

3D Printing to Produce BioComposite

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Current environmental concerns have led to a search of more environmentally friendly manufacturing methods, thus, natural fibers have gained attention in the 3D printing industry to be used as biofilters along with thermoplastics. The utilization of natural fibers is very convenient as they are easily available, cost-effective, eco-friendly, and biodegradable. Using natural fibers rather than synthetic fibers in the production of the 3D printing filaments will reduce gas emissions associated with the production of the synthetic fibers that would add to the current pollution problem. As a matter of fact, natural fibers have a reinforcing effect on plastics. This review analyzes how the properties of the different types of polymers vary when natural fibers processed to produce filaments for 3D Printing are added. The results of using natural fibers for 3D Printing are presented in this study and appeared to be satisfactory, while a limited number of studies have reported some issues.

Keywords: natural fibers ; biofilters ; FDM ; 3D Printing ; mechanical properties

1. Introduction

Three-dimensional (3D) printing is manufacturing a 3D object from a computer-aided design model by sequential addition of materials added one layer at a time. It is also named as additive manufacturing (AM)^[1]. The first 3D printing method was patented in 1986 by Charles W. Hull^[2], and it was then known as stereolithography. Earlier in the 1990s, 3D printing techniques were used only for the creation of functional or visual prototypes and were more often referred to as rapid prototyping^[3]. Currently, the comprehensive 3D printing market is growing in a fast-paced manner and is expected to expand even more in the next few years. 3D printing is being applied innovatively in multiple areas, including biotechnology, energy, medical devices, and many more^{[4][5][6][7][8]}.

The reason behind this fast-paced growth is that objects can be designed digitally and manufactured precisely in a layer-by-layer manner with no molds, dies, or lithographic masks^{[9][10]}. The technology is now being rapidly adopted in both industrial and household settings, because of its many advantages, such as suitability for small scale production, effortless part acquisition, limited waste, energy efficiency, and no need for expensive tools^[11]. 3D printing offers automation and reproducibility to a great level. It allows the uninterrupted production of structures that can only be produced with much more effort using traditional subtractive manufacturing procedures^{[12][12]}. 3D printing has a potential for providing prototypes, customer-specific designs, high structural complexity, and rapid on-demand fabrication of small production lines at affordable rates^[13]. Therefore, it is regarded as the next revolution in manufacturing.

With 3D printing, it is possible to fabricate objects of complicated shapes and thickness, which may be inaccessible to the standard polymer manufacturing techniques^{[14][15][16]}. The printing techniques can broadly be divided into four categories: (1) extrusion-based methods, such as fused deposition modeling (FDM) where layers of material are fused in a pattern to create a printed object, (2) particle fusion-based methods, such as selective laser sintering which uses a laser to sinter powdered material, aiming the laser automatically at points in space defined by a 3D CAD model, binding the material together to create a solid structure, (3) stereolithography (SLA), the production parts are printed in a layer by layer fashion using photochemical processes by which chemical reaction causes the formation of polymers, this is mostly used for the production of prototypes and patterns, (4) inkjet printing which prints by depositing liquid materials or solid suspensions, (5) digital light process (DLP) is similar to SLA as both cure liquid resin using light. The primary difference between the two technologies is that DLP uses a digital light projector screen. In contrast, the SLA uses a UV laser, (6) multi jet fusion (MJF) which builds functional parts from powder instead of using a laser to sinter the powder MJF uses an inkjet array to apply fusing agents to the bed of powder, and (7) electron beam melting (EBM), is a metal 3D printing technology that uses an electron beam controlled by electromagnetic coils to melt the metal powder^{[13][17][18]}. Although some materials can be used for printing using different 3D technologies, the compositions of the printable material vary considerably^[13]. Among the various 3D printing techniques, one of the most popular techniques is the fused filament fabrication (FFF) or extrusion-based method because it is simple, cost-effective, and does not require hazardous solvents or glues^[19]. Also, the printing apparatus is small in size that can be accommodated on a tabletop^[20]. In this technique, an object is built by

selectively depositing melted material layer-by-layer along a pre-determined path. The materials used are thermoplastic polymers that come in a filament form. The following printing parameters are used for the extrusion-based technique: (1) extruder-related (nozzle diameter and filament width), (2) process-related (temperatures and speed of printing), and (3) structure-related (layer thickness and infill geometry).

