3D Printing of Continuous Natural Fibre Reinforced Biocomposites for Structural Applications

A. Mugeshwaran, Nalini Ranganathan, R. Joseph Bensingh, and Sanjay K. Nayak

Abstract In last few years, there is a constant demand from several industrial fields for using light-weight components to exhibit high mechanical performance. This demand is met by employing composite materials, especially the fiber reinforced polymer composites. Compare to conventional process shape complexity, infill density, and manufacturing lead times are no longer barriers with additive manufacturing. Therefore, the fabrication of light weight fiber reinforced polymeric composites using additive manufacturing remains the protagonist. In this chapter different types of fiber reinforce composites developed using different additive manufacturing technique are classified based on the length of fiber, aspect ratio, orientation and performance i.e. short, long and continuous fiber reinforced composites. Further the type of continuous natural fiber reinforced composites that are developed using various additive manufacturing technique such as fused deposition modelling (FDM) or Fused Filament Fabrication (FFF), stereolithography (SLA), selective laser sintering (SLS), selective laser melting (SLM), and Direct ink writing (DIW) were reviewed and discussed. In addition, different materials, drawbacks, and strengths associated with different additive manufacturing processes were detailed. Few examples were also presented in the chapter were the 3D Printed structural components has its own real time application in various manufacturing sectors namely automotive, aerospace and aviation.

Keywords Additive manufacturing · Natural fiber · Reinforcement · Polymeric matrix

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1 Introduction

Continuous Natural Fiber (CNF) and their biocomposite are being widely used in almost all the structural application due to its massive natural characteristics like lower density, lower cost, and they are renewable and biodegradable with the environment (Pickering et al. 2016). “Green” composites, based on natural fibers derived from plants and biodegradable resins, are in high demand to meet regulatory requirements of recyclability. Several researchers confirmed that the continues natural fiber composites shows significant improvements in stiffness, strength and toughness of composites over fiber reinforced composites, which tends them to utilize in high-tech applications in various engineering sectors such as automotive, aerospace, defense etc. However, properties of the CNF composites depend on various factors matrix intrinsic properties, fiber properties, fiber volume fraction, and interfacial bond strength between the fiber and the matrix, fiber aspect ratio as well as fiber orientation (Ranganathan et al. 2015).

In past decades, the CNF based composite have been developed through various manufacturing processes such as vacuum forming (Delvasto et al. 2010), extrusion (Grande and Torres 2005), injection moulding (Gao and Mäder 2006), and compression (Sreekumar et al. 2007). These conventional fabrication methods for composites require expensive facilities and equipment, such as autoclaves and complex rigid molds, hindering the wide applications. They were also constrained and limited by complexity structure, high mould cost, orientation, and alignment of fibers (Mohanty et al. 2004; Goodship et al. 2015; Faruk et al. 2012). In order to overcome the limitation, the novel method i.e. the 3D Printing/Additive technology for continuous fiber reinforced composites is being developed and studied by many researchers. 3D printing techniques are one of the most promising processes which enhance the light-weight and low-cost processing approaches for reinforced biocomposites when compared to several other composite fabrication methods (Compton and Lewis 2014).The research revolving additive manufacturing to develop structural components using continuous natural fiber composites is in its young stage and burgeoning exponentially throughout the globe. Several additive manufacturing techniques are being involved for the development of structural components which includes fused deposition modeling (FDM) or Fused Filament Fabrication (FFF), stereolithography (SLA), binder jetting (BJ), multi-jet fusion (MJF), selective laser sintering (SLS), selective laser melting (SLM), electron beam melting (EBM), electron beam additive manufacturing(EBAM), big area additive manufacturing (BAAM) (Dugbenoo et al. 2018); Out of which the FDM/FFF, SLA and SLS technologies are being used for developing polymeric composites.

