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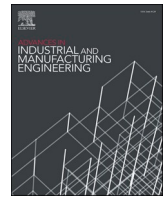
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# Material design factors in the additive manufacturing of Carbon Fiber Reinforced Plastic Composites: A state-of-the-art review

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## ABSTRACT

Materials design advancements are now paramount to further the course of additive manufacturing (AM) of carbon-fiber-reinforced plastic (CFRP) composites. This is due to the increased prospect of such composites in a wide range of applications, ranging from space to automotive subjected to stringent mechanical performance requirements. A synergy of the high strength-to-weight ratio of the CFRP composites coupled with design freedoms inherent in AM techniques offers several interesting opportunities to customize and increase access to mechanical parts. However, several challenges are currently preventing the AM fabrication of the composites from realizing satisfactory mechanical properties compared to some of the traditional methods such as autoclave molding, extrusion molding, compression molding, etc. The challenges can be improved with a better understanding and appropriation of materials design factors that define the controllable material features which could be suitably varied to obtain desired mechanical performances. This paper reviews the literature on the material factors that influence the mechanical performance of parts composed of short-fiber CFRP composites fabricated through the AM technique. Thermoplastic matrix compositions, chain arrangements, and structural morphology effects are discussed in relation to the ease of processing and the final mechanical performance of fabricated composites. Operating environmental effects on mechanical performance were reviewed and also works of literature on the current state of development in the simulation modeling of material factors in the AM fabrication of CFRP composites were discussed.

## 1. Introduction

### 1.1. Additive manufacturing of Carbon Fiber Reinforced Plastic Composites

Opportunities inherent in realizing mechanical parts with carbon-fiber-reinforced plastic (CRFP) composites are necessitating investigations into the principles that guide materials designs for the additive manufacturing (AM) of CRFP composites. Previously, CRFP composites have been majorly manufactured through autoclave molding in which the fiber and the plastic matrix are laid on a tool in a desired sequence and spot welded to ensure that the stacked plies do not shift positions relative to each other before vacuum bagging and autoclave forming (Wang et al., 2011). Other traditional methods such as injection molding in which molten composite is injected into predefined molds under high pressure to form the shape of the cavity, and compression molding in which molten composite is condensed within the male and

female cavities of a mold to into a shape, etc. have also been used (Rajak et al., 2021). Constraints within these traditional composites' manufacturing limit the quality of emergent composite parts.

AM technique enables new capabilities for composite parts offering benefits in terms of design customization and optimization, prototyping time reduction, access to parts, materials waste reduction, service reparability, and many more when compared to other traditional manufacturing processes. Some of these benefits of AM fabricated CRFP are depicted in Fig. 1. According to Huang et al. (2013), AM techniques do not require fixtures, cutting tools, and other auxiliary resources to fabricate parts, expanding the potential for part optimization and customization. Tymrak et al. (2014) found that AM techniques could potentially reduce the quantity of part inventory held by the aerospace industry. Attaran (2017) agrees, emphasizing AM's potential to minimize lead times, and time-to-market, hence fulfilling orders quicker than other traditional manufacturing methods. Klahn et al. (2015) highlighted AM's design freedom since it is less restrictive in terms of

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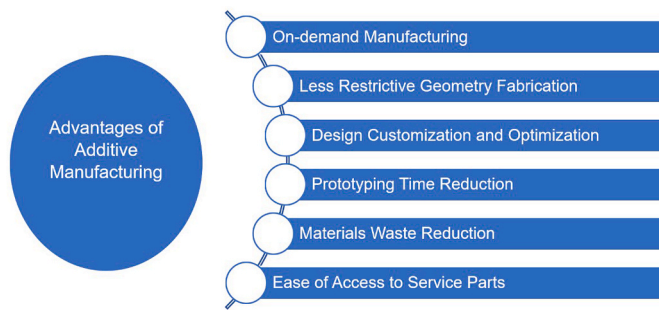


Fig. 1. Summary of some of the advantages of Additive Manufacturing the Traditional Manufacturing Methods.

geometry designs while Ford and Despeisse (2016) discussed the benefits of AM from the cradle to the grave of products i.e., across the entire product life cycles. These include product and process design, material selection and processing, make-to-order component, and product manufacturing.

These benefits are proliferating the use of AM in the customization of advanced materials like CFRP composites. CFRP composites realized with AM techniques are finding application in several industries (Petrovic et al., 2011), including automotive (Pervaiz et al., 2016) aerospace (Kroll and Artzi, 2011), and the military (Angrish, 2014; Mattox, 2013). For example, AM is being used to manufacture the wing structures of Unmanned Aerial Vehicles (UAVs) as observed by Goh et al. (2017). They described recent developments in the exploitation of AM techniques to realize light-weighted CFRP flapping wings for UAVs. Also, Goehrke (2015) used AM fabricated CFRP composites to develop small-scale wind turbines for a relatively economical value of power source at remote locations, while ongoing development is exploring AM fabricated CFRP composites for energy harvesting applications in wind turbine components (Transforming Wind Turbine Blade Mold, 2022). This is aimed at leveraging previous successes in using these techniques to fabricate molds for wind turbines as detailed in (Hassen et al., 2016; Kunc et al., 2017). For space-bound applications, Mitchell et al. (2018), discussed the exploitation of AM techniques to realize a lunar base, deployment mechanism for solar panels, and the realization of a miniaturized satellite. The portability of certain AM systems allows their deployment in space stations where they could be used to manufacture replacement parts.

The AM technology as a material joining process applies three-dimensional model data, to make parts usually in a layer-upon-layer build as opposed to subtractive manufacturing and formative manufacturing methodologies (ASTM International, 2000). Different AM methods such as Fused Deposition Modeling (FDM), Selective Laser Sintering (SLS), Stereolithography (SLA), Inkjet, and Laminated Object Manufacturing (LOM) are available for the AM fabrication of plastics (Dikshit et al., 2020; Singh et al., 2020; Parandoush and Lin, 2017). However, among these arrays, the FDM method, which schematics are illustrated in Fig. 2 is currently the most viable for realizing the AM fabrication of CFRP composites, with minimal investigations reported on the use of the other AM methods in the fabrication of CFRP composites.

The essential elements of the FDM system include the material feed mechanism, liquefier, print head, gantry, and build platform. A Stereolithography (STL) file containing the features of a part to be printed is sliced into successive two-dimensional layers which are further used to generate translational G-codes. The G-code guides the print head's horizontal (x, y) and vertical (z) movement. The nozzle moves according to the G-code, depositing a thin layer of an extruded composite called "road". This solidifies quickly upon contact with the heating bed or an adjacent layer (Bellini and Güçeri, 2003). A variant of the FDM technique feeds a prepreg composite filament material through, the printer head, gantry, and unto a build platform. The filament is transformed into

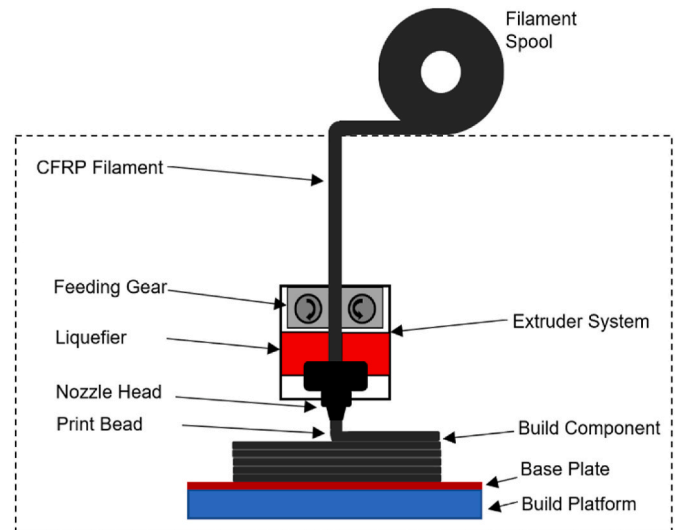


Fig. 2. Key elements of the fused deposition modeling (FDM) process. Based on vaes and puyvelde (Vaes and van Puyvelde, 2021).

a semi-liquid state and then re-extruded before deposition in a layer-to-layer manner. The location of filament deposition is guided by features in an STL file (Mohamed et al., 2015).

Fig. 3 shows a layer-to-layer printing of AM fabrication of CFRP composite.

### 1.2. Current challenges in the AM fabrication of CFRP composites

AM fabricated CFRP composites are yet to fully feature in several commercial applications especially those with stringent requirements. This is a consequence of relatively poorer mechanical performance compared to the more standard parts. These have recently led to investigations into several aspects of AM fabrication of CFRP composites. Much of this work focused on fused deposition modeling, part of which this review of literature contributes to improving. The poorer properties of an AM fabricated CFRP composite have been linked to materials and process limitations (in the form of part porosities), weak fiber-matrix adhesion, uncontrolled fiber dispersion alignment, and anisotropy (Tekinalp et al., 2014; Shofner et al., 2003; Duty et al., 2017; Zhang et al., 2018).

Tekinalp et al. (2014) compared AM and compression-molded (CM) CFRP samples in which they used scanning electron microscopy (SEM) images of fractured surfaces to contrast pore formation issues in parts

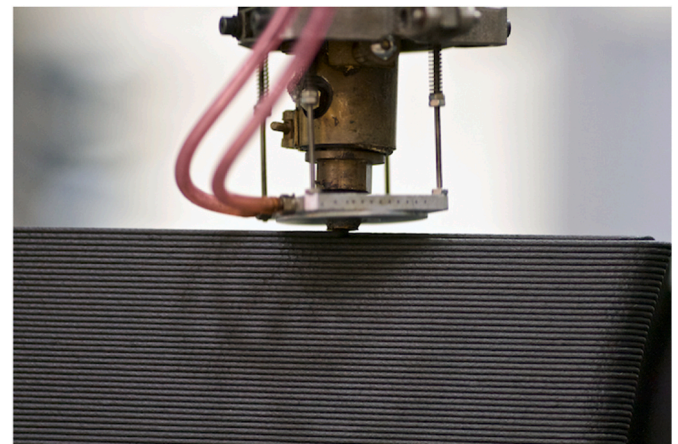


Fig. 3. Large-scale AM printing of CFRP composite (US Department of Energy, 2014).

manufactured with the AM technique. They showed the issue of porosities in gaps between the print beads in AM fabricated thermoplastics with and without the effects of fibers. Before this, Shofner et al.'s (Shofner et al., 2003) study found the fracture mode in AM fabricated CFRP matrix to be more brittle than ductile due to interfacial fiber-matrix bonding issues. Similarly, they found fiber dispersion and misalignment issues during processing led to material anisotropy, thereby, compromising mechanical properties. Duty et al. (2017) found similar issues, also associating the mechanical properties to porosities, interfacial bonding, inflexible fiber placement, anisotropy, etc. They observed that CFRP printed parts exhibit anisotropic behavior across the axial (x-axis) and transverse (z-axis) directions. This was attributed to a variation in the extent of fiber-matrix bonding along the axial build direction when compared to the weaker interlayer bonding across the transverse axis of the composite.

