REPORT

3D Printing of polymer composites: A short review

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Abstract
Composite or reinforced materials, especially in the class of polymers, are becoming prominent materials for the diversified range of engineering and scientific utilities because of their physical, chemical, mechanical, and structural excellences. Indeed, the main credit for the widespread acknowledgment of polymer matrix composites (PMCs) goes to the various types of natural and synthetic reinforcements, which resulted in good interfacial chemistries. However, the intrinsic challenges of traditional manufacturing technologies always presented restrictions in the development of application specific PMCs. Reportedly, with an evolution of three-dimensional printing (3DP) technologies, the applications of PMCs have been transformed as these enabled the production of near-net shapes with high degree of control over the design, reinforcements, and processing parametric while maintaining the waste associated at minimal level. However, the industrial benefits of 3DP technologies for still limited owing to the lack of awareness among the young professionals and industrialists. Moreover, the literature available in this regard is either too scattered making it tedious for the professionals to go through or limited to only fundamental concepts. Therefore, the concept of this review paper is to provide brief research-based insights of different 3DP technologies (namely, fused deposition modeling, selective laser sintering, stereolithography, laminated object manufacturing, and inkjet printing), material systems, applications, and the future paradigms of research as a short and precise document.

KEYWORDS
3D printing, applications, composite, polymer, processing, reinforcement

1 INTRODUCTION

Generally, the polymer systems have always attained the attention of manufacturers because of their unique characteristics such as ease of processing, light weight, low cost, long life, and often ductility.1 However, in comparison of the metals and ceramics, polymers offer significantly less modulus and strength because of which the unique features of the later were always outweighed.2,3 Many decades ago, researchers found a novel method of eliminating the challenges of polymeric systems through the reinforcement of secondary metallic, ceramic, or polymeric inclusions in the form of fibers, whiskers, platelets, or particles.4,5 The embedment of such inclusions in a host polymer matrix makes it a composite,6 commonly referred as polymer matrix composites (PMCs),7 which offer enhanced material properties that have been used for demanding engineering applications.8 Conventionally, PMCs were reinforced with microsized inclusions9; however, the recent paradigms have shifted this trend towards the use of nanoscale inclusions, with an average size...
ranging between 1 and 100 nm. It has been proved by various researchers that the inclusion of nanoscale improves the mechanical, metallurgical, electrical, thermal, and other desirable quality characteristics of the developed PMCs. This was the phase wherein the conventional applications of the PMCs were already established in automobile, aerospace, and structural domains, and bloom in the field of nanoscience and nanotechnology has encouraged the scientific community to start using PMCs in other important fields including computing, sensors, optical, marine, biomedical, and many other smart applications. Figure 1 illustrates the various industrial applications of three-dimensional printing (3DP)-based PMCs.

Indeed, the advancements in the aforementioned fields were primarily depended on the ability to synthesize nanoparticles of various materials, sizes, and shapes, as well as the processing routes, for instance, 3DP, which can assemble and position these, efficiently, into the complex architectures. Printing of polymer composites combines the matrix and reinforcements to achieve a system with more useful structural or functional properties nonattainable by any of the constituent alone. Generally, the use of particle, fiber, or nanomaterial reinforcements in polymeric matrices allows the fabrication of PMCs, which are characterized by high mechanical performance and excellent functionality. In today's scientific practices, the literature is advocating that carbon tubes, graphene, and graphite are the three most extensively practiced reinforcements for the fabrication of PMCs of high efficacy.

In the contemporary era, the rapid consumption of petroleum-based polymeric systems and noticeable polymeric waste associated with the manufacturing technologies have also received a lot of criticism, owing to which increased environmental awareness reached a new levels of practices with much emphasis on the employment of naturally driven polymeric-based materials in place of routine synthetic materials as well as to use efficient manufacturing routes to reduce the residual wastes. Natural resources, including oil, gas, potable water, clean air, fertile soil, rare metals, minerals, wood, and biodiversity, are essential for human survival and for keeping the global economy functioning. Meanwhile, a new class of reinforcements, based on the waste, has also come into practice. During the same, a new terminology “Green Composites” came into existence, which referred to the materials with benefits to the companies, natural environment, and end-customers. The most widely employed natural-organic fillers, wood flour and fibers, are easy and cost-effective to achieve from sawmill wastes and provide excellent thermo-mechanical properties.