This chapter presents a detailed review of types of natural fibers and 3D printing techniques that are adopted by the researchers for the development of additive manufactured CNF-reinforced biocomposite parts. Additionally, based on the current technological significance and the limitation have been defined.

2 Classification of Short, Long and Continuous Natural Fibers Biocomposites

The natural fibers of cellulose/lignocelluloses fibers are derived from the plant, such as kenaf, hemp, jute, flax, ramie, nettle, pineapple leaf, sisal, date palm, cotton fire, coconut fibers, kapok, bamboo (Al-Oqla and Salit 2017; AL-Oqla and Sapuan 2018; TG YG et al. 2019). On the other hand, wool, silk, hair, and feathers are extracted from the largely consisted proteins of animals and some of the other waste of renewable resources (Saheb and Jog 1999). The natural fibers are classified as short, long and continuous fibers by its aspect ratio (fibers length and diameter) fibers, these fibers are reinforced with polymeric matrix and form biocomposites. Biocomposite materials are defined as composite materials in which at least one of the constituents is derived from natural resources. It is broadly categorized into three types.

i. Reinforcement of synthetic fiber such as carbon fiber, glass fiber, kevlar fiber into bio-matrix such as PLA, PHA, PHB etc.
ii. Reinforcement of natural fibers such as jute, coir, sisal fiber etc. in synthetic/petroleum-derived polymers such as polyethylene (PE), polypropylene (PP) etc.
iii. Reinforcement of bio/natural fibers such as jute, coir, sisal fiber etc. into bio-matrix such as PLA, PHA, PHB etc.

In particular, the natural fiber biocomposites fall under the category of (2) and (3), i.e. the biocomposite materials made from the combination of natural fibers reinforced petroleum-derived polymers or with bio-polymers. Globally, the cellulosic fibers are becoming very interesting for bio-based composite development as they possess advantages with their mechanical properties, low density, environmental benefits, renewability, and economic (Ngo 2018). The bio-based composites are majorly classified as short, Long and continuous fiber composites. In case of short fiber composites, the average size of fiber tends to range below 0.5 inches, whereas in long fiber composites the fibers are normally 0.5, 1.0, or 2.0 inches long (Collett and Campbell 2004) in both the cases namely the short fiber composites and long fiber composites, the fibers are discontinuous. But, in continuous fiber composites the fibers tend to be continuous owing its unique properties. Continuous-fiber composites normally have a preferred orientation, while discontinuous fibers generally have a random orientation. Therefore, the continuous fiber composites are used where higher strength and stiffness are required (but at a higher cost), and discontinuous-fiber composites are used where cost is the main driver and strength and stiffness are less important (Chevali et al. 2010); The benefits and its properties of each category are described in Table 1.

Table 1 The benefits and its properties of each category

Description	Short Fiber Composites	Long Fiber Composites	Continuous Fiber Composites
Processing	Can be processed by many methods	Cannot be processed by many methods	Cannot be processed by many methods
	Compression molding, Extrusion, Hand lay-up, Injection molding, Resin transfer molding, and Sheet molding, Additive Manufacturing	Compression molding, Direct long fiber extrusion technique (D-LFT), Hand lay-up, Resin transfer molding, and Sheet molding	Compression molding, Filament winding, Hand lay-up, Pultrusion, Resin transfer molding and Additive manufacturing
Length of fiber	Lesser than 0.5 Inches	0.5–2.0 Inches	Larger than 2.0 Inches
Aspect ratio	Lower aspect ratio (<100), provide moderate strength, stiffness or creep under loads and with lower endurance compare to LFC or CFC	Higher aspect ratio about 200–3000 (Chevali et al. 2010) provides long fiber composites with increased strength, stiffness or creep under loads and higher fatigue endurance with minimal compression	Longer aspect ratio (Preferable)
Type and orientation	Discontinuous (More randomly dispersed and oriented across the part)	Discontinuous (Not more randomly dispersed and oriented across the part)	Continues (preferred regular orientation arranged across part)
Mechanical performance	They offer lower mechanical performance compare to long and continuous fiber composites, hence could be used forsemi-structural components	They offer lower mechanical performance compare to continuous fiber composite, but higher properties compare to short fiber composites	They provide extreme mechanical performance, thereby indenting to utilize the composite for metal replacements or for structural applications
	Short fiber reinforced polymers were developed to fill the property gap between continuous fiber laminates used as primary structures by the aircraft and aerospace industry and unreinforced polymers used largely in non-load bearing application	The long fiber composite bridges the gap between short and continuous fiber composites, offers better mechanical properties than SFC, but retain their processing conditions (Phelps 2009)	Continuous-fiber composites are normally laminated materials in which the individual layers, plies, or laminae are oriented in directions that will enhance the strength in the primary load direction (Campbell 2010)