### 1.3. Motivations for the review paper

A conceptual analysis of material design factors in innovative technologies like the AM fabrication of CFRP composites is needed to cultivate the right knowledge for the sustainability of the manufacturing technique. This should involve evaluating the material design factors that would improve the sustainability of CFRP realization using AM techniques. A review of relevant literature that promotes an understanding of materials design factors that affect the mechanical performance of short-fiber CFRP composites fabricated through the AM technique is needed. The paper is aimed at presenting such a review by providing an insight into state of the art on fundamentals knowledge needed to improve the mechanical performance of CFRP through the balance of fiber content, fiber length, fiber orientation, fiber morphology, fiber chemistry, aspect ratio, fiber diameter, fiber-matrix adhesion, plastic matrix chemistries, monomer chain arrangements, polymer structural morphologies, etc. These aspects form the basis needed to address AM fabricated CFRP composites' performance issues.

### 1.4. Review outline

This review discusses relevant literature that has investigated various material effects on the mechanical performance of AM fabricated CFRP composites. Specifically, it addressed short-fiber thermoplastic CFRP composites fabricated through the FDM method.

It reviewed the literature on short fiber and matrix material effects on the ease of fabrication, material mechanical properties, and environmental performance. It finally examined the literature on the simulation models of material design factors.

## 2. Material design factors

Material design factors define the controllable material features that can be suitably varied to obtain the desired performance of the final material. In combining plastic matrices with carbon fiber (CF) as reinforcements, CFRP composites achieve improved strength-to-weight ratios, stiffness-to-weight ratios, corrosion resistance, thermal conductivity, etc. (Zhu et al., 2018; Lin et al., 2020). However, composite properties can be optimized with the consideration of materials design factors.

Material features such as fiber and matrix composition, fiber and matrix material morphologies, fiber sizes, fiber-matrix aspect ratios, etc. would affect the physical and chemical interactions. This will influence the flowability of the composite during manufacturing and the emergent mechanical properties of the fabricated part which are gained as a compromise between those of the constituent part materials as illustrated in Fig. 4. In the fabricated part, the CF generally provides strength and stiffness while the plastic matrix protects the fiber, and transfers load through the composite. The matrix material also provides ductility and toughness as such in combination with the fiber to improve the

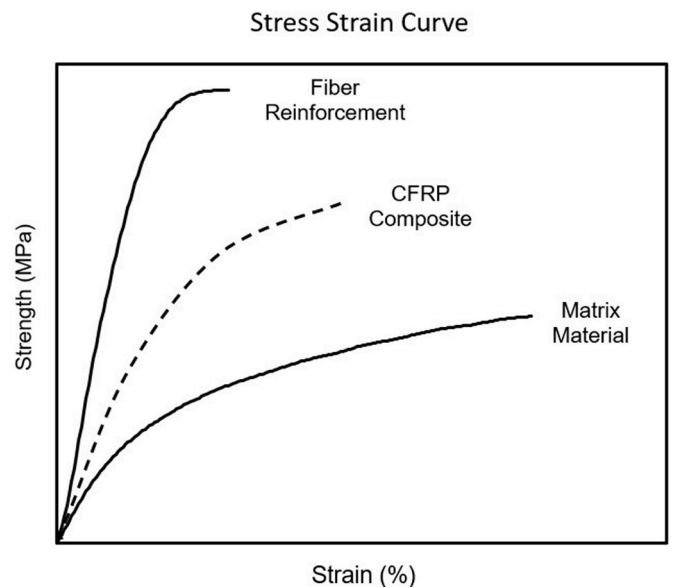


Fig. 4. Mechanical performance expectations of carbon fiber reinforced plastic composites over individual carbon fiber and thermoplastics matrix materials.

mechanical properties of the composite. Enhanced integration of the fiber with the matrix would result in effective load transfer that could withstand higher mechanical stresses.

The composite benefits from a synergy of the ductility of the matrix and the high strength of the fiber. Mechanical strength inherent in the composite would be much higher than that of the unreinforced matrix, yet more ductile than the carbon fiber. By optimizing design parameters and changing the matrix material, the characteristics of the composites can be tuned for strength, modulus, ductility, and other properties relevant to an intended application. Fig. 5 presents the dependence of tensile strength of some CFRP composite on fiber content for a few thermoplastic matrices including Acrylonitrile-butadiene-styrene (ABS), Polyamide6 (PA6), Polyamide12 (PA12), and Polyetherimide (PEI) as reported by Duty et al. (2017), Zhang et al. (2018), Ning et al. (2015), Mohammadzadeh (Mohammadzadeh et al., 2021), Liao, et al. (Liao et al., 2018), Love et al. (2014), etc.

### 2.1. Carbon fiber reinforcement

Carbon fiber (CF) strength and stiffness make it suitable for improving the mechanical properties of plastic materials in a quest to realize composite with higher impact resistance, chemical resistance, and thermal stability. Its high strength-to-weight ratio is nearly twice that of 6061 Aluminum (UnterwegerOliver Bruggemann, 2014) As such it is one of the strongest and stiffest synthetic fibers with viable reinforcement options for most short fiber applications. CF can mitigate warping and distortion while offering a satisfactory level of insulation owing to its low coefficient of thermal expansion (Tekinalp et al., 2014; Duty et al., 2017).

In the broader field of composites, short fibers are preferred to continuous fibers in applications requiring intricate shape fabrication. Besides from the solid-filled infill parts which are more highlighted (Zhang et al., 2018; Ning et al., 2015; Mohammadzadeh et al., 2021; Liao et al., 2018), geometrically complex honeycomb and lattice structures are more easily fabricated with short fibers and with impressive mechanical performance (Santiago et al., 2021; Quan et al., 2016; Austermann et al., 2019; Hao et al., 2019; Goh et al., 2022). Santiago et al. (2021) applied short carbon fibers in fabricating complex lattices and stochastic foams with overhanging features from CF-PEEK by AM in which they found a dramatic improvement of the mechanical properties of the CF-PEEK composite which finds viable applications as custom

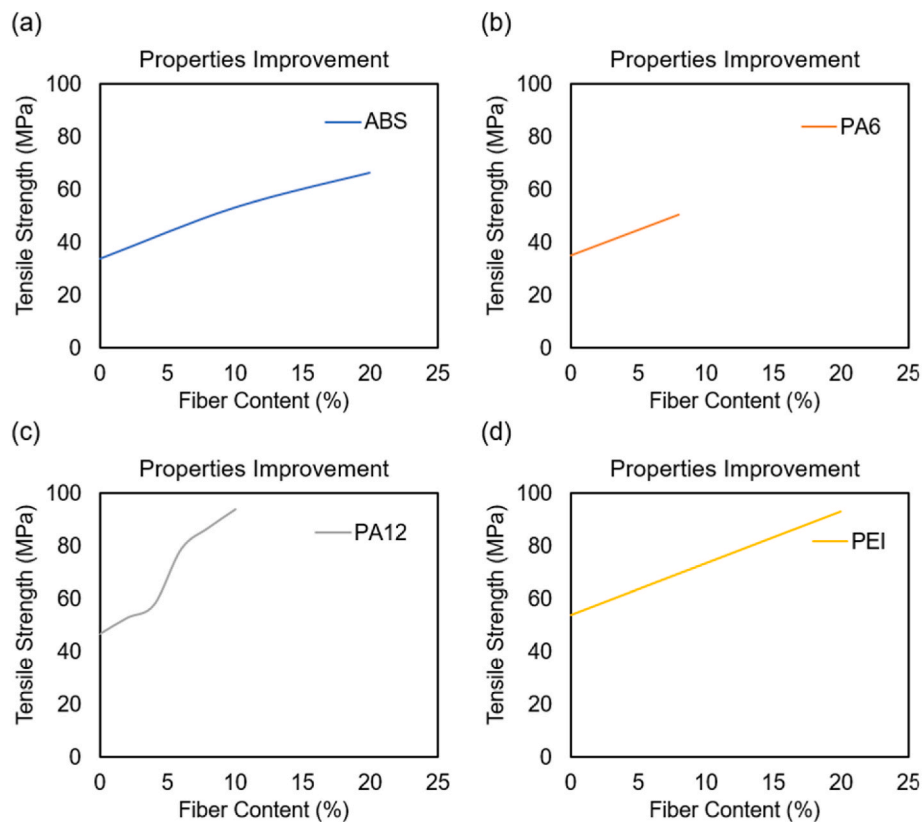


Fig. 5. Tensile properties improvement trends with carbon fiber addition in different thermoplastic matrixes of AM fabricated CFRP composites (a) ABS (b) PA6 (c) PA12 (d) PEI (Tekinalp et al., 2014; Duty et al., 2017; Mohammadzadeh et al., 2021; Liao et al., 2018).

implants or aerospace structures. More than 100% improvement in tensile and more than 900% in flexural was observed over pure PEEK lattice structure, which was attributed to the short carbon fiber, that is known to enhance mechanical performance and, in this case, acted as filler to reinforce the matrix.

Quan et al. (2016) evaluation of a different matrix material found the short carbon fiber design to influence the mechanical properties of orthogonal CF-ABS composite lattice preforms. They found the compressive properties of CF-ABS composite and that of its silicone-infused composite to demonstrate much higher mechanical properties, giving credence to the feasibility of applying honeycomb and lattice structures to CFRP composite structural applications. The results from Austermann et al. (2019) in their new process development to fabricate hybrid CFRP composite lattice sandwich structures found compressive and flexural properties: strengths and modulus improvements. Contributions from the carbon fiber, part geometry, and relative density were found as determinants. While the material composition defines the basic properties, the topology and geometric features also contributed.

AM fabricated short fibers are seen to offer good acoustic properties. Goh et al. (Hao et al., 2019) in evaluating various short fiber layouts of face sheets and core designs of AM fabricated composites found acceptable values for the indentation resistance and acoustic absorption performance of fabricated lattice structures. Similarly, Hao et al. (Goh et al., 2022) found short fiber-reinforced composite lattice structures tested for tensile strength and modulus to exhibit higher properties and more stable repetitive energy absorption in the lattice structures with the short fiber-reinforced than for unreinforced composites. On the other hand, they found higher strength and modulus properties not to apply in the compression mode but observed more stable repetitive energy absorption.

The fiber features with important influences on mechanical

properties have been determined (Fu et al., 2009; Huang et al., 2021; Breuer and Stommel, 2020). Among these, the parameters most relevant to the AM fabrication of CFRP composites are depicted in Fig. 6. The link between the parameters is aimed at showing the strong interrelationships among them. While studying short fiber plastic composites, Fu et al. (2009) found the fiber-matrix interface features, fiber length distribution, fiber orientation, fiber volume fraction, etc. to be critical to the mechanical performance of the composites. Breuer et al. (Breuer and Stommel, 2020) elaborated on the contributions of fiber packing, fiber shape, fiber-matrix bonding, fiber length distribution, and fiber orientation on the mechanical performance of thermoplastic composites and went further to simulate the properties of the composite with micro-mechanical models using representative volume elements (RVE).

### 2.1.1. Fiber morphology

The CF morphologies include the diameter, cross-sectional shapes, voids, surface consistency, etc. of the fiber. The most common short carbon fibers that are used in AM applications include the precision-cut chopped (PCF) and the milled (MLF), while the less commonly used ones include carbon nanotubes (CNT) and graphite fibers (GRF). The PCF offers more reinforcement properties marked by overall better mechanical properties, while the MLF offer improved dimensional stability and electrical conductivity. Designing for fiber morphology would influence material properties and could help tailor to meet certain AM fabricated CFRP composite requirements.

