A review on 3D printed polymer-based composite for thermal applications

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Abstract. Recently, 3D printing techniques for the polymer-based part has become popular in the industry and academic area. Fused deposition modelling (FDM) is one of the most popular methods among 3D printing techniques because of its low cost, ability to fabricate objects with complex structures and geometries, specific functions, ease in processing, safety, reliability, and availability of various thermoplastic materials. This technique's application is widespread on the electronic application, medical application, and rapid tooling applications. This paper aims to review fused deposition modelling (FDM) 3D printed polymer-based composite techniques for thermal management applications. This paper will provide an overview of fused deposition modelling (FDM) techniques in 3D printing, conducting polymer base and conducting nanofiller additive material used, and current trend research in this area.

Keywords: Fused Deposition Modelling; Thermal Management; Conducting Polymer; Conducting Nanofiller Additives.

1. Introduction
Higher-dimensional accuracy and shorter time to construct prototypes have become the reason to use 3D printed techniques in modern industries. There are several types of 3D printed techniques that have been commonly used in modern industry such as stereolithography (SLA), selective laser sintering (SLS), fused deposition modelling (FDM), laminated object manufacturing (LOM), and direct metal laser sintering (DMLS). The most commonly used 3D printed technique in the industry is fused deposition modelling because of its ability to construct a complex structure in a shorter time and shorter cost. It has been widely used in applications such as medical [1, 2], electrical and electronics [3-7], rapid tooling [8], automotive sector [9], toys [10] and aircraft industries. Traditional FDM systems only used to fabricate parts with thermoplastics. Current FDM systems can produce parts in a wide range of engineering and industrial thermoplastics such as acrylonitrile butadiene styrene (ABS), polylactic acid (PLA), etc.
1.1. FDM basic principles
The FDM system's basic principle is by depositing extruded materials layer by layer through a nozzle of feedstock filament offers the great potential of filament extrusion for a wide range of other materials such as carbon, metals and ceramics as shown in figure 1. FDM system comprises filament winder, drive wheels, filament feedstock, extrusion heads, nozzle, and building platform to depositing layer by layer extruded materials of 3D printing. The 3D printing filament feedstock can be either a single polymer, polymer blends, or composite materials, combining the base polymer and filler materials depending on the preferred application. Metals, ceramics, and carbon are an example of filler materials. The new composites materials from a combination of base polymers and filler materials in feedstock filament can be developed and used in the FDM process with suitable size and properties. A. Dorigato et al. [11] has made up filament for FDM feedstock from combinations of acrylonitrile-butadiene-styrene (ABS), and multi-walled carbon nanotubes (MWCNT). Liu W. et al. [12] has made a hybrid composite by combining two filler materials, silicon carbide (SiC) / C / PLA as feedstock filament of FDM and produced 3D printed polymer composite parts. FDM technique has been used in improving mechanical properties [13-20], improving thermal properties [11, 20-23], improving electrical conductivity [11], improve process ability [21, 24-26], and fabricate complex parts [27-29].

![Figure 1. Schematic diagram of FDM primary mechanism.](image)