3 Continuous Natural Fibers Composites by Additive Manufacturing

Natural fibers are becoming a preferred alternative in reinforced polymer composites, due to the intensive properties such as renewability and biodegradability; in addition, they also possess excellent tensile properties.

Several studies have shown that short (discrete or chopped) fibers extruded as one filament successfully integrated with matrix material are applicable in several AM techniques (Campbell 2010), whereas printing of fibers and matrix materials continuously layer-by-layer have presented successful research. The continuous fiber 3D Printing were started since 2014. The continuous fiber composites in 3D printing may be defined as spool of fibers used to embed very long strands of fiber into parts as they are printed. These fibers in 3D printing provide substantially more strength and stiffness than other short/particulate fiber composites. Tian et al., study has reported that, the use of biocomposite filaments reduces the material cost and environmental impact (Tian et al. 2017). Several studies reported that the continuous fibers are available in different forms such as fiber bundle, yarn, hessian cloth or mat, in case of additive manufacturing technology the continuous fibers are used as fiber bundle with slight twisting. Only limited research studies have been done in 3D printing of the natural fiber-reinforced composite received from fruits and plants. Table 2 reveals about the 3D printed continuous fiber reinforced composites. The development of composite using additive manufacturing involves implementation of fibers after the nozzle directly into the print job using the fibers and matrix. In this process, the fibers are implemented in two ways one is incorporation of fiber 'inside the nozzle' or placing 'after the nozzle'. Generally, the following disadvantages are present in implementing the continuous fiber reinforced composites using 3D printing process.

i. Difficult process controls due to the fiber infiltration parallel to printing and the handling of the fibers.
ii. Possess challenges with resolution, surface finish and in controlling the interfacial (or) thermal bonding between the fiber and matrix.
iii. The inferior mechanical properties and anisotropic behavior.

A study by (Tekinalp et al. 2014). highlighted the challenges associated with 3D printing fibre reinforced composites and evaluated the load bearing potential of composite parts made from carbon fibre and ABS resin feedstock. On the other hand, they have various benefits such as freedom of design, mass customization, waste minimization and the ability to manufacture complex structures, as well as fast prototyping etc.

Table 2 Classification of composite materials used in additive manufacturing technique

SI. No.	Short natural fiber	Polymeric matrix	Additive manufacturing technique	Author/References
1	Sugar cane baggasse	Polyethylene (PE) Polypropylene (PP) Acrylonitrile butadiene styrene (ABS) Polylactic acid (PLA)	Fused deposition modeling (FDM)	Navarrete et al. (2018)
2	Hemp fiber	Polylactic acid (PLA)	Fused deposition modeling (FDM)	Stoof et al. (2017)
3	Recycled cotton fibers	VisiJet-SL (Clear)	Stereolithography (SLA)	Liu et al. (2017)
4	Green tow flax fibers	Geopolymer matrix	Injection molding made to simulate 3D printing using a vibrating table	Korniejenko et al. (2019)
5	Hemp fiber	Recycled polypropylene	Fused deposition modeling (FDM)	Milosevic et al. (2017)
6	Hemp fiber	Polylactic acid (PLA)	Fused deposition modeling (FDM)	Stoof et al. (2017)
SI. No	**Continuous natural fiber**	**Polymeric matrix**	**Additive manufacturing technique**	**Author/References**
1	Flax fiber	Polylactic acid (PLA)	Fused deposition modeling (FDM)/Fused filament fabrication (FFF)	Duigou et al. (2019)
2	Twisted jute fiber	Polylactic acid (PLA)	Fused deposition modeling (FDM)	Matsuzaki et al. (2016)
3	Cotton fibers	Amylopectin	Direct ink writing (DIW)	Jiang et al. (2019)