Savandaiah et al. (2021) comparison of fiber morphological sizing effects on some of the mechanical and thermomechanical properties of AM fabricated CF-PP composites using PCF and MLF are summarized in Fig. 7. They reported the 275  $\mu\text{m}$  average length PCF to offer much higher tensile strength and modulus (about 30% and 75%, respectively) than the 200  $\mu\text{m}$  average length MLF (Fig. 7a). Similarly, flexural strength was much higher more than 50% for the PCF (Fig. 7b). Fiber

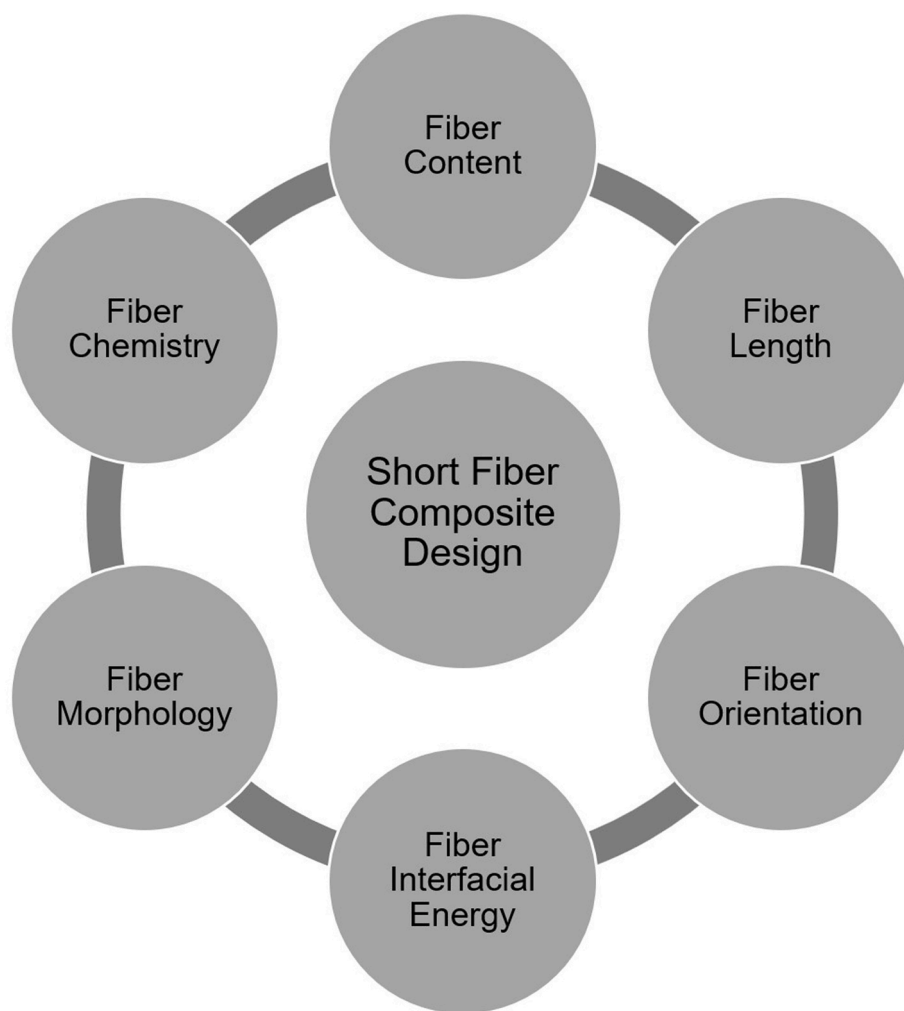


Fig. 6. Short fiber composite design factors for AM fabricated CFRP composites.

morphology differences didn't result in any significant differences in the torsional storage modulus over a broad range of temperatures from  $-40\text{ }^{\circ}\text{C}$  to  $+120\text{ }^{\circ}\text{C}$ . However, a pronounced difference in the absolute  $\tan \delta$  value above the glass transition temperature,  $T_g$ , was observed with the values for the MLF higher than those of the PCF (Fig. 7c), which can be ascribed to the fiber morphology's compatibility with the matrix. The mean void volume for the MLF was significantly lower ( $4.01 \times 10^{-5}\text{ mm}^3$ ) compared to that of PCF ( $7.02 \times 10^{-5}\text{ mm}^3$ ), which reflects in the impact properties (Fig. 7d) and signifies the role of fiber morphology on the fiber-matrix compatibility.

The investigations by Ning et al. (2017a) comparing the mechanical performance of the PCF versus GRF also found more void formation in the AM fabricated PCF-ABS compared to the GRF-ABS composite. However, the PCF-ABS exhibited better tensile properties than the GRF-ABS because of the higher fiber strength of the PCF and the better matrix bonding between adjacent layers. Zhang et al. (2017) in comparing AM fabricated PCF-ABS and CNT-ABS composites found the PCF to be more effective in reducing shrinkage compared to CNT. However, the CNT-ABS composite exhibited lower porosity due to the higher contact area between the matrix and the CNT morphology. Considerations of the CF morphology influences on material porosity, shrinkage, conductivity, etc. could help to control for better material properties in the quest to improving the mechanical performance of AM fabrication of CFRP composites from its current state.

### 2.1.2. Fiber chemistry

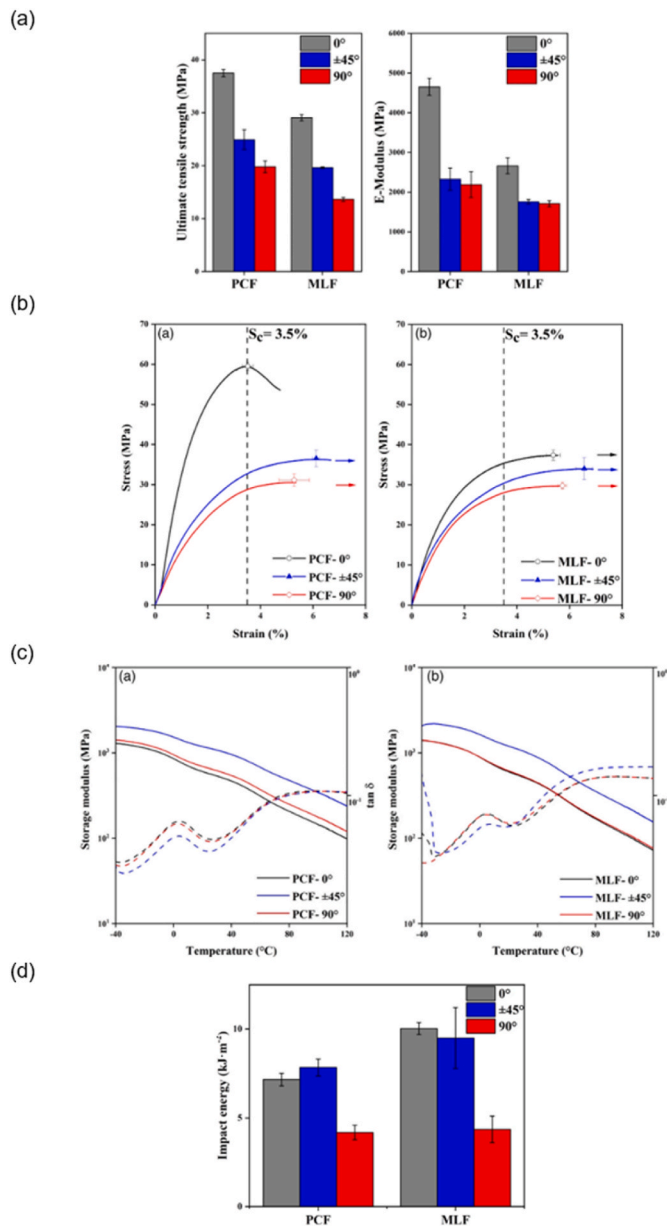
The fiber chemistries are majorly influenced by the precursor

characteristics used in manufacturing the fiber, which also determines the fiber output strengths and modulus. Based on these chemistries, the synthetic CF commonly used in CFRP composites is classified as pitch or PAN-based, which Huang et al. (Huang, 2009) categorized according to modulus values into five grades namely: the ultra-high modulus (UHM)  $> 500\text{ GPa}$ , high-modulus (HM)  $> 300\text{ GPa}$ , intermediate modulus (IM)  $> 200\text{ GPa}$ , standard modulus (SM)  $> 100\text{ GPa}$ , and high strength-high strain (HT)  $> 4\text{ GPa}$ .

Beckman et al. (2021) discussed the PAN-based IM CF chemistry offering the highest tensile strength, and the pitched-based UHM and HM CF offering the highest tensile modulus in composite applications. However, there are limited investigations to confirm these effects on the mechanical performance of AM fabricated CFRP composites. Such a theory that the PAN-based CF generally achieves a higher modulus with a compromise of lower strength, and the pitch-based a lower strength, but higher modulus is yet to be confirmed for AM fabricated composites. However, should be considered in materials design for the AM fabrication of CFRP composites. Fig. 8 shows the tensile strength and modulus differences between some of the commercially available PAN-based and pitch-based CF.

### 2.1.3. Fiber content

Fiber content effects are one of the most discussed topics in the mechanical performance of AM fabricated CFRP composites. Ensuring the optimal fiber content is important to the effective exploitation of AM fabricated composites and this will vary for contrasting matrix materials. The consensus from most of the AM fabricated CFRP composite



**Fig. 7.** Properties Comparison for PCF and MLF Reinforced AM Fabricated CF-PP Composites at Different Print Orientations (a) Tensile (b) Flexural (c) Torsional The Storage Modulus ( $G'$ ) and Loss Factor  $\tan \delta$  (d) Impact Properties. Reproduced by Permission (Savandaiah et al., 2021).

publications to date is that CF composites with 20% fiber content can be reached with a gradual increase in tensile performance up to this percentage in most thermoplastic composites. Further increase in the content of fiber requires further development. Table 1 summarizes recent publications that relate fiber content to mechanical performance. Most were only able to test CFRP composites up to 20% CF, except for Tekinalp et al. who were able to investigate one specimen sample at up to 40% CF content.

Duty et al., 2015, 2017 showed that by increasing the CF content to 20%, the tensile strength and modulus of CF-ABS could reach a tensile strength and modulus of up to ~66 MPa and ~12 GPa, respectively, however, this comes with some degree of anisotropy. Ning et al. (2015) agree with this observation till 15% CF content which was the limit of the study. On the contrary, they observed that the composite's toughness and ductility are compromised by increasing CF content. Tekinalp et al.'s (Tekinalp et al., 2014) investigation of up to 40% CF content in

ABS matrix found up to 115% and 700% increase in tensile strength and modulus, respectively over unreinforced thermoplastics, which they ascribed to the high orientation of the CF along with the print orientation. Also, Love et al.'s (Love et al., 2014) investigation of 13% CF claimed the addition of CF to polymer feedstock significantly increases the strength, stiffness, and thermal conductivity, decreased the coefficient of thermal expansion, and greatly reduced the distortion of the parts. Similarly, Mohammadzadeh et al. (2021) reported an increase of up to 40% in the tensile strength and 80% modulus properties for AM fabricated PA matrix reinforced with 8% CF.