1.2. Overview of 3D printed polymer composites
The classification of 3D printed polymer composite can be divided into a single polymer, poly-polymer, and composite. Single polymer 3D printed materials are the materials that using single polymer for 3D printed application. In contrast, poly-polymer 3D printed materials are the materials that blend two or more polymers together in their research. In composite classification includes single polymer composite, polymer blends, hybrid filler, and base-filler hybrid. Polymer blends composite can be achieved by blending two or more polymers, together with the addition of filler material. A brief schematic of 3D printed polymer composite classification using the FDM technique for thermal application is illustrated in figure 2. In contrast, figure 3 elaborated on the polymer-based composite research trend in thermal application acquired from 2001 to 2020.
The purpose of blends polymer together is improved the processability of the composite. Blanco I. et al. [21] have blended two polymers together, polyetherimide and polycarbonate to be used in advanced applications. Single polymer composite uses one type of polymer and one type of filler only while hybrid filler composite is acquired by combining two or more fillers with a polymer. Hybrid composite purposes are to enhance the properties while maintaining the others to the composite materials. To enhance the crystallinity, thermal, electrical, and mechanical properties of composite, Kotsilkova R. et al. [20] have made filament feedstock using graphene and carbon nanotubes. A Base-filler hybrid composite is a composite that combining two or more polymers with two or more fillers to become a hybrid composite. Researchers choose composites over single polymers or poly-polymer because composite materials give different characteristics to the materials depending on the polymer and filler used [4, 11, 13, 21, 30].
The graph from figure 3 shows the increasing trend of polymer-based composite research for thermal application. The trendline shown is covered from 2001 to 2020, including the data collected from more than 150 research papers, which means that this research topic is an interesting topic to be explored and has a wide range of applications. There has been increasing in the research field of 3D printed polymer composite materials from 2011 until now because of the fast advancement of nanotechnology and microelectronic device that demand new materials for their wide range of application. MXene is an example of new material described in the year 2011 that has become supermaterial with excellent thermal and electrical conductivity, chemical stability, and high mechanical properties which are believed to lead to a drastic increase of polymer composite research in material engineering that can be compared with graphene. Graphene also has excellent properties such as thermal properties and has great potential in energy storage technology and electronic device [31].

In this review, the trend of advances in 3D printed polymer composite for the thermal application was presented. Fused deposition modelling (FDM) of 3D printing techniques has been selected among other methods because of ease of manufacturing, low cost, and fabricating complex shapes. This paper aims to analyze and summarize the processing parameter and response parameter of the FDM technique such as base materials, filler materials, processing temperature, and experimental study. This review outcome outlines the progress in developing materials in 3D printed polymer composite based on recent literature. Current or future trends of this area of study and the limitations of current technologies will also be discussed.

2. Thermally conductive nanofiller additive materials

Polymers such as polylactic acid (PLA), ABS, polypropylene (PP), polyethylene (PE), polyetherimide(PE), and others have been widely used in industry for decades because of its flexibility, processability, lightweight, low cost, excellent chemical stability, and excellent mechanical properties. The only technological barrier of these polymers to some applications such as electrical and electronic applications [32], rapid prototyping [33], energy storage [12], and biomaterial, is that polymers have low thermal conductivity. The effort to enhance polymers’ thermal conductivity is crucial for these applications that have become an active research topic in the last decades.

2.1. Single filler

Lots of filler materials can be used to enhance thermal conductivity, as shown in table 1, such as metal, ceramic, and carbon filler material. The metal filler used for thermal application such as silver (Ag), copper (Cu), gold, zink (Zn), stainless steel (SS), aluminium (Al), iron (Fe), titanium dioxide, and zirconia but copper has attracted researcher more due to its high thermal conductivity among other metal fillers. Typical high thermal ceramic fillers are aluminium oxide (Al2O3), silicon carbide (SiC), aluminium nitride (AIN), and boron nitride(BN). Ceramic-based fillers are more preferred than metallic-based fillers for high thermal conductivity and electrical insulation properties and thermal stabilities. However, the electrical conductivity of metallic filler can be tailored by surface treatment or oxidation. Ceramic nanofiller such as boron nitride (BN), silicon carbide (SiC), alumina (Al2O3), beryllium oxide (BeO), silica (SiO2), aluminium nitride (AIN), and zinc oxide (ZnO) also can be used to overcome the problem because of high thermal conductivity and high electric insulation properties. Compared to ceramic-based fillers and metallic-based fillers, carbon-based fillers have received more attention due to their high thermal conductivity. Graphene, carbon nanotubes (CNT), carbon black, and carbon fibres are the carbon-based fillers used in thermal application. Even being used at the smallest filler content from 1 wt% to 8 wt%, the addition of CNT still very effective in enhancing the thermal and electrical conductivity compared to neat ABS [11]. However, graphene is regarded as the most promising candidates with high thermal conductivity, high mechanical strength, and chemical stability. As Vijay T. et al reported, graphene's addition into ABS enhances the thermal stability with thermal decomposition increased by 4.9% [30].
2.2. Hybrid filler
The hybrid nanocomposite combines two or more nanofillers that are employed together as reinforcement in polymer composite. The nanofiller combination can be with carbon, ceramic, cellulose, metal, salts, and others. Generally, the purpose of combining two nanofillers is to enhance the properties of the composite such as thermal, mechanical, electrical properties, and morphologies of the composites. One of the researchers combined ceramic and carbon nanofiller [34, 35] in the research as filament feedstock of FDM 3D printing using silicon carbide (SiC) and carbon (C). The other researcher used silicon carbide (SiC) and Aluminum oxide (Al₂O₃) [8, 36] as hybrid fillers in their research for rapid tooling application. To enhance thermal stability, electrical conductivity, and crystallinity of nanocomposite, graphene nanoplatelets (GNP) and carbon nanotube (CNT) have been combined as hybrid nanofiller for the feedstock of FDM 3D printing [20]. Another effort for thermal enhancement using ABS as polymer and silicon carbide (SiC) and zink (Zn) as hybrid nanofiller [37] for novel 3D printing feedstock.