4 Additive Manufacturing: Continuous Natural Fiber Reinforced Composites

Various 3D printing techniques are being used for the development of continuous reinforcement biocomposites such as fused deposition method (FDM), stereolithography (SLA), selective laser sintering (SLS) and binder jet (3DP). For structural applications, beyond these techniques, FDM, SLS, and 3DP are largely used for the fabrication of CNFRBC (Ngo et al. 2018). The most widely used 3D printing techniques and manufacturing methods and challenges in the 3D printing of the biocomposite are discussed below.

4.1 Fused Deposition Modeling (FDM) in the Natural Fibers-Reinforced Composite

FDM is the nozzle extrusion-based technique where the feedstock material is mounted in the form of spool. Figure 1a, b, shows the schematic representation of Fused Deposition Method (FDM) for developing the continuous natural fiber composite.

Figure 1a, shows that the feedstock material is fed through the printer nozzle and filaments are extruded from a nozzle by the application of controlled heat (Turner et al. 2014) and (Ranganathan et al. 2019). The extrudate will be delivered through the nozzle under controlled melting temperature to build the object by selective depositing; meanwhile, continuous natural fiber is fed from the fiber supply coil and goes through the inner bore of the extrusion head to the nozzle.

Figure 1b, shows that the continuous fiber is immersed in the liquid matrix which is kept below the spool of fiber. In both the cases the, continuous natural fiber is infiltrated and coated by the molten thermoplastic polymer inside the nozzle, and the impregnated composites can be extruded out from the egress of the nozzle. When the extruded material reaches the surface of the part, it solidifies rapidly and adheres to the previous layer so that the fiber can be pulled out continually by the foregoing fiber inside the part. On the other hand, the extrusion head, which is connected to X–Y motion mechanism, can and generate single layer of the part. After one layer is completed, the building platform placed on a lift mechanism moves in the Z-axis direction by an increase equal to the layer thickness. The process is repeated until the part is finished (Murphy and Collins 2018) and (Yang et al. 2017). The schematic representation of FDM process is shown in Fig. 1. FDM is the most widely used method for printing continuous natural fiber reinforced composites.

Various researchers are utilizing the FDM/FFF process for developing continuous natural fibers-reinforced biocomposite for the structural application. The efforts

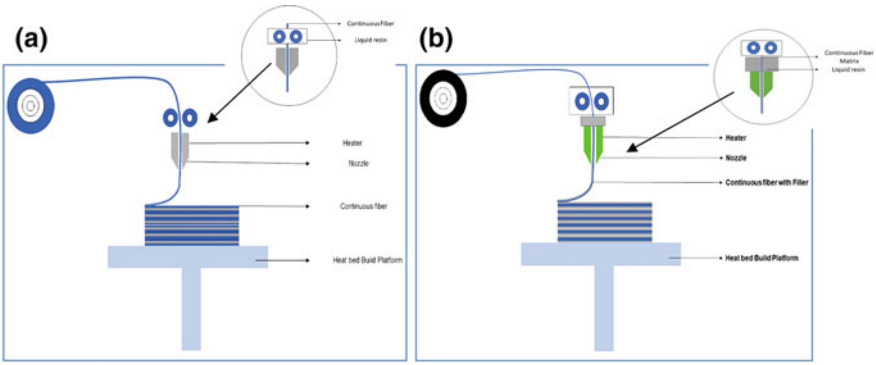

Fig. 1 **a** Fused Deposition method in the continuous natural fiber composite and **b** Fused Deposition method in the continuous natural fiber composite immersed in the liquid matrix