Tekinalp (Tekinalp et al., 2014) related the increase in fiber content to a decrease in the interlayer porosities, but some increase in the inter-bead porosities. Their brief scanning electron microscopy (SEM) image analysis of the polished surface of failed gauge sections shown in Fig. 9 (a) to (d) found the void volume fraction to fluctuate between 16% and 27% independent of fiber content. They attributed this to the competing effects of changes in large interlayer voids to smaller inter-bead voids with increasing fiber content. However, the decrease in die-swell with the increasing CF content resulted in smaller beads and smaller inter-layer porosities. Thermal conductivity increased with CF content which also helped to improve interlayer adhesion between successive print layers to provide an overall improved bond strength.

#### 2.1.4. Fiber length

The concept of critical fiber length,  $L_{cr}$ , given by equation (1) has shown some relevance in designing AM fabricated CFRP composites of suitable mechanical properties. The critical length is the minimum length beyond which the maximum allowable fiber strength can be achieved. It determines how likely the fibers can handle the stress from the matrix via shear. Longana et al. (Gerrit Blok et al., 1320) described the critical length as the length at which the failure mode changes from fiber pull-out to fiber breakage, which equates to the length that allows the full strength of the fiber. The theory is that fibers below this critical length will be ineffective in reinforcing the matrix.

$$L_{cr} = \sigma_f d / 2 \tau_y$$

$$\text{Equation 1}$$

where  $L_{cr}$  is the critical fiber length,  $\sigma_f$  is the fiber ultimate tensile strength ( $\text{N/mm}^2$ ),  $d$  is the fiber diameter, and  $\tau_y$  is matrix-fiber bond strength or matrix shear yield strength ( $\text{N/mm}^2$ ) whichever is greater.

The critical length of CF is important to its reinforcing ability and has been determined to range from 500 to 600  $\mu\text{m}$  depending on the matrix material (Unterweger et al., 2020; Graupner et al., 2016; Mortazavian and Fatemi, 2015; Capela et al., 2017). The length in the AM fabrication process may be limited by the filament and printing extrusion, where the extruder heads may break down the length. Fiber length design should be explored around the extrusion head to optimize the fiber length performance. Savandaiah et al.'s (Savandaiah et al., 2021) analysis of the fiber length distribution before and after the filamenting extrusion process of CF-PP composites found 50% of the CF composition to have their lengths decreased from 275  $\mu\text{m}$  to 200  $\mu\text{m}$  for milled CFs and from 5953  $\mu\text{m}$  to 275  $\mu\text{m}$  for precision-cut chopped CFs because of the filamenting process. Their analysis of the before and after length distributions for the respective fibers using a proprietary image processing software, Image-Pro plus, and FASEP 3E is illustrated in Fig. 10.

The investigations by Ning et al. (2015) which compared 150  $\mu\text{m}$ –100  $\mu\text{m}$  fiber length averages found the higher 150  $\mu\text{m}$  to provided higher tensile strength and modulus, respectively in the excess of 10% and 40%, over the 100  $\mu\text{m}$  supporting the theory that further length reduction below the critical length translates into reducing mechanical properties (Unterweger et al., 2020; Graupner et al., 2016; Mortazavian and Fatemi, 2015; Capela et al., 2017).

#### 2.1.5. Fiber orientation

An advantage of short fiber over continuous fiber is the ability to easily embed it into intricate features of part design. However, this

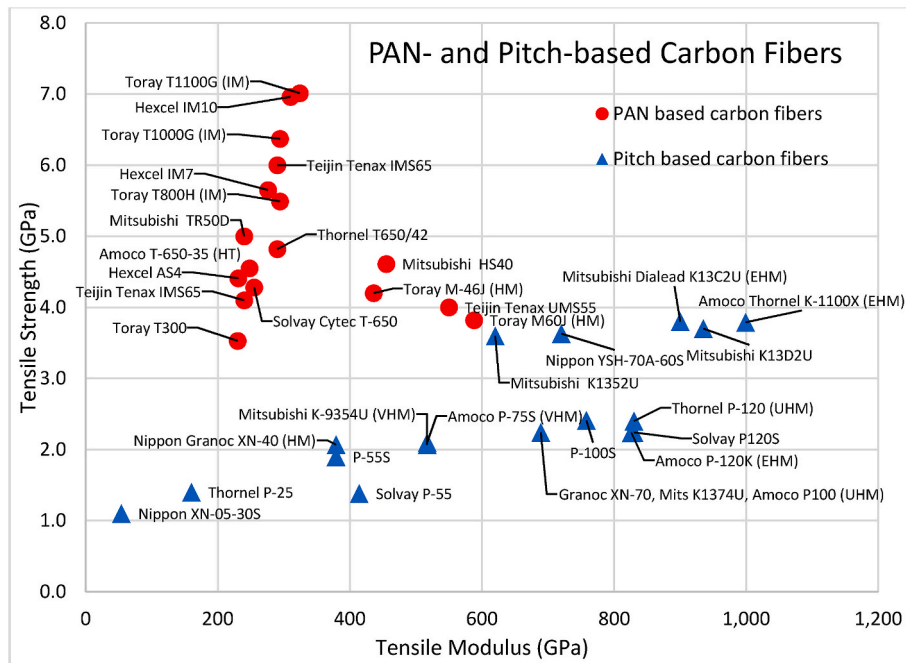


Fig. 8. Strength and Modulus Comparison Trends of Commercially Available CF PAN and Pitch-Based Chemistries. Reprinted with permission (Beckman et al., 2021).

Table 1

Some of the previous investigations of fiber content effect on the tensile properties of AM fabricated short-fiber CFRP composites layered in the axial (x) print orientation of test specimens.

Matrix Material	Fiber Percentage (%)	Author
ABS	0, 13, 20	Duty et al., 2017
	0, 3, 5, 7.5, 10, 15	Ning et al., 2015
	0, 10, 20, 30, 40	Tekinalp et al., 2014
	20	Hill et al. (2016)
	0, 13	Love et al., 2014
	0, 15	Zhang et al., 2018
PA6	0, 8	Mohammadzadeh et al., 2021
PA12	0, 2, 4, 6, 8, 10	Liao et al., 2018
PEI	0, 20	Duty et al., 2017

usually minimized the contributions of fiber in the intricate region to the mechanical properties of the composites due to fiber misalignment. Although, the ability to adaptively orientate the CF fibers along the

cartesian coordinate of the print bed could potentially optimize the mechanical properties of AM fabricated CFRP composites, however, such control is yet to fully feature on AM composite printing equipment. Table 2 presents the typical mechanical properties of the carbon fiber along the cartesian coordinate. where  $E_{ij}$  is the corresponding young's modulus in the x, y, and z-direction of the cartesian coordinate system. It should be noted that fibers inclined at an angle to any of the Cartesian axes will not maximize their strength along that axis.

Inclusion of features that allow some control of the orientation and alignment of the fiber will increase the viability of AM fabricated short fiber CFRP composites. Such et al. (2014) in their review of the methodologies of short fiber composites' manufacturing discussed how developing highly aligned and oriented discontinuous fibers can be used to overcome the geometrical complexity restrictions of continuous fiber, while theoretically allowing for similar mechanical properties. They developed a model, illustrated in Fig. 11, which predicts that highly aligned short fibers would provide the optimum balance of processability and performance in thermoplastic composites.

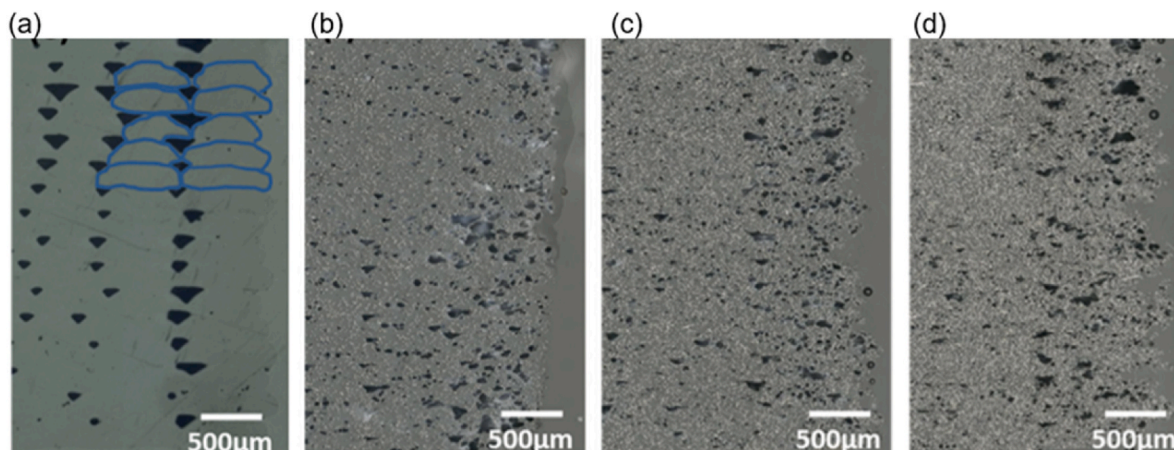


Fig. 9. Sem images of polished surfaces of AM fabricated CF-ABS with different fiber contents (a) Neat ABS (b) 10% CF (c) 20% CF (d) 30% CF (Tekinalp et al., 2014) (Reprinted with Publisher's permission).



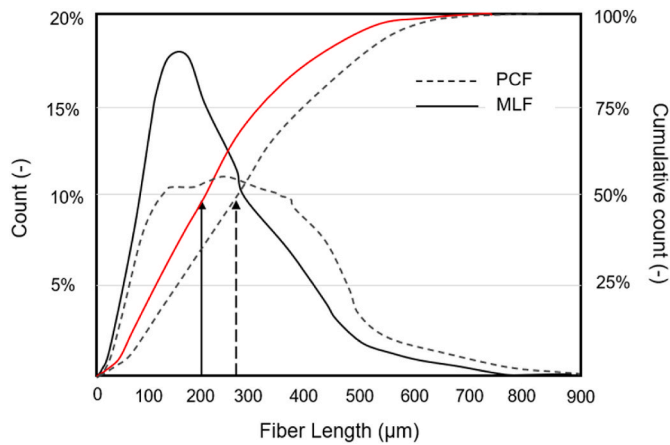


Fig. 10. Fiber length distributions of milled and precision-cut carbon fiber measured from a CF-PP filament spool (Savandaiah et al., 2021) (Reprinted with Publisher’s permission).

**Table 2**  
Carbon fiber materials properties in the cartesian coordinate. Based on (Mattsson et al., 2008).

Material Property	Cartesian Coordinate	Value
Elastic Modulus (GPa)	E <sub>11</sub>	230
	E <sub>22</sub>	20
	E <sub>33</sub>	20
Poisson’s Ratio	v <sub>12</sub>	0.2
	v <sub>13</sub>	0.2
	v <sub>23</sub>	0.2
Shear Modulus (GPa)	G <sub>12</sub>	20
	G <sub>13</sub>	20
	G <sub>23</sub>	8.3

Tekinalp et al. (2014) support this theory by attributing the high tensile strength (67 MPa) and modulus (13.7 GPa) observed at 40% CF in a CF-ABS to the high alignment of the CF along the cartesian print direction. Maintaining high orientation and distribution will improve the interlayer bond strength and the overall viability of AM fabrication of CFRP composites.