3. The conducting base polymers
Thermal conductivity is an essential key to the world of research because it characterizes the materials' ability to conduct heat flow when operating with high-temperature procedures. Thermoplastics' composite thermal conductivity is crucially determined by polymer crystallinity that varied from 0.14 W/mK for amorphous polymers such as polystyrene (PS) to 0.53 W/mK for high-density polyethylene (HDPE) which is known as a highly crystalline polymer. Semi-crystalline polymers thermal conductivity, on the other hand, is reported to increase with crystallinity.

3.1. Single polymers
Polymers can be categorized in many ways such as according to the origin of materials; natural or synthetic, based on their thermal response properties; thermosets or thermoplastics, and polymer chain structure; linear, branched, cross-linked, and network. For thermal application composite materials, polymers are categorized based on their thermal response properties: thermosets and thermoplastics. Thermoplastics polymers composite dominated thermosets polymer composite over 93%. Most of the polymer composite experiments for thermal applications using thermoplastics such as ABS, PLA, PEEK, HDPE, PHA, PVC, PC, and TPU compared to thermosts epox. This is due to the brittleness and less tough of thermosets materials which made thermoset are not the choice for composite materials. For thermal application, the commonly used polymers are ABS and PLA, as shown in table 2. The polymers' application includes electrical and electronic, medical, rapid prototyping [21], and aerospace application [28].