have begun to overcome the geometrical limitation and complexity of the product. Le Duigou et al. developed continuous flax fibers (FF) reinforced PLA composites using FDM process (Duigou et al. 2019). In their study, they have compared and reported the mechanical properties and failure mechanism of FDM printed cFF/PLA part and thermo-compressed continuous synthetic fiber reinforced polymer composites. Similarly, (Duigou et al. 2016) has also investigated the FDM processed continuous wood fiber reinforced thermoplastic composites. The author studied the fiber's anisotropic behavior by printing in different orientations (0 or 90°). Matsuzaki et al. have also utilized the FDM Process to manufacturing continuous natural fibers reinforced PLA composites (Matsuzaki et al. 2016). In their study, they have examined and compared the mechanical properties of the continuously reinforced biocomposite with commercial 3D printing polymer composites.

4.2 Selective Laser Sintering (SLS) in Continuous Natural Fibers-Reinforced Composite

SLS process operated under the principle of laser sintering i.e. high-intensity laser beam such as CO_2/Nd: YAG are used to sinter the powder particles, this process is also known as powder bed fusion, shown in Fig. 2. Reinforcements for SLS-fabricated FRPC are mostly found in the form of discontinuous fibers; synthetic fibers namely carbon fiber, glass fiber or aramid fibers are commonly used. In this process, the raw material or the polymer matrix will be in powder form i.e. polyamide (PA) spherical particles and synthetic fiber will be in uniform diameter around 10 µm. Besides PA, some other bio-polymer composites for instance polycapralactone (PCL)/hydroxyapatite (HA), PEEK/HA and polyethylene (PE)/HA were using SLS (Wang et al. 2017). In this technique the laser-scanning speed, the intensity of the source and layer thickness; needs to be optimized for printing the biocomposite.

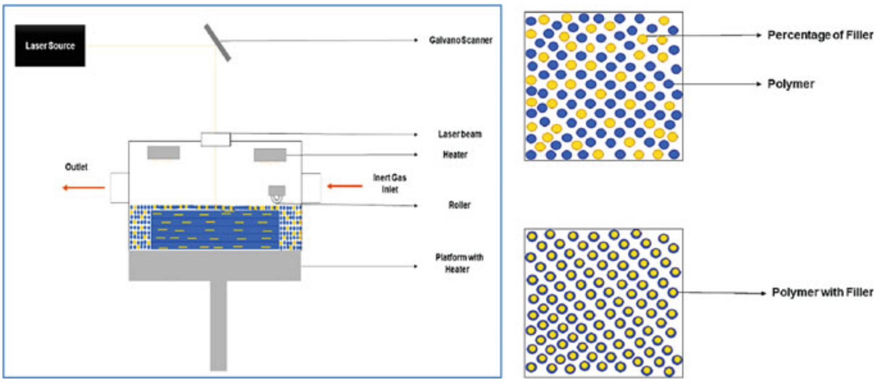

Fig. 2 Selective laser sintering (SLS) for natural fiber reinforced composites

In addition, the build chamber has to be maintained with an inert atmosphere to create the required oxygen-free environment (Goh et al. 2019) and (Goodridge et al. 2011); so that the powdery materials used for printing will be free of moisture, thereby the voids or defects in the printed parts can be avoided.