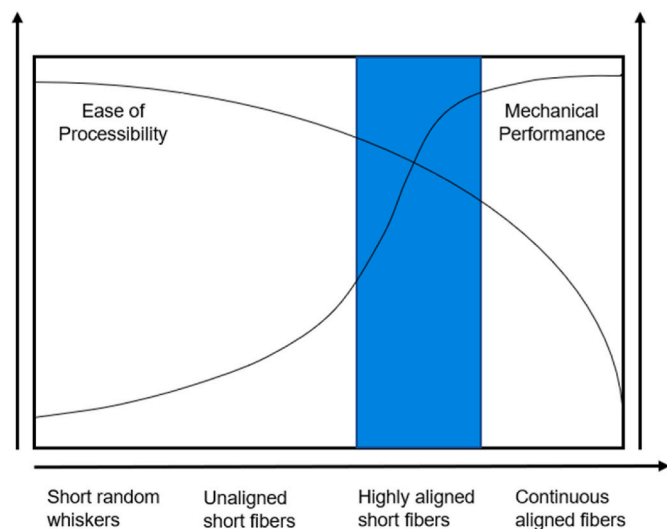


Fig. 11. Ease of Processability and Mechanical Performance ‘Sweet Spot’ of highly Aligned Short fibers. Based on (Such et al., 2014).

### 2.1.6. Interfacial energy

Wide surface energy gaps between the CF and a matrix material would result in poor interfacial bonding within the print beads (Gray et al., 1998). A balance of fiber surface energy with the matrix material surface energy is necessary to achieve a good mechanical performance of AM fabricated CFRP composites. According to Ning et al. (2017b), poor interfacial bonding between the CF and matrix results in fiber pullout from the matrix, thereby causing lower mechanical performance in the composite. Selecting CF grades with the least surface energy differences with the matrix or vice versa can help to optimize interfacial bond strength.

The option of binding additives has also been prescribed to improve the issue of fiber-matrix interfacial energy compatibility. Savandaiah (Savandaiah et al., 2021) applied 1.8 vol% of processing aid and 0.9 vol % of coupling agent in their processing to AM fabricated CF-PP composites to improve interfacial bond adhesion between the fiber and matrix. Rangisetty et al. (Rangisetty, 2017) proposed fiber treatment for CF to increase interfacial bonding with the matrix material. Fiber treatments can be used to tailor interfaces between organic and inorganic materials by simply modifying wetting and adhesion to enable comparable surface energies of the CF and matrix.

### 2.2. a.m. thermoplastic matrix materials design

The choice of the plastic (also referred to as polymer) matrix has a huge influence on the overall mechanical performance of the composite. The matrix bonds, protects and propagates load in-between fibers and into the entire composite. They also provide ductility and toughness, synergizing with the fibers to form a composite with superior properties to those of its constituents. An appropriate matrix also offers good processability, thereby, minimizing any adverse effect of the AM process on the emergent properties of the fabricated composite.

Two classes of matrix materials are commonly used to realize CFRP composites. The first, thermoplastics, is more amenable to offering reversible chemical changes, especially where temperature changes occur, while the other is more difficult to reprocess as such, they are terms as thermosets (Visakh et al., 2016). The recyclability of thermoplastics at elevated temperatures and later solidification at lower temperatures makes them good candidates for the FDM. According to Jiang et al. (2020), weak van der Waals forces within the structure of thermoplastics enable them to soften when heated above their glass transition temperature ( $T_g$ ) where they become viscous. This process is completely reversible on cooling below the  $T_g$  owing to the absence of chemical de-bonding and bonding. As such the preference for thermoplastics over thermosets as matrix materials is a consequence of their ease of processability, reparability, maintenance offering, etc. These are particularly essential features as we seek ways to enforce a sustainable future. Also, thermoplastics do not require cure cycles which complicate the fabrication process, with their reversible physical state often allowing easier and cheaper reparability of parts (Mohamed et al., 2015; Van de Werken et al., 2020). According to Fidan et al. (2019), AM thermoplastic matrices must be thermally, physically, and chemically stable; emphasizing the need to be compatible with the fiber without reacting with them. They also highlighted their need to be able to keep the fibers in place, transfer stress between fibers, protect the fibers from adverse environmental conditions, prevent surface abrasion, and support the fibers under loading application.

Depending on their functionality, thermoplastics used as matrix materials can be classified as commodity, engineering, or high-performance grades. Fig. 12 shows some of the common examples for the different classifications. The commodity grades are majorly used for non-load bearing functions and find applications where low mechanical performance is required. The engineering grades are the most used in AM fabricated CFRP composites’ load-bearing applications whenever exceptional mechanical properties such as strength, stiffness, ductility, toughness, and impact resistance are desired. The high-performance

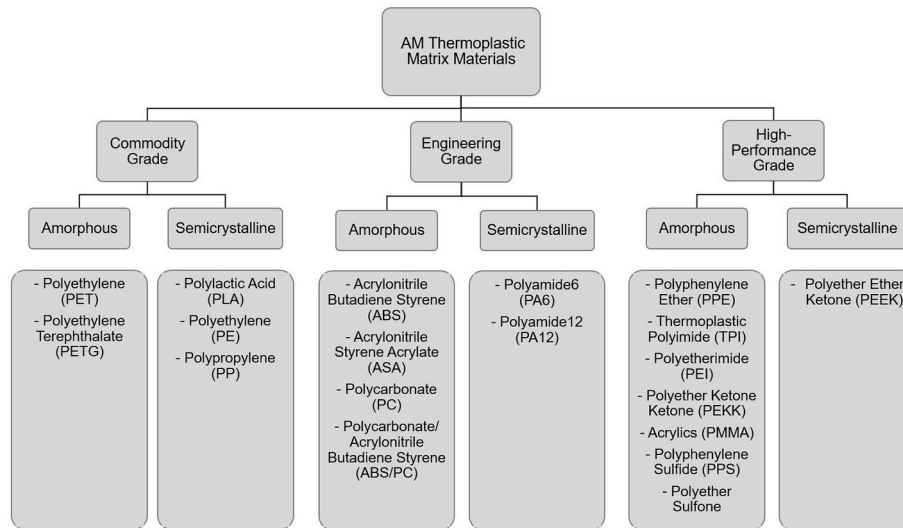


Fig. 12. Developed Thermoplastic Matrix materials for AM Fabricated CFRP Composite Applications.

grades also find applications in load-bearing functions, especially where higher temperature stability and chemical resistance are required. However, could be limited by the ease of processing through the FDM method.

Material features such as polymer composition, chain arrangements, structural morphologies, etc. define the polymer properties and mechanical performance capacities. Table 3 shows the mechanical properties of the common engineering and high-performance grades used in AM fabrication of CFRP composites.

2.2.1. Matrix composition effects

The polymer molecules form an important constituent of the AM fabricated CFRP composite. Mechanical and thermal features increase with the degree of polymerization, arrangement of carbon and hydrogen atoms (e. g. numbers of aromatic over aliphatic units), linking of aromatics with oxygen, (Flory and Vrij, 1963), etc. The degree of polymerization (DP) is given as the mean number of monomer units in the polymer. This is the ratio of the molecular weight of the polymer to the molecular weight of repeat units (Cowie and Arrighi, 1991; Allcock and LFWMJP, 2003). A difference in the degree of polymerization of plastics of similar compositions will result in differences in their physical properties and this should duly be considered in matrix composition design.

According to Jain et al. (2019) by increasing the choice of matrix

material’s molecular weight, thermoplastic degradation during the AM fabrication process can be mitigated. Alternatively, chemical modification of thermoplastic composition with supramolecular is another way to enhance the printability of thermoplastic materials. Consequently, Chen et al. (2019) found that challenges experienced when printing polyethylene terephthalate (PET) due to, rapid crystallization after extrusion can be mitigated by incorporating phenylacetylene (PEPN) groups featuring  $\pi-\pi$  interactions as a side chain. They proposed the incorporation of the pendant PEPN chain prevented recrystallization upon cooling down due to the destroyed chain regularity by the pendant and that the decreased differential temperature between the melting temperature ( $T_m$ ) and glass transition temperature ( $T_g$ ) of the enhanced composition enabled a rapid solidification during the printing process.

2.2.2. Polymer Chain Arrangements effects

Aromatic structures composed of carbon and hydrogen atoms in ring structures versus aliphatic structures in straight or branched arrangements tend to offer better bond strength within the polymer, hence, better mechanical properties. Jain et al.’s (Jain et al., 2019) systematic study of the relationship between saturated, pendant side chain length and ease of AM printability showed that increasing side chain lengths (aromatics) configuration of polymers could reduce viscosity and enable extrusion at low temperature and pressure, while the long-chain lengths (aliphatic) configuration could reduce the ease of formability. Fig. 13

Table 3 Summary of the average mechanical property values of common AM Thermoplastic Matrix materials (3DXTECH Additive Manufacturing, 2022).

Thermoplastic Grade	Thermoplastic Material	Tensile Yield (MPa)	Tensile Modulus (GPa)	Tensile Ductility (%)	Flexural Yield (MPa)	Flexural Modulus (GPa)
Engineering	Acrylonitrile-Butadiene-Styrene (ABS)	42.0	1.95	1.0	76.0	1.99
	Acrylonitrile-Styrene-Acrylate (ASA)	45.0	2.01	1.0	78.0	2.00
	Polyamide-6 (PA6)	55.0	1.98	10.0	76.0	2.05
	Polycarbonate (PC)	62.0	2.41	7.0	78.0	2.20
	Polycarbonate/Acrylonitrile-Butadiene-Styrene (PC/ABS)	62.0	2.41	7.0	78.0	2.20
High-Performance	Polyphenylene Ether + Polystyrene (PPE + PS)	67.0	2.25	12.0	85.0	2.30
	Thermoplastic Polyimide (PEI)	74.0	2.95	6.0	110.0	2.86
	Polyetherimide (PEI)	56.0	2.5	3.0	110.0	2.51
	Polyether Ether Ketone (PEEK)	100.0	3.72	28.0	130.0	2.70
	Polyether Ketone Ketone (PEKK)	105.0	2.75	5.0	95.0	2.68
	Acrylics (PMMA)	68.9	2.65	16.5	90.8	2.51
	Polyphenylene Sulfide (PPS)	50.0	2.55	18.0	52.0	2.54
	Polysulfone (PSU)	52.0	2.1	8.0	87.0	2.05
	Polyphenyl Sulfone (PPSU)	55.0	2.31	3.0	11.0	2.22

illustrates a comparison of the aromatic and aliphatic polymer chain configurations.

The degree of oxygen functionalization in the chain arrangement influences the ease of processibility (Warren and Ditor, 1996). Similarly, the degree of hydrogen bonding particularly influences printability. Rupp et al. (2019) demonstrated this, by comparing ease of printability based on linear and three-arm star supramolecular polymers, which they used to support the theory for AM fabricated thermoplastic matrices. The polymer printability is based on reversible thermal and shear-induced dissociation of the supramolecular polymer network. Thermoplastic viscosity is influenced by the composition and blend formulation of the polymer chain to determine the printing window and the structural stability of printed parts. The molecular architecture of the plastic matrix material is determined by the hydrogen bond configuration which would affect the composite printability from the plastic material viscosity control under heated temperature.