3.2. Polymer blends
It is called polymer blends when a system combines or blends two polymers to improve specific properties to the composite while maintaining others. The examples of polymer blends in FDM 3D printing are polyetherimide (PEI)/ polycarbonate (PC), polylactic acid (PLA)/polyhydroxyalkanoate (PHA), polycaprolactone (PCL)/ hydrolyzed collagen (HC), and polyethylene terephthalate (PET)/ polypropylene (PP). Using polymer blends is to improve the processability [21, 38] for the 3D printing filament. Blanco [21] has shown that polymer blends can lower thermal degradation resistance and improve the filament processability by TGA analysis. Other than that, FDM 3D printing was also used to print pots using polymer blends using PCL and HC for biodegradable items for applications in agriculture and plant nurseries where the observed HC thermal stability showed the suitability to be printed with PCL without having thermal degradation [38]. Polymer blends can also be a cost-effective way to reduce plastic usage by recycling plastics such as PET in water bottle bodies and PP in water bottle caps. The recycled plastics are proved as viable printing stock of FDM 3D printing and ease in the process while its tensile strength comparable with HIPS, lower-end commercial filaments [39].
| Fillers Category | Fillers | Composition (wt%) | Thermal Conductivity (W/m.K) | Application | Ref. |
|------------------|---------|-------------------|-----------------------------|-------------|-----|
| Metal            | Silver (Ag) | 40 - 55           | 450                         | Electrical and electronic application | [32] |
|                  | Copper   | 5- 40             | 398                         | Rapid prototyping | [33, 40] |
|                  | Copper oxide | 8- 14            | -                           | -           | [40] |
|                  | Zink     | 10 - 30           | -                           | Rapid prototyping | [8, 37] |
|                  | Stainless steel | 10 - 23        | 247                         | -           | [18] |
|                  | Aluminium oxide | 5- 15         | 30                          | Rapid prototyping | [8] |
|                  | Iron     | 5 - 50            | -                           | Rapid prototyping | [33, 36] |
|                  | Titanium dioxide | 30            | -                           | Rapid prototyping | [36] |
|                  | Zircornia | 30 - 40           | -                           | Biomaterial application | [41] |
| Carbon           | Graphene | 0.2- 8            | 4840-5300                   | Rapid prototyping, energy storage, electrical and electronic application | [5, 6, 12, 15, 30, 42] |
|                  | Graphite | 20-50             | 100-400                     | -           | |
|                  | Carbon nanotube | 1- 15        | 2000-6000                   | Electrical and electronic application | [11, 20, 25, 37] |
|                  | Carbon black | 1.5 - 32.3   | 6-174                       | Electrical and electronic application | [43-45] |
| Ceramic          | Hydroxyapatite | 4             | -                           | Medical application | [46] |
|                  | Barium titanate | 6- 45         | -                           | Electrical and electronic application | [35] |
|                  | Boron nitride | 5- 40          | 250-300                     | Electrical and electronic application | [34, 47] |
|                  | Silicon carbide | 5- 40         | 270                         | Rapid prototyping, energy storage | [8, 12] |
Table 2. Polymer base materials, and their thermal conductivity.

| Base Category | Base Material                          | Abbrev. | Thermal Conductivity (W/(mK)) | Application                | Ref.    |
|---------------|---------------------------------------|---------|-------------------------------|----------------------------|---------|
| Thermoplastics| Acrylonitrile-butadiene-styrene        | ABS     | 0.1                           | Rapid prototyping          | [48-50] |
|               | Polylactic acid                        | PLA     | -                             | Electronic device          | [7, 42] |
|               | Polypropylene                          | PP      | 0.11-0.17                     | Medical device             | [46]    |
|               | Polycarbonate                          | PC      | 0.19-0.21                     | Rapid prototyping          | [21]    |
|               | Poly-ether-ether-ketone                | PEEK    | 0.25                          | Aerospace, medical device  | [28]    |
|               | Polyvinylchloride                      | PVC     | 0.13-0.29                     | Medical device             | [46]    |
|               | High-density polyethylene              | HDPE    | 0.33-0.53                     | -                          |         |
| Thermoset     | Epoxy resin                           | -       | 0.19                          | -                          |         |

4. Nanofiller in thermal application

4.1. Preparation of nanofiller composite
The important in preparing the nanofiller composite is the materials such as a base polymer, the fillers, the composition of the materials, and the apparatus used. The materials' choosing process can be done by reviewing the other publications in the applied application. One of the preparations of nanofiller composite is a solution blending method. The solution blending method uses a solvent liquid such as acetone [18, 37, 51] or chloroform [52] to melt or blend the base polymer before mixing with the nanofiller.

4.2. FDM 3D printing processing parameter and response parameter
To ensure a good quality finishing product with enhanced thermal or mechanical properties of FDM 3D printing, the processing parameter choice must be suitable for its application for 3D printing machine, as shown in table 3. Table 3 shown the optimum process setting parameter for FDM 3D printing including nozzle diameter, nozzle diameter, bed temperature, nozzle speed, filament diameter, infill pattern, infill density, and layer height for thermal application. The filament diameter used by the researchers is 1.75mm [23], 2.85 mm [53], and 3 mm [7] diameter filament feedstock but commonly used for 3D printing is 1.75 mm. As for layer height of FDM deposition are 0.1 mm [54], 0.2 mm [25], and 0.3 mm [4] of layer thickness. On the other hand, bed temperature varied from 60°C [27], 110°C [43], and 120°C [55] depends on the melting temperature of the materials used. Temperature control is critical to control residual stress as residual stress accumulated during layer by layer build-up leads to warping and delamination during the 3D printing process [54].
Table 3. Printing process parameter for FDM 3D printing.