Thermoplastics are being widely used in extrusion-based techniques, since they have a low impact on the environment, as they are recyclable and are available in a great variety of materials. However, polymers, such as polylactic acid (PLA) and acrylonitrile butadiene styrene (ABS) or nylon can be hazardous and not adequately environment-friendly, as volatile organic compounds and ultrafine aerosols may be generated during 3D printing [21]. However, less toxic 3D printing materials are forthcoming. To reduce safety risks and unpleasant odors associated with synthetic polymers, the industry is now more inclined toward natural polymers, which are environmentally friendly and renewable [22]. Research is now more focused on developing printable biopolymer composites with improved performance. Environmentally friendly and inexhaustible biobased materials are now being investigated. This includes cellulose derived from plants, biomass from marine, wood, and agricultural residuals, and other abundant renewable feedstocks, which are potential alternatives to fossil resources [23][24][25].

The factors affecting the cost of the printed objects are the cost of materials being used and the time taken to print. Some filaments are expensive as compared to other filaments and printers may pose restrictions in their usage. The cost of filaments can be reduced by the addition of economical filler materials, which may improve the flexural stiffness, mechanical properties, and stability after solidification. However, to achieve these benefits, a suitable chemical treatment to the fibers may be required and a suitable coupling agent for material formation may also be needed. Moreover, the use of fillers will assist in mitigating the environmental impact.

Natural fibers have recently been widely used as additives in extrusion-based filaments [26]. To produce a good-value natural fiber thermoplastic filament, the biofilter should be mixed with a polymeric matrix. This can be achieved through compounding using a co-rotating twin-screw extruder, which allows dispersive and distributive mixing [27]. The latter homogenizes additives evenly within the matrix, while the former eliminates additive clusters and is particularly relevant for natural fibers, as they tend to attract one another. Mechanical performance is improved by the chemical treatment of fibers, and it positively affects the load transfer capability of the biofilter-polymer interface [11]. Even though the use of fiber reinforcement appears feasible and promising, it has various challenges that need to be overcome, such as the effect of fibers on the resolution, agglomerate formation, heterogeneous composite formation, blockage of printer heads, non-adhesion, and increased curing times [5].

The polymer matrices can be categorized as either biodegradable or non-biodegradable, or, based on their origin, as a virgin, recycled, or hybrid, respectively. Few reviews have been published. The review of Wang et al. [26] summarizes different materials used for 3D printing, their properties, and application in fields of biomedical, electronics, and aerospace engineering. Another review by Mazzanti et al. [11] reviews the mechanical properties of 3D printed objects of polymers containing natural fillers. This review aims to cover recent advancements in the FDM process of polymers with AM techniques to inculcate natural fibers as fillers, their fabrication strategies and parameters, and their effects on the mechanical properties of the resultant 3D-printed parts.

2. 3D Using Natural Fibers

When using natural fibers, the processing needs to be done diligently, or else it may lead to low-quality filament or poor outcomes. These fibers must be dried carefully in the initial phase even before compounding as it is very important to reduce the water content in them, which, if not done correctly, could lead to hydrolytic degradation. The temperature used should be monitored carefully to avoid thermo-oxidative degradation. If the material viscosity increases, it is important to have high extrusion temperatures. Still, high extrusion temperature also reduces the permanence time of the melt inside the heated chamber, which may prevent the degradation of the biofilters due to low heat conductivity in polymers. This needs to be handled by increasing the printing speed so that the permanence is reduced at very high temperatures.

Chemical treatments and toughening agents have proved useful for the improvement of tensile and flexural strengths. These agents may also tend to fill in the voids and cracks, reducing porosity and, thus, improving the strength. Some treatments are given before filament production to the fibers to improve their quality and make it easier to blend in with polymer for filament production.