### 2.2.3. Polymer morphological effects

The crystallinity of the thermoplastics plays a huge effect on the mechanical properties of the composite. Thermoplastics of similar compositions and grades but different morphological structures tend to exhibit widely varying strengths, modulus, ductility, and toughness. Based on the morphology, thermoplastics can be categorized as amorphous or semi-crystalline. According to Rubinstein (2003), the amorphous consists of entangled chains without order or crystal formation, while the semi-crystalline consists of densely packed ordered crystalline regions with lamellar morphology packed parallel to each within some disordered amorphous regions. Fig. 14 illustrates the amorphous and semicrystalline structural morphologies.

The amorphous morphology offers superior modulus over the more ordered and closely packed semicrystalline matrix due to the loose packing of their molecules. The loose molecules also allow for more gradual softening of the material when heated above the glass transition temperature, which corresponds to the temperature at which a sudden change in molecular mobility occurs, and the material transitions from a glassy, brittle to a more rubbery state (Gofman et al., 2013). The dense packing and ordered crystalline regions in the semicrystalline morphologies set them apart from the amorphous structures. The crystals stay in their orderly packed lamellar structure up until their melting point at which they transform to the semi-liquid state, thus, able to support much higher service temperatures than the amorphous thermoplastics. They typically experience a dramatic reduction in mechanical properties at glass transition temperatures,  $T_g$  (Throne, 2017). Common examples of semicrystalline thermoplastics include Polyamides (PA), polyethylene (PP), polypropylene (PP), Acetal, etc. while the common examples of the amorphous include Acrylonitrile Butadiene Styrene (ABS), Polycarbonate (PC), PETG, Polystyrene (PS), Poly Vinyl Chloride (PVC), etc.

Amorphous thermoplastics in addition to the good mechanical

properties exhibit several interesting processing features such as high fluidity, low  $T_g$ , and a wide window of processing temperatures which makes them excellent for AM fabrication. Fabricated parts may however be characterized by weaker intermolecular forces from the loose packing of the thermoplastic monomer chains. On the other hand, the crystallinity of the semicrystalline thermoplastics provides them with some distinct features which usually offer superior mechanical properties but can yet present some difficulties to the processing of the thermoplastics (Vaes and van Puyvelde, 2021). Self-nucleation in the crystals can impact the printing process in the form of insufficient heat transfer and melting and high shear deformations upon extrusion, which limits chain mobility. This inhibits the development of interlayer strength and dimensional accuracy owing to excessive shrinkage.

Benedetti et al. (2019) highlighted the main cause of shrinkage as crystallization, which is the rearrangement of the chains into a structure with lower volume. They discussed the requirement for a broader processing window for semicrystalline thermoplastics to overcome the issue of rapid crystallization and distortion and explained how a higher degree of crystallinity such as in the semicrystalline versus the amorphous leads to higher shrinkage. Their theory is illustrated in Fig. 15 which compares the shrinkage for the two thermoplastic morphologies when cooled below their  $T_m$  and  $T_g$ .

This behavior of the thermoplastic matrix due to crystallization and distortion was yet confirmed by Adeniran et al. (2022), who investigated the influence of the thermoplastic matrix on the mechanical performance of AM fabricated composites. Their comparison of the meso-structure formation of amorphous CF-ABS and semicrystalline CF-PA6 of similar 15% fiber content and fabrication temperatures shown in Fig. 16 revealed excessive shrinkage in the interlayers of the semicrystalline CF-PA. Such shrinkage leads to huge interlayer porosities which are the causes of the lower modular properties exhibited in semicrystalline form as opposed to the amorphous CFRP composites. Hence, confirming the influence of matrix morphology on AM fabrication of CFRP composites.

### 3. Materials design for manufacturability

The feasibility of certain materials for AM fabrication of CFRP composites should be taken seriously by identifying suitable process parameters that can be used to influence the properties of the materials. Selecting unsuitable materials for any particular manufacturing process would compromise the ease of processing and the quality of the manufactured part. Researchers have therefore demonstrated the importance of optimizing the material design to suit process parameters and vice versa for AM fabrication of CFRP as seen in (Vaxman et al., 1989), (Ning et al., 2017a), Gray IV et al. (Gray et al., 1998), Zhang et al. (2017), Parandoush and Lin (2017), Alafaghani et al. (2017), etc. The consensus was that void formation increases with fiber concentration, fiber length, extrusion rate, extrusion temperature, and decreasing draw ratio. Such issues can be mitigated with careful design of material and process

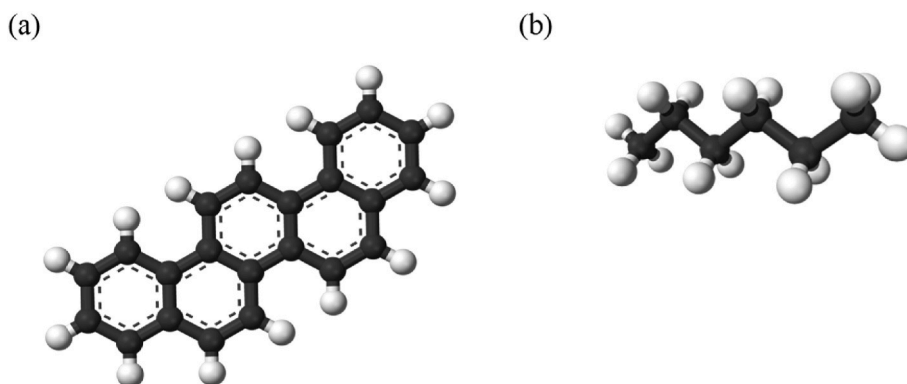


Fig. 13. Polymer chain arrangements (a) aromatic (b) aliphatic (Madhusa, 2017).

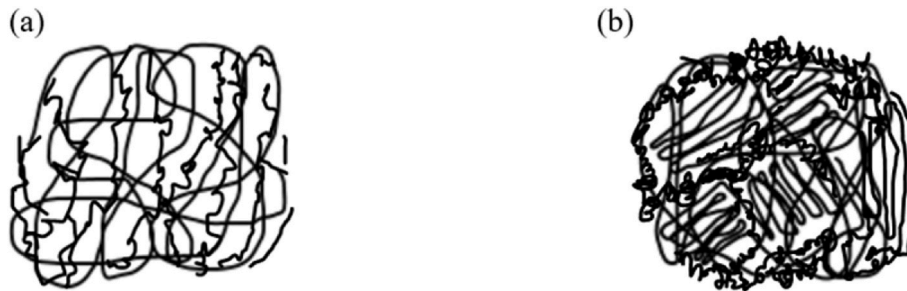


Fig. 14. Illustration of the thermoplastic morphology arrangements (a) amorphous (b) semicrystalline.

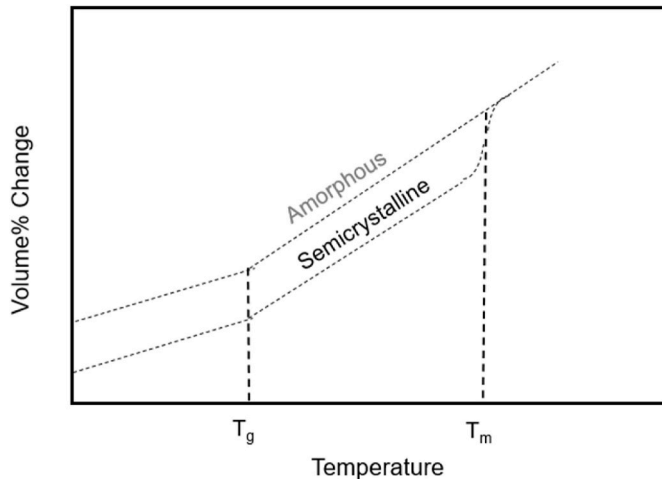


Fig. 15. The difference in Volume% Change Between Amorphous and Semicrystalline thermoplastics matrix Upon Part Cooling. (Based on (Benedetti et al., 2019)).

factors. Optimized process parameters such as part build orientation, raster angle, infill speed, layer thickness, nozzle diameter and temperature, print bed temperature, etc. Design considerations should also be given to material processing operations precluding the printing process such as the composite compounding and feedstock filamenting which could also be a determinant of the quality of fabricated parts.

### 3.1. Composite compounding

Due considerations should be given to determining the optimum process conditions for the miscibility of the fiber and matrix. The role of melt-mixing temperatures and pressures in the success of the composite miscibility of the melting temperatures of the thermoplastic constituent has been discussed by researchers such as Tekinalp et al. (2014), Guo et al. (2013), Giancola and Lehman (2012), Zhang et al. (2011), etc. Tekinalp et al. (2014) incorporated up to 40% of 3.2 mm length-sized chopped Hexcel AS4 CF and ABS copolymer (GP35-ABS-NT) for which they controlled process temperatures and speed, respectively to 220 °C and 60 rpm rotor speed in a Brabender Intelli-Torque Plasti-Corder prep-mixer to ensure miscibility. Guo et al. (2013) also applied the torque co-rotating twin-screw extrusion process to improve the miscibility of Multiwalled Carbon Nanotubes (MWCNTs)-Polylactic Acid (PLA). They described the selection of the temperature and torque to be suitable to prevent material degradation, and empirically adjusted over the heat zones to ensure miscibility and extrusion. Giancola and Lehman (2012) discussed the criticality of the temperature of the solids conveying zone to ensure good wetting of the polymer on the extruder wall, which is important to provide sufficient shear stress to the extrusion process.

Other AM composite material designs for fabrication may require the

introduction of binder additives to ensure better fiber and matrix miscibility. The investigation by Wang (2017) which also used the melt-mixing compounding process to formulate CNF-polypropylene (PP) found the need for binder addition to improving miscibility. Their addition of up to 3% maleic anhydride polypropylene (MAPP) increased the crystallization of the PP thermoplastic matrix by 14% which they attributed to the MAPP function as a nucleating agent which helped the PP to form smaller spherulites and more spherulitic sites. They also related the MAPP binder additive to improving the compatibility between the CNF and PP, distributing the CNF better in the PP, thus, enhancing the ability of the CNF to nucleate.

### 3.2. Feedstock filamenting

The fiber and matrix material melt mixing and surface energy compatibilities should be considered in optimizing both the compounding and filamenting processes. The composite output of the compounding operation is processed in a filamenting device which breaks down and remelts the composite chunk in a heated screw extruder into a continuous slender thread-like filament. The process temperature and speed will vary depending on the matrix material and fiber content and considerations should be given to these factors to achieve miscible, porosity-free, and consistent thickness filaments. Fig. 17 illustrates the filament fabrication process which consists of composite compounding and filamenting.

Various designs to achieve the miscible, porosity-free, and consistent thickness requirements have been reported. Tekinalp et al. (2014) reported using a plunger-type batch extrusion unit heated to 220 °C and with a slit-shaped die to extrude their filament their highly aligned CF-ABS composite which reported the highest tensile properties improvements of AM fabricated CF-ABS to date. Adeniran et al. (2021) also successfully used a similar filamenting process in an extruder machine (Felfil Evo, Turin, Italy), with a single screw filament extruder of 1.75 mm, die-head with the temperature set to 220 °C and speed at 7 rpm to process up to 30% CF-ABS composites. While most of the commercially available filament sizes are 1.75 mm and 2.85 mm, larger filament sizes can be made with larger nozzle sizes. These are currently being realized for custom applications and form the basis for several ongoing research.

