, the conductivity was increased to 0.235 W/mK. The enhancement in the conductivity when compared to neat PLA is about 27%, which is quite good. However, thermal conductivity studies on FDM- FDM-fabricated PLA-reinforced CNF composites are rare to find. Although reference[22] did report the thermal conductivity of FFF-printed PLA/CNF composites, the measurements conducted were at 70°C and 90°C. A direct correlation between this reference and the present work might be incorrect. While exploring the values, the neat PLA showed values for both temperatures that were almost the same (0.16 W/mK), but when 1% of carbon fibre was added to PLA, the value increased to 0.18 W/mK, which was the same for both temperatures. Despite the addition of a high amount of carbon fibres, the increase was quite minimal. In the current study, a minor increase of CNF (1%) yielded an enormous rise in the conductivity of heat. The increment in the thermal conductivity of the PLA matrix post-addition of CNF can be attributed to the high thermal conductivity of the latter phase. The precise CNF heat conductivity is unknown, and many studies assume its value is somewhere in between 600 W/mK and 3000 W/mK. The bracketing of thermal conductivity is based on the fact that CNF belongs to the carbon family. Since it has a layered structure similar to graphite and a size that of carbon nanotubes, the thermal conductivities of these materials are used as lower and upper limits. However, some isolated studies have shown a thermal conductivity value of 1950 W/mK for heat-treated CNF[25,26]. Nonetheless, this value of CNF is several times higher than that of neat PLA. By this, one can expect that the thermal conductivity of the combination of these two materials will at least be better than neat PLA. The principal mechanisms responsible for PLA composites' increased thermal conductivity are thermally conductive linkages and filling of voids in the microstructure by CNF. Moreover, the thermal conductivity value grew to 0.245 W/mK as the CNF concentration was elevated to 2%. This value is almost 32.4% higher than that of neat PLA, while compared to the PLA/1% CNF composite, the increment is quite marginal. The composite with the highest CNF content of 3% showed the highest value of 0.285 W/mK, and the increase in heat conductivity over plain PLA was 54%. This pattern suggests that PLA composites' thermal conductivity increased as the CNF level increased. The enhancement of thermal conductivity suggests that as the CNF content is increased from 1% to 3%, the empty spaces, or more specifically, the CNF-free zones, are reduced. The filling of spaces due to the availability of more CNFs and the reduction in distance between CNFs led to an increase in the number of conductive tracks. Thus, composites with higher CNF content possessed a higher number of conductive tracks, which eventually led to better phonon transmission and an increase in thermal conductivity. This observation is well in line with the observation reported by Ali *et al.*,[27] on carbon fibre-reinforced epoxy composites. The presence of a greater number of conductive links with an increase in carbon fibre content and a reduction in empty spaces led to an enhancement in the thermal conductivity. In the second scenario, the opted processing technique, fused deposition modelling, has also proven to be effective as it dictates dispersion and bonding between the PLA matrix and CNF. The process dispersed CNF uniformly throughout the PLA matrix with good interfacial bonding. CNF and PLA matrix have a significant interaction that reduces the phonon scattering at the interface, which is one more reason for improved thermal conductivity for PLA composites. A similar observation was reported by Karippal *et al.*,[28] on epoxy/CNF composites. The authors found strong interaction between functionalised CNF and epoxy, minimising phonon scattering and giving rise to enhancement in the thermal conductivity for composites. Overall, in the present work, the addition of the CNF phase and the fused deposition modelling processing technique used to develop PLA composites has resulted in an enhancement in the thermal conductivity, making them potential candidate materials for thermal management applications.

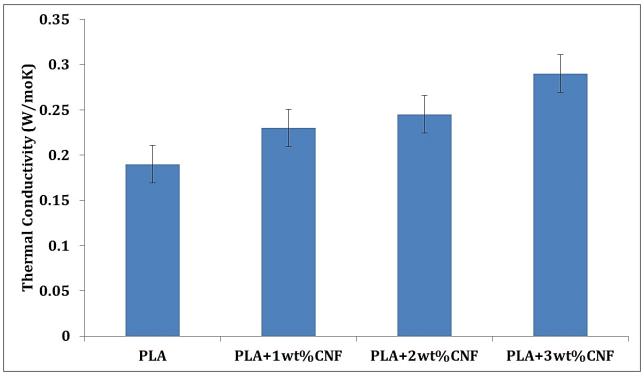


Figure 4. Graphical representation of the variation in thermal conductivity of fused deposition modelling (FDM) printed PLA+CNF polymer composites.

4.0 Conclusions

The investigation's findings led to the following outputs:

1. The thermal conductivity of neat PLA was measured at 0.185 W/mK, aligning with values reported in the literature for different processing techniques.
2. The addition of 1% CNF increased the thermal conductivity by 27% to 0.235 W/mK, indicating the beneficial effect of CNF reinforcement.
3. 2% CNF loading further improved the conductivity by 32.4%, to 0.245 W/mK, although that from 1% CNF was minimal.
4. The maximum enhancement was seen for 3% CNF, resulting in a 54% increase over neat PLA, reaching a final value of 0.285 W/mK.
5. The improvement in thermal conductivity is due to enhanced CNF dispersion, high interfacial bonding, and the creation of thermally conductive pathways.
6. These results indicate that CNF-reinforced PLA composites are viable for applications in advanced thermal management, such as electronics cooling, aerospace parts, and heat exchangers, where efficient heat dissipation is essential.

5.0 Declaration

5.1 Competing Interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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