Saroia et al. employed laser sintering process to manufacture carbon nanofibre reinforced polyamide-12 composite. The mixture of carbon fibers and polyamide was prepared by melt mixing, and the characterization of laser sintered parts was studied (Saroia et al. 2020). Salmoria et al. reported that, the 3D printing of continuous fiber reinforcement composite using SLS is a real challenge (Salmoria et al. 2011). The researchers are working for the development of a proper and standard paradigm for 3D printing of the continuous fiber polymer composite.

Recently, few researchers have utilized the different natural reinforced biocomposite to develop a structural component. They reported that, by optimizing the process parameters the SLS process could be adapted for developing natural fiber reinforced composites. Zhao et al. has optimized the process parameters of SLS, to improve the mechanical properties of the developed biocomposite (Zhao et al. 2017). Zeng et al. developed SLS printed biocomposite components using bamboo flour and co-polyester (BFCP) for various industrial applications (Zeng et al. 2013). Likewise, Simposon et al. developed biocomposites using wood powder as reinforcement in polymeric matrix (Simpson 2018). The powder material processing for the SLS process is quite expensive than other printing techniques. Further, the difficulties persist with material manufacturing and controlling the fiber orientation, interfacial adhesion etc. However, the developed biocomposites found environmentally friendly, non-toxic, and biodegradable materials with low cost.

4.3 Stereolithography (SLA) in Continuous Natural Fibers Composite

Vat photopolymerization selectively cures photopolymer in a vat using ultraviolet (UV) light may be referred as Stereolithography (SLA). In otherwords, the stereolithography (SLA) is a commonly used vat photo-polymerization technique to print FRPC. The Process is also known as direct light processing (DLP). In the SLA process, the low watt laser Nd-Yag laser/UV source is used to cure the liquid photopolymer resin. The build platform is submerged into the liquid photopolymers and is maintained at one-layer height. Then a UV/laser creates the next layer by selectively curing and solidifying the photopolymer resin. This process is called photopolymerization (Feng et al. 2017) and (Guillaume et al. 2017a). Most of the existing SLA systems implement a bottom up approach, scanning each layer, then subsequently moving the base downwards and wiping an additional layer of polymer on top before again photo-polymerizing, represented in Fig. 3. As shown in the figure the platform of the SLA system moves downwards to an incremental amount, by depositing layer by layer. The wipers smooth the top of the deposited resin before the laser

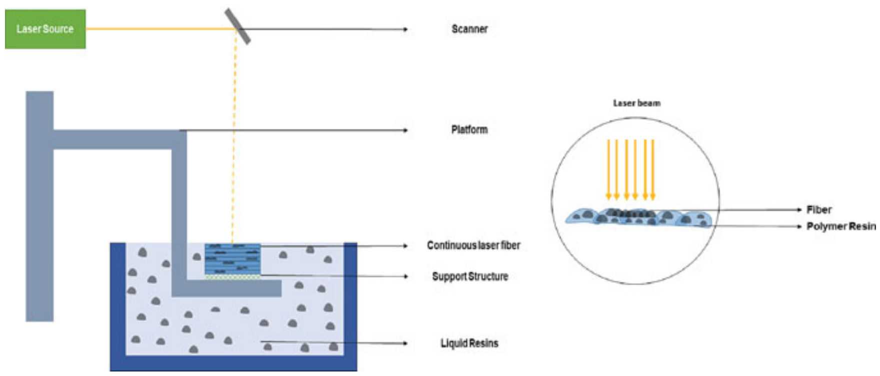

Fig. 3 Stereolithography (SLS) process

cures the corresponding layer patter. The woven mat of fibers are incorporated on bottom layer of solid resin, and then coated with resin. Vat photopolymerization can create composite parts from chopped, woven, or continuous fibers. The creation of composite parts consists of submerging the reinforcing fibers in the UV curable resin, then curing the resin. This technique has the ability to print the higher resolution component. However, the availability of the commercial material is very limited for this process. Research has been carried out on continuous FRPC using glass and carbon fiber bundles and carbon fiber mats as reinforcement (Karalekas and Antoniou 2004). In this continuous FRPC development using SLA, the manual lying or incorporating fiber lying mechanisms (Goodridge et al. 2011).