### 3.3. Printing process

The FDM method has been widely researched with many established process parameters for different fiber and matrix material features. Thermoplastic matrix suitability for viscoelastic formability is very fundamental to the success of the process. Such et al. (2014) explained the strong influence of the thermoplastic matrix rheological and thermal phenomena during printing on its manufacturability which in part is influenced by the process parameters. The addition of fiber reinforcement to the thermoplastic matrix increases the molten viscosity of the bulk composite, increasing the heat capacities and conductivity to change the temperature profile during and after deposition. The contrast in viscosities between the matrix and fiber could compromise the

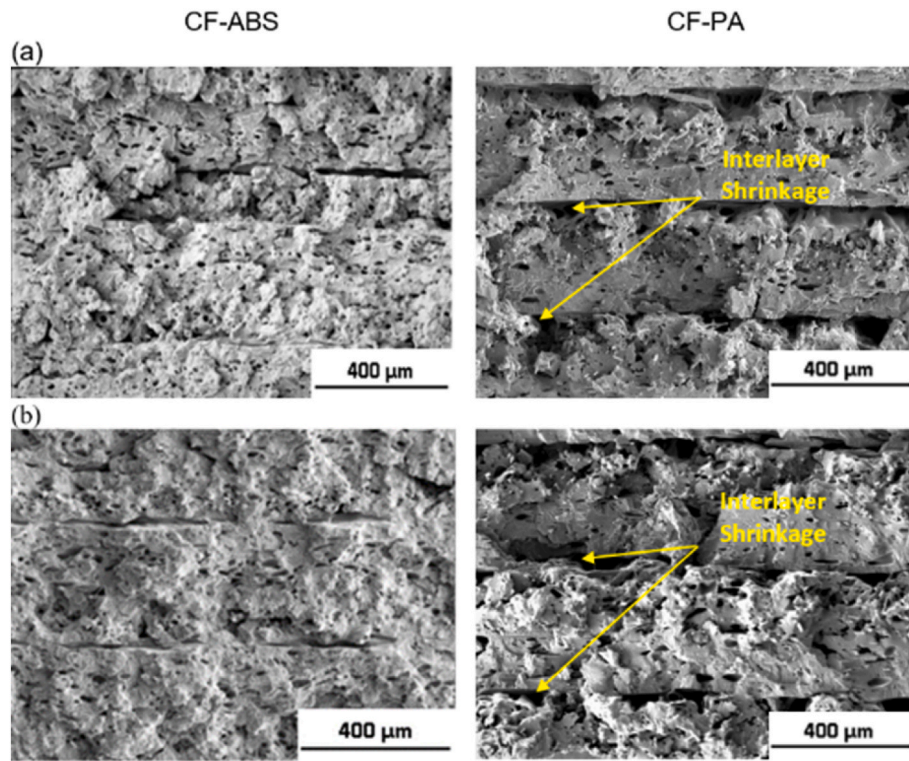


Fig. 16. Thermoplastic volume change and cooling effects on mesostructure formation in amorphous CF-ABS and semicrystalline CF-PA composites fabricated at (a) 250 °C and (b) 270 °C.

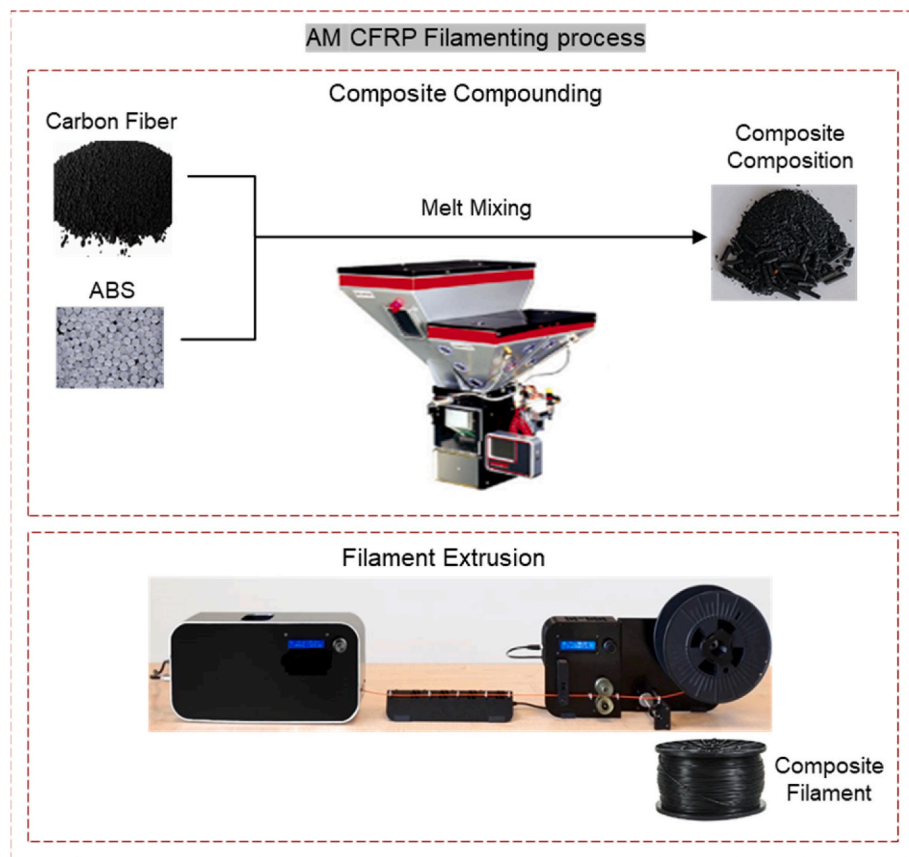


Fig. 17. Cfrp composite filamenting process for AM fabrication.

bonding process within the print bead and between the interlayers, thereby, negatively affecting the mechanical properties. Thus, it is important to consider the compatibility of the fiber and matrix material before manufacturing.

Mohamed et al. and several other researchers (Mohamed et al., 2015; Ning et al., 2017b; Ahn et al., 2002; Goh and Yeong, 2018) analyzed the effect of printing process parameters such as part build orientation, raster angle, raster width, contour width, infill speed, infill pattern, infill density, layer thickness, etc., and machine setup parameters such as nozzle temperature, nozzle diameter, print bed temperature, etc. on the interlayer properties to influence the mechanical performance of AM fabricated CFRP composites. Mohamed et al. (2015) highlighted the absence of perfect "one size fits all" optimal process conditions for all types of material designs and explained the need to determine the balance of process parameters for different material designs to ensure the tradeoff between material quality and production time. Ning et al. (2017b) found that the raster angle, infill speed, nozzle temperature, and layer thickness will affect the tensile strength, modulus, toughness, and ductility of the printed material and that the properties of the AM fabricated composite can be controlled by the process parameters. Ahn et al. (2002) also demonstrated that process raster angles can influence material porosities which affect the tensile strength, but that road width, printing temperature, and material color have minimal effects.

Zhang et al.'s (Zhang et al., 2018) characterization of the interfacial bond strength of AM fabricated CF-ABS composite using in-plane tensile shear and double notch shear test methods showed the raster angles to have some effects on the interlayer bond strength where they found  $0^\circ$  raster angle to offer the least porosity, followed by  $90^\circ$  and then  $\pm 45^\circ$ , with direct translations to the tensile strength. Elevated bed temperature settings were also applied to explain the presence of smaller voids at the bottom of the specimens as higher temperatures, above the glass transition of a matrix is believed to result in more fusion effects at the bottom layers.

Young et al. (2018) applied a modified double cantilever beam (DCB) and a single-end notch bend (SENB) test to examine printing parameter effects on the fracture toughness of chopped CF-ABS composites. The data revealed the fracture toughness of CF-ABS composite can be limited by poorly wetted chopped carbon fibers and good wetting should be ensured for the efficient application of fiber reinforcement benefits. Similarly, Goh and Yeong's (Goh and Yeong, 2018) investigation of the Mode I interlaminar fracture toughness of AM fabricated CF-PA composite to understand the effect of process temperatures on fracture toughness showed increases in the Mode I interlaminar fracture toughness with the nozzle and bed temperatures but a decrease with printing speed leading to the recommendations for the careful consideration of process parameters hand in hand with materials design to optimize AM fabricated CFRP composites' mechanical performance.

As the fiber content effect in increasing the mechanical performance of the composite increases. However, consideration should be given to the possible negative effects in processing, the major one of which is the printhead nozzle clogging which results from random fiber orientation and the reduced viscosity of the semi-liquid composite flow. Most of the reported investigations were not able to test more than 20% fiber volume which may be due to the clogging issue typical above this fiber volume. Advances to matrix materials of 30% fiber content which was associated with nozzle clogging. Adeniran et al. (2021) ran into a similar nozzle clogging issue in their investigations of the carbon fiber volume effects of the compressive and tensile properties in which the nozzle got clogged and couldn't support the cycle time of a tensile workpiece beyond 20% CF volume.

Understanding and paying attention to designing fiber critical fiber length into the manufacturability of AM fabricated CFRP composites is also an important factor since the printing process may limit the fiber retention of the optimal critical length in the course of the printing process through the nozzle. Fiber length should be chosen close to the critical length, around the nozzle size, and other process conditions to

minimize breakage and nozzle clogging on extrusion. Gray et al. (1998) expressed the possible difficulties of extruding short fibers through the die head and still maintaining a high aspect ratio fiber (i. e.  $L/D > 100$ ) due to the small diameter capillary die action (0.3 mm) common in AM fabrication. However, the closer the fiber size to the critical length, the better the mechanical properties get.

The effect of matrix material melt processing on nozzle head flow and clogging should be considered and carefully designed. Ajinjeru et al. (2018a) specified the need to identify the relationship between matrix material design and the appropriate processing conditions. Their investigations of the dynamic rheology behavior of high-performance polyetherimide (PEI) thermoplastic to its CFRP composite (Ajinjeru et al., 2018b) found the composite exhibiting viscous liquid characteristics in the extrusion process. The addition of CF to PEI enhances the shear-thinning but significantly increased the viscosity. Their comparison of the rheology behavior of a high-performance grade polyphenyl sulfone (PPSU) with engineering-grade acrylonitrile butadiene styrene (ABS) thermoplastic found the ABS behaving more like an elastic solid while the PPSU was more like a viscous liquid and showing a potential variation of  $2-3 \times$  over the range of expected extrusion shear rates.

Duty et al. (2018) presented a practical model for evaluating thermoplastic matrix feedstock materials as candidates for AM fabrication across a variety of extrusion-based platforms. They discussed the series of fundamental conditions that should be met to consider a thermoplastic material for successful utilization in AM fabrication of CFRP composites. They explained the material's need to be able to meet a pressure-driven extrusion through a given diameter nozzle at a specified flow rate, the need for the ability to form and sustain the desired shape, the need for the ability to bridge a specified gap, and the ability to serve as a mechanically sound foundation for successive deposits and the need to be dimensionally stable during the transition to the final state. The matrix material's viscosity and the ease of processibility should be carefully considered in the matrix material design for ease of fabrication and to achieving the desired mechanical performance of AM fabricated CFRP composites.