Various categories of conventional manufacturing methods for PMCs usually use mold tooing made of metallic materials as well as nonmetallic materials, which are welcoming for the different types of reinforcements. However, except fibrous reinforcements, it is utmost difficult for the manufacturers to control the distribution or placements of the fillers within the polymeric architectures. Nonetheless the materials’ issue, tool fabrication itself consumes noticeable labor and machining leading to high costs and long lead times. Here, the use of 3DP technologies has demonstrated considerable cost and lead time reductions while providing numerous other advantages such as immense design freedom and rapid iteration, nearly regardless of part complexity. Furthermore, it also provides the opportunity to produce lightweight tooing, directly simplifying transportation and storage operations within manufacturing facilities. Advancements in the development of composite 3D printers motivated the growth in preblended materials with fillers such as nanoparticles, carbon nanotubes, fibers, and graphene in order to achieve unique characteristics and capabilities. Fiber reinforcement, in particular, appeared to be an attractive filler to improve the properties of polymers. However, one of the biggest challenges in developing practical applications with 3DP, using PMCs, is that, till
now, the duo has been used mostly for either producing conceptual prototypes or performing academic research. Such drawbacks restrict the wide industrial application of 3D-printed PMCs.\textsuperscript{28}

In Kumar and Kruth,\textsuperscript{43} it has been highlighted that since 2010, when the very first document in the area of 3DP of PMCs has been published by Olatunde Olakanmi et al,\textsuperscript{44} no significant breakthroughs have been presented. Further, they\textsuperscript{43} suggested that there should be simultaneous developments executed in terms of the both 3DP systems and PMCs, as their feedstock, in order to deliver distinct physicochemical properties into the resultant product. Surely, this will exhibit unique characteristics and capabilities.\textsuperscript{44} Till few years ago, 3DP techniques have suffered from underappreciated similarity to those of old-style composite materials, as both are integrally based on assembling a series of distinct layers. However, today, it is consequently judicious to propose that efficacious adaptation of 3DP technologies to composite materials could enable a simple composite manufacturing method with lower production cost and a high degree of automation, as reinforcements can be accurately placed, the laminated structure of composite parts can be further optimized in each layer, allowing for an increase in design freedom and mechanical performance.\textsuperscript{45}

In view of the cited literature, this review is focused to brief the various 3DP technologies, materials, and applications associated with the PMCs along with roadmaps for the future research efforts required in the considered domain of interest.

2 | 3DP TECHNOLOGIES FOR PMC SYSTEMS

Todays' manufacturing using 3D printing is taking up with the phenomenon of using multimaterial printing opportunities wherein the different materials could be deposited in a designed fashion as well as using a preblended composite feedstock consisting of different types of fillers.\textsuperscript{46,47} The choice of the composite formation generally depends on the types of printing systems; however, both approaches are capable of delivering distinct physicochemical properties into the resultant materials.\textsuperscript{44,48,49} This domain of composite printing is presenting a combination of superior technologies and material systems with an immense potential, affordability, simplicity, and unaccountable advantages.\textsuperscript{50} Table 1 describes the recent applications of 3DP systems for different end-user applications by utilizing PMC systems.

2.1 | Fused deposition modeling

Fused deposition modeling (FDM) is one of the most widely used 3DP systems that has been extensively considered by most of the domain researchers, across the world. The statistics reported in Parandoush and Lin\textsuperscript{42} highlighted that the commercial FDM systems held 41.5% of the market share, with the total of 15,000 machines sold by the end of 2010. However, today, it is almost impossible to count the sale of FDM systems as after the collapse of the technology’s patent, which earlier held by Stratasys Inc, various firms started the production of cost effective printers. Technically, FDM follows extrusion principle as discussed in plethora of publications.\textsuperscript{91-95} Further, the key elements of FDM\textsuperscript{96} and process parameters\textsuperscript{97-99} are highlighted by others. Traditionally, FDM was suitable for only pure thermoplastics; however, nowadays, reinforced feedstock are also commercially available. But the authenticity of such feedstock is never guaranteed, and most of the practitioners willingly develop their own feedstock by using suitable processing routes.\textsuperscript{52} For more information on the technological aspects, experimental schematics, materials, and applications, articles\textsuperscript{28,41,42} could be referred.