It is also important to mention that nozzle type and size needs to be chosen with extra caution and understanding. A very narrow nozzle may lead to sieving of the filament during extrusion, which may lead to uneven material flow and may introduce defects in the printed sample. Similarly, a wide nozzle may tend to release more than the required amount of

melted matter on to the plate, making it difficult to shape precisely, thus resulting in a very deformed and irregular printed sample.

Sometimes researchers tend to design their own custom nozzles, to meet a particular requirement of their protocol. For instance, Jassmi et al.^[28] invented a compound nozzle for a cement 3D printer to produce thermally insulated composites, this nozzle can also be used with natural fibers to create insulation through the printing process.

Prior to printing, the filament quality must be assessed to check for any voids or cracks, and its composition should be cross-checked regarding the additives, their quantity, and distribution. For this assessment, various methods are used and scanning electron microscopy imaging is commonly used. This method can also be applied for verification and testing the accuracy of 3D printed samples.

The study by Liu et al.^[29] proposed a novel method of 3D printing—the free-hanging 3D printing method for manufacturing CRFTP lattices. This method is different from the conventional layer-by-layer approach and uses direct extrusion of the overhanging and undercut structures with the guided spatial movement of nozzles. The method produced a better truss surface and bearing capacity, which negated the requirement of a complex support structure.

However, this method is only suitable for continuous fiber material because the continuous fiber enables continuity and stiffness structure to the printed object^[30]. In another study^[31], TiO₂ was used with ABS and extruded filaments to expand the chemical capabilities of the 3D printed structures, which were developed through thermoplastic printers. Zhang et al.^[32] fabricated PLA with hydroxyapatite and compared the osteogenic and biodegradation property. Results showed that they had good osteogenic capability and biodegradation activity with no difference in inflammation reaction, showing the potential to be used in bone tissue engineering. Using 3D printing in bone tissue engineering with natural composites is gaining popularity. One study determined the practical setup of parameters to increase the properties of objects when using additives in powder form for tissue engineering^[33]. Another study^[34] created a cement-free 3D printed concrete by using desert sand, the cement was replaced by 10% silica fume and 30% fly ash. Along with a superplasticizer which was added in the range of 1 to 3%, by binder mass.

Even though some thermoplastics release hazardous gases when modified, when used as polymers with natural fibers as fillers, they tend to be less harmful to the environment. Non- biodegradable polymers like ABS can be recycled. Biodegradable polymers such as PLA can be safely degraded. The main advantage of using natural fibers as fillers is that the industrial waste or other discarded materials from factories can be put to efficient use in the creation of filaments, which in turn can be converted to newly printed objects. For instance,^[30] uses wood powder waste collected from the furniture industry to create their filament with PLA. Many studies used jute fiber, flax fiber, or sugarcane bagasse for producing filaments in combination with different polymer, which in turn is giving a way to judicious waste management. To further reduce the environmental impact, researches are being conducted to produce printable biopolymer composites. Natural hydrogels, based on collagen, gelatin, and keratin, are being used to prepare scaffolds, which may be beneficial for tissue engineering. Industrial wastewaters and cellulose-based material are being put to good use, they are getting recycled into bio-based polymers and benefiting our environment in the long run.

Despite the importance of previous research efforts, there is some limitations. One limitation is that the fiber structure information was incomplete in many of these studies and average particle dimensions were generally used. Precise details would have enabled better understanding and determination of their performance as fillers. Many chemical agents were used as toughening agents and compatibilizers, which also affected the properties of printed objects. When used as additives, they created a multi-phased structure that reduced the concentration of stress and absorbed energy on impact. Several issues arise during printing, such as increased viscosity and fiber-matrix interface issues. Moreover, it has been observed that there is a lack in the studies related to the impact of implementing different internal 3D printing structures on the mechanical properties^[35] of 3D printed fiber-reinforced composite as well as the influence of the open-source 3D printer^[36] on the quality, consistency and the process capability^[37] of the natural fiber 3D printed composite objects or using hybrid composite 3D printing technology enhanced with hard particles^[38].

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