Process environmental control is also a necessary consideration for an effective fabrication process since the presence of moisture in the filament and print vicinity would result in material porosities. Researchers Leite et al. (2018), Halidi and Abdullah (2012), and Nidagundi et al. (2015a) discussed the effects of processing environmental conditions on the print quality of thermoplastics and their composites. They reported on the hygroscopicity of ABS and how the material moisture absorption can lead to porosity defects. The same issue also applies to thermoplastic matrixes such as polyamides, polycarbonates, acrylics, etc. Measures should be put in place to limit and get rid of absorbed moisture in filaments before and during printing. Measures such as storing the filaments in a heated oven at below melting temperatures to get rid of moisture absorption should be employed. A measure by Adeniran et al. (2022) illustrated in Fig. 18 was employed to control the printing environment temperature to reduce the effect of moisture. Thus, improving the melt flow and interfacial bonding by employing a heated printer enclosure system to control the temperature to less than  $50^\circ\text{C}$  and relative humidity to less than 20% RH.

A fundamental understanding of the relationships between material design and ease of fabrication is needed to be put into consideration to achieve the effectual optimization of the composites' mechanical properties.

#### 4. Materials design for environmental performance

Emerging applications for AM fabricated CFRP composites under various operating environmental conditions require improving materials design for mechanical performance. Already, AM fabricated plastic composites are being used in critical exterior components of Cosmic Antenna Array parts in space applications (Stratasys, 2006) and for wind turbine applications (Post, 2016), and many more emerging

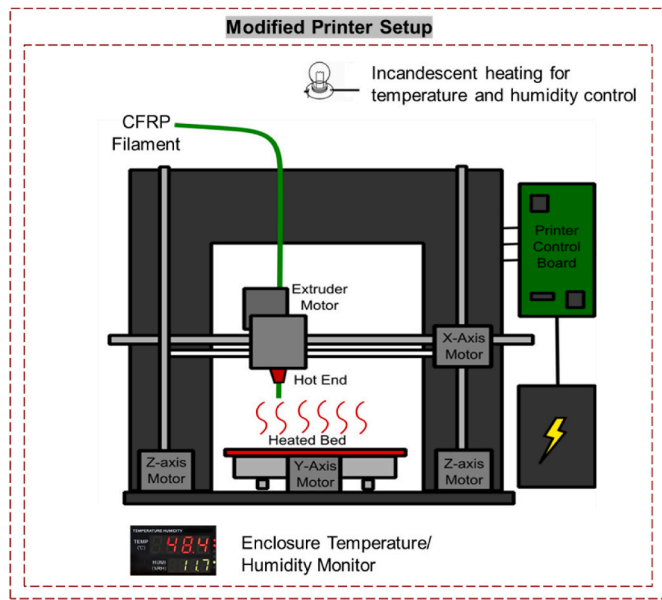


Fig. 18. Modified printing setup with incandescence heating enclosure for moisture control (Adeniran et al., 2022).

applications. According to Mitchell et al. (2018), there is an outlook towards potential space applications, in areas including deployable structures, antennas, hinges, etc. that would be subjected to some form of environmental effects. This calls for improved developments that will be spurred by effective designs for material mechanical performance to achieve sustainability under such applications.

Environmental elements are known to affect a material's mechanical properties. This is particularly true for plastic-based materials since they are susceptible to moisture absorption at low and elevated temperatures. Kim et al. (Kim et al., 2016) compared the environmental effects on the traditional injection molded and AM fabricated thermoplastics. After the part was immersed in water at 60 °C, more degradation was found in the AM fabricated parts which led to these parts having a tensile strength of 26%–56% of the injected molded parts at room temperature; and 67%–71% at the hot wet environment. They associated this with more porosities in the AM fabricated part.

A prior understanding of CFRP composites' environmental requirements would enable a better adaptation of the parts for different applications. Many developments are ongoing in the industry some of which involve major players like Stratasy, Markforged, etc. These are at the forefront of developing materials for various outdoors, space, and environmental applications. It should be noted that carbon fiber by itself has very good environmental properties, as such combining it with highly anhydrous matrix materials would improve melt flow, interfacial bonding, heat transfer, etc. in the AM fabrication of CFRP composites (Dabiri et al., 2014; Yin et al., 2018; Costa et al., 2019; Adeniran et al., 2022).

## 5. Simulation modeling of material design for AM fabricated CFRP composites

Developments in theoretical and computational modeling are fostering materials designs and selection in the general materials science field, which can rightly be extended to AM fabricated CFRP composites. At the current stage, simulation modeling of the composite still needs development as limited literature is available on AM composites, rather than only for plastic materials. More accurate predictions are also needed from the need to incorporate the AM process factors such as interlayer porosity, inter-bead porosities, fiber-matrix interface bond strength, etc. into the models.

Developed and more accurate models would help to better predict the mechanical performance for existing and emerging applications which would improve the acceptance and the pace of development of the composite in a wider range of industrial applications. Already a sizable number of experimental investigations have reported on the mechanical properties of the composites (Hassen et al., 2016; Duty et al., 2017; Ning et al., 2015; Van de Werken et al., 2020; Adeniran et al., 2021, 2022). However, a limited number is still reported on simulation modeling. To date, the handful of simulation models reported on mechanical performance only focused on the matrix material and the interlayer effects while ignoring the carbon fiber influences (Zhang and Chou, 2006; Nickel et al., 2001; Zhang and Chou, 2008; Li et al., 2002; Nidagundi et al., 2015b; Al Rashid and Koç, 2021; Liu and Shapiro, 2016; Kulkarni and Dutta, 1999; Magalhães, 2013). Of these models, the Classical Laminate Theory (CLT) and the micromechanical model approach by Finite Element Analysis (FEA) have been the most applied.

### 5.1. Classical Laminate Theory modeling of AM fabricated CFRP composites

According to Shokrieh and Shahri (Shokrieh and Kamali Shahri, 2021), CLT makes it possible to evaluate complex interactions between composite laminates where it can be used to predict displacement, strains, and curvatures that develop as the laminates are mechanically or thermally loaded. Li et al. (2002) presented one of the earliest constitutive models which utilized CLT to simulate the effective modulus of the AM fabricated CFRP composite. They applied experimental results of the matrix material stiffness for 0° (axial), 90° (transverse), and 45° raster angles to simulate the theoretical modulus for the composite, presenting SEM images of the sample cross-sections and their appending interlayer porosity volumes to predict the mechanical properties based on an empirical model. However, their model did not touch on carbon fiber effects, making it less relevant. A similar approach was seen with Magalhaes et al. (Magalhães, 2013) which only discussed the matrix interactions with no reference to the carbon fiber effect as reported in the many other CLT model investigations (Nidagundi et al., 2015b; Al Rashid and Koç, 2021; Liu and Shapiro, 2016; Kulkarni and Dutta, 1999; Brenken et al., 2019).

According to Cuan-Urquizo et al. (2019), applying the CLT to determine the effective modulus of AM fabricated plastics and their composites requires a more accurate specification of Young's modulus of the AM layers in both axial and transverse axes. This is only possible with the extent of interlayer connections and porosities between the individual print beads determined, which for the most part is still a challenge. More developments to establish these interactions more accurately in AM parts will offer the CLT approach more relevance in predicting mechanical performance parts fabricated with the composite and process.

Fig. 19 illustrates the fiber and matrix interaction within AM fabricated CFRP composites and shows a schematic of the representative volume element (RVE) used to model the AM parts using micro-mechanical evaluation.

### 5.2. Micromechanics approach by Finite Element Analysis

The fundamental micromechanics computation of the fabricated structure using FEA allows for the derivation of analytical expressions from which effective Young's modulus and effective shear modulus can be calculated from the structure-property relationship. The approach provides some edge over the limitations of the CLT. In this case, the composite is examined as repeating unit cells in a representative volume element (RVE), allowing for the simulation of Young's modulus and porosity volumes into the RVE, and convergence analyses are subsequently conducted to validate the consistency of modeled mechanical properties. The FEA approach is not without its limitations since certain features such as perfectly bond print beads, negligible interface layers,

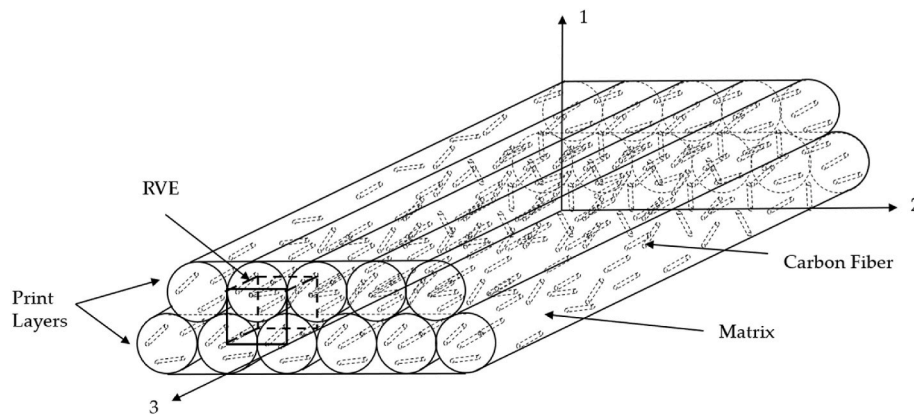


Fig. 19. Illustration of print layers of AM fabricated CFRP composite laminates including the schematics of representative volume element (RVE) used in micro-mechanical models.

Young's modulus consistency across an axis, etc. need to be assumed, but may not necessarily be correct.

Three different approaches to the FEA model are found in the literature which includes the microscopic, macroscopic, and mixed-modeling technique. The microscopic approach simulates the microstructure explicitly with as much resemblance to the fabricated structure as possible (Wendt et al., 2017; Somireddy and Czekanski, 2017) at the expense of high computational time and cost. The macroscopic approach on the other hand models the fabricated parts as solid continua with some homogenized effective properties (Domingo-Espin et al., 2015), while the mixed modeling approach combines the microscopic and macroscopic modeling as a stack of series of macroscopically modeled discrete reinforced layers, in which each layer has orthotropic properties.

Just like for the CLT, most of the micromechanical models reported only investigated pure plastics with limited research on the mechanical performance of AM fabricated CFRP composites (Zhang and Chou, 2008; Liu and Shapiro, 2016; Huang and Singamneni, 2013, 2014; Crococolo et al., 2013; Favaloro et al., 2017) and still need development as these can serve as powerful tools to develop the growth of AM fabricated CFRP applications.

## 6. Concluding remarks

In this review, short fiber composite design factors necessary to improve the current situation of the mechanical performance in the AM fabrication of CFRP composites have been discussed. The following are the observations and theoretical concepts that can be appropriated to further advance the applications of the material:

1. The AM fabrication of CFRP composites offers huge potential due to the low strength-to-weight and strength-to-modulus offering of CFRP composites combined with the many advantages of the AM fabrication technique.
2. There still exists some mechanical properties issues such as material porosities, fiber-matrix interfacial adhesion, anisotropy, etc. for parts fabricated through the method which can be improved with the proper appropriation of materials design factors as provided in this review.
3. Material designs should take into consideration the ease of processibility of composite constituent composition since material processing could influence the mechanical performance of the composites.
4. The presence of interlayer voids in the composite from the layering process of AM fabrication makes the materials more susceptible to environmental influences, which can be improved by appropriating materials design.

5. Further developments in materials and process design will be needed to improve the current situation of AM fabrication of CFRP composites to improve mechanical performance for the existing and the vast opportunities that lie ahead.
6. There is still a huge knowledge gap in the simulation modeling of the mechanical performance of AM fabricated CFRP composites which need to be developed to foster the growth of the composite.

## Declaration of competing interest

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.

## Data availability

Data will be made available on request.

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