| Parameter          | Value      | References                                      |
|--------------------|------------|-------------------------------------------------|
| Nozzle Diameter    | 0.4mm      | [1, 4, 13, 24, 27, 29, 42, 43, 45, 56-60]        |
| Nozzle Temperature | 230°C      | [15, 32, 45, 49]                                |
| Bed Temperature    | 60°C       | [27, 42]                                        |
| Nozzle Speed       | 30mm/s     | [13, 55]                                        |
| Filament Diameter  | 1.75mm     | [4, 5, 10, 12, 13, 15, 17, 21, 23-25, 30, 32, 35, 40, 43, 44, 50, 55, 61-67] |
| Infill Pattern     | Linear     | [4, 13, 15, 24, 27, 37, 49, 54, 57, 62]         |
| Infill Density     | 100%       |                                                 |
| Layer Height       | 0.2mm      | [13, 25, 29, 35, 43, 49, 61, 66]                |

The response parameter includes thermal properties, physical properties, mechanical properties, morphological, and characterization studies. FDM 3D printing's thermal properties are thermogravimetric analysis (TGA) [13, 21, 30], differential scanning calorimetry (DSC) [13, 24, 30], thermal conductivity [4, 11, 13], electrical conductivity [43], heat capacity [11], electrical resistivity [11], and thermal diffusivity [65]. Still, thermal conductivity, DSC and TGA are the most commonly testing conducted in literature for FDM 3D printing for thermal application. The ASTM testing standard for thermal conductivity in literature is ASTM D5470 for filament thermal conductivities [12]. Simultaneously, physical properties include density [11, 68] and dimensional accuracy [55, 69]. On the other hands, morphological and characterization in literature studies are scanning electron microscope (SEM) [4, 13, 24], transmission electron microscope (TEM) [19, 62], and computed tomography (CT) scan [24], x-ray diffraction (XRD) [24, 26, 62], x-ray photoelectron spectroscopy (XPS) [70, 71], and Fourier transform infrared spectroscopy (FTIR) [72, 73]. Several mechanical properties studies were conducted from literature, such as tensile [4, 11, 23, 24], flexural [13, 24, 25], impact [19, 54, 74], and hardness [27]. Still, tensile, flexural, and impact tests are commonly conducted in literature for mechanical properties for FDM 3D printing technique for thermal application. The ASTM standard testing method for tensile test is D638 Type V (63.5mm × 9.53mm × 4mm) [12, 18, 24], for flexural test is ASTM D790 (127mm × 12.7mm × 3.2 mm) [62, 75], whereas for impact test is ASTM D256 (64mm × 12.7mm × 3.2 mm) [26].

5. Limitations of thermal conductivity of FDM 3D printed composites

The problem with the thermal application for a 3D printed composite is low thermal conductivity. Most of the base polymers have a lower thermal conductivity which is not sufficient for the high heat conduction applications such as electronic application. The addition of heat conductive fillers to polymer-based composite enhances the composite materials thermal conductivity. They typically heat conductive fillers are introduced into polymers to increase the thermal conductivity such as graphene, carbon nanotube, boron nitride, silicon carbide, and metal particles. Sonnalla et al. has experimented with 3D filament contributing ABS as polymer base, Zn flakes, SWCNT, MWCNT, and SCMW as the filler materials, for the enhancement of thermal conductivity of the 3D filament as the sufficient thermal conductivity of filler are more than 100 times than polymer base thermal conductivity to achieve the highest thermal conductivity at any level of filler loading [37]. The addition of graphene nanoplatelets to the base polymer shown increment in glass transition temperature with the increasing graphene volume suggests that thermal stability enhanced with graphene [30].