Recently many researchers are attempting to develop new continuous biocomposites using natural fibers in the SLA process, due to the easy availability of liquid resin. Guillaume et al. a developed poly (trimethylene carbonate) (PTMC) and nano-hydroxyapatite (HA) based cytocompatible 3D porous structure using SLA manufacturing process (Guillaume et al. 2017b). Similarly, Garg et al. developed SLA processed the continuous fiber reinforced composite using Accura60 Resin and carbon fiber (Garg et al. 2001). However, the vat photo-polymerization process has several limitations such as (1) the mechanics of fiber addition, (2) fiber settling, (3) formation of bubbles (voids), (4) increase in resin viscosity (causes poormixing, leading to poor interfacial properties, and (5) laser diffraction and laser energy.

4.4 Direct Ink Writing Technology in Developing Continuous Fiber Reinforced Composite

Direct ink writing (DIW) technology is used to develop continuous natural fiber reinforced thermoplastic composites. In the direct ink writing technology four syringes containing a different composition of magnetic material are used to produce inks with different formulations, thereby the multi-material prints are attained. To produce the composite inks, SR powder (amylopectin), water, and cotton fibers, without any other chemical or binders were used by (Jiang et al. 2019). In this process the components are added in the proper ratio, mixed well and then subjected to a thermal treatment (causing gelatinization), thus forming the 3D composite pattern. SR composite with 10 wt% cotton fibers from a 400 μm nozzle and (inset) a cellular structure printed via this process were developed and studied by the author. Similarly, the direct ink writing (DIW) system was also used by Sullivan et al. In their study they have used silver nanoparticle ink with a custom-built cell for producing an electrophoretic system to enable deposition of conductive electrodes with controlled thickness with homogeneous mixing of thermite composites (Sullivan et al. 2019).

5 3D Printed Components in Structural Application

The 3D-printing technology emerged to address the rapid prototyping needs of the industry. The technology has outgrown its intended purpose and has its potential for structural applications. Various manufacturing industries have been leveraging the ability of 3D printers to lighten structures by developing lattice structures using fiber reinforced composites. These components are used in various for sectors such as automotive, aerospace and aviation. For example:

i. M/s. AMFG Autonomous Manufacturing Company has produced 3D-printed spacer panels and installed on commercial A320 aircraft.
ii. M/s. Dutch Architecture Company used 3DP to design facades integrated with solar panels, where the angle of the solar panel were optimized automatically based on the location.
iii. M/s. Arevo technology, used continuous carbon fiber/polyetheretherketone (PEEK) for Arevo frame: battery-assisted "ebike".
iv. M/s. Apis Cor is a San-Francisco-based start-up company has used mobile construction 3D printers that print structural components by adopting additive manufacturing, the construction industry had several advantages like better creativity, design flexibility, less material waste, lower carbon footprint, and more robust structures.

6 Conclusion

In this chapter, the development of CNF and their biocomposite for structural application by using the additive manufacturing process are discussed. The chapter clearly demonstrates the types of natural fibers biocomposites namely short, long and continuous by its aspect ratio (fibers length and diameter) fibers and the different types of fibers (Syntenic fiber, natural fiber and bio added natural fiber) are used as a reinforced with polymeric matrix to form biocomposites. Further the type of continuous natural fiber reinforced composites that are developed using different additive manufacturing technique was explored. In addition, the chapter also addressed the major advantages and difficulties involved in the printed natural fiber composite by using the various additive manufacturing process the such as fused deposition modelling (FDM) or Fused Filament Fabrication (FFF), stereolithography (SLA), selective laser sintering (SLS), selective laser melting (SLM), and Direct ink writing (DIW). The ability to develop the structural component using natural fiber reinforced composites by using the additive manufacturing process was also discussed detailed.

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