Other than that, lower filler loading also can lead to low thermal conductivity. However, too much filler can also cause poor composite, low mechanical properties, and high cost. The best option is to use nanofillers such as graphene, silicon carbide, boron nitride, and other, potent materials if only used in a small number of materials. Recent studies of FDM 3D printing have shown that thermal properties can be influenced by the addition of CNT nanofiller to the ABS base polymer by increasing 55% of
thermal diffusivity and 30% increment in thermal conductivity 6wt% of CNT filler loading [11]. The viscosity of extruding filament for the 3D printing cannot be too high which will agglomerate and clogged the nozzle suggests that 4wt% of graphene is the optimal value as reported by S. Dul et al. [15].

The other issue with the 3D printed composites' limitation is the low interaction between the polymer and the filler itself, leading to high thermal interfacial resistance and reducing the polymer composite's thermal conductivity. By improving the interaction of polymer-filler is believed can increase the thermal conductivity of the overall composite. The coupling agent is one of the solutions to improving polymer filler interaction. The commonly used coupling agent is a silane and titanate coupling agent. The titanate coupling agent can improve the polymer-filler interactions by increasing polymer-filler adhesion and increasing thermal conductivity and lowering thermal expansion [37]. Compatibilizing agent, in contrast, functions to stabilize and modifies interfacial between two polymer or polymer blends. The examples of compatibilizing agents in FDM 3D printing are maleic anhydride and styrene ethylene butylene styrene (SEBS). SEBS was proved to improved bonding between phases and led to an increase in glass transition temperature thus increase the performance of the window materials although not significantly increase the tensile strength [39].

Printer orientation of 3D printing is horizontal, vertical, and flat, which is also taking part in thermal conductivity enhancement in a filament. Horizontal printing orientation of 3D printing is believed to give lower thermal conductivity enhancement than vertical printing orientation of filament. It leads to a larger number of layer junctions, more considerable interlayer resistance, and thermal transport along the shortest dimension of filler particle. While vertical orientation reduces thermal interfaces, filler alignment is in the direction of heat flow, and shear forces align fillers in the print directions which leads to enhancement of thermal and electrical polymer composite proved by Sonsalla et al. that filament printed by vertical give 70% thermal enhancement than horizontal print orientation [37].

6. Prospect and recommendation
Thermal conductivity enhancement for 3D printing of thermal application was investigated in this article. All of the experimental and theoretical approaches have been considered in this paper. Since nanofiller materials can influence the thermal conductivity, future research can be extended by combining two nanofiller with higher thermal conductivity in producing filament 3D printing. Also, combining two different base polymers with higher thermal conductivity can be applied for 3D printing thermal applications for new materials.

7. Conclusion
In conclusion, the processing parameter and response parameter of FDM techniques has been summarized. The processing parameter of FDM 3D printing techniques are the materials used and the processing temperature. The most commonly used base material for FDM 3D printing for thermal application is ABS. In contrast, the filler material is graphene, regarded as the most promising candidates with high thermal conductivity, high mechanical strength, and chemical stability. The processing temperature, such as nozzle temperature and bed temperature, also has been reviewed. The nozzle temperature commonly used is 230 °C, bed temperature used is 60 °C, the nozzle speed is 30mm/s, and layer height is 0.2mm. The response parameter includes thermal properties, physical properties, mechanical properties, morphological, and characterization studies. Thermal properties conducted in literature are DSC, TGA, and thermal conductivity while physical properties are density and dimensional accuracy. Tensile, flexural, and impact tests are the mechanical testing conducted in literature. On the other hand, morphological and characterization studies include SEM, TEM, CT scan, XRD, XPS, and FTIR. The limitations such as low thermal conductivity, the material used such as the base materials and filler materials, and experimental parameters such as nozzle temperature, nozzle diameter, nozzle speed, layer thickness, and others have been discussed through this paper and taking consideration for the prospect of FDM 3D printing for thermal application.
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