2.2 | Selective laser sintering

Selective laser sintering (SLS) is a 3DP process wherein the polymeric powder particles are fused with the help of high laser power.\textsuperscript{53,54} Similar to the other types of 3DP technologies, the laser fuses the polymer powder at specific locations for each layer specified by the design.\textsuperscript{100} In SLS printing, the quality characteristics of the resulting print depend on the processing condition, especially the laser power and scan speed.\textsuperscript{55,56} The working procedure of SLS is well explained in Mazzoli.\textsuperscript{67} The different classes of polymers available for SLS process are discussed in Drummer et al\textsuperscript{57} and Schmid et al.\textsuperscript{58} Particularly in biomedical engineering, the scope of SLS is huge.\textsuperscript{59-64} Further, blended polymers as well as PMCs are also suitable for SLS processing.\textsuperscript{65,66,101,102} In Bourell et al\textsuperscript{103} and Singh et al,\textsuperscript{104} authors discussed the various issues of SLS process.
### TABLE 1 Description of recent trends in 3DP of PMCs for various applications

<table>
<thead>
<tr>
<th>Schematic of Technology</th>
<th>Thermoplastic/Thermoset Matrix</th>
<th>Fillers</th>
<th>Application</th>
<th>Ref</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fused deposition modeling (FDM)</td>
<td>Thermoplastic (PLA)</td>
<td>Bronze</td>
<td>Structural</td>
<td>52</td>
</tr>
<tr>
<td></td>
<td>Thermoset (EVA)</td>
<td>Graphite</td>
<td>Electrical</td>
<td>53</td>
</tr>
<tr>
<td></td>
<td>Thermoplastic (ABS)</td>
<td>Copper/Iron</td>
<td>Thermal</td>
<td>54</td>
</tr>
<tr>
<td></td>
<td>Thermoplastic (PCL)</td>
<td>HAP</td>
<td>Biomedical</td>
<td>55</td>
</tr>
<tr>
<td></td>
<td>Thermoplastic (ABS)</td>
<td>Graphene</td>
<td>Structural and thermal</td>
<td>56</td>
</tr>
<tr>
<td></td>
<td>Thermoplastic (ABS)</td>
<td>Carbon fiber</td>
<td>Load bearing</td>
<td>57</td>
</tr>
<tr>
<td></td>
<td>Thermoplastic (PA-12)</td>
<td>Carbon nano-fiber</td>
<td>Mechanical</td>
<td>68</td>
</tr>
<tr>
<td></td>
<td>Thermoset (PA-11)</td>
<td>Carbon black</td>
<td>Mechanical</td>
<td>69</td>
</tr>
<tr>
<td></td>
<td>Thermoplastic (PCL)</td>
<td>Silica</td>
<td>Mechanical</td>
<td>70</td>
</tr>
<tr>
<td></td>
<td>Thermoplastic (PE)</td>
<td>Hydroxyapatite</td>
<td>Biomedical</td>
<td>71</td>
</tr>
<tr>
<td></td>
<td>Thermoplastic (PA)</td>
<td>Hydroxyapatite</td>
<td>Biomedical</td>
<td>72</td>
</tr>
<tr>
<td></td>
<td>Thermoplastic (PA-12)</td>
<td>Glass beads</td>
<td>Biomedical</td>
<td>73</td>
</tr>
<tr>
<td></td>
<td>Thermoplastics (Acrylate and methacrylate)</td>
<td>Yttria stabilized zirconia</td>
<td>Biomedical</td>
<td>76</td>
</tr>
<tr>
<td></td>
<td>Thermoset (Acrylate resin)</td>
<td>45S Bioglass</td>
<td>Biomedical</td>
<td>77</td>
</tr>
<tr>
<td></td>
<td>Thermoset (Polyramic (SPR-212))</td>
<td>SiOC and SiC</td>
<td>Mechanical</td>
<td>78</td>
</tr>
<tr>
<td></td>
<td>Thermoset (Methacrylate oligomers)</td>
<td>Boron nitride</td>
<td>Biomedical</td>
<td>79</td>
</tr>
<tr>
<td></td>
<td>Thermoplastic (PMMA)</td>
<td>Urea, resorcinol, and ammonium chloride</td>
<td>Smart materials</td>
<td>80</td>
</tr>
<tr>
<td></td>
<td>Thermoset (Envision-TEC)</td>
<td>Graphene</td>
<td>Electrical</td>
<td>81</td>
</tr>
<tr>
<td></td>
<td>Thermoset (Epoxy)</td>
<td>Glass fiber</td>
<td>Mechanical</td>
<td>83</td>
</tr>
</tbody>
</table>

(Continues)
2.3 | Stereolithography

When compared with other 3DP systems, stereolithography (SLA) has an edge as it possesses a comparatively high resolution and can create mechanically stable and watertight polymeric channel by using a liquidized photo-curable resin as feedstock,[105] to be cured by a UV light.[106] As mentioned in Bártolo and Gibson,[107] the applications of 3DP, again, are widespread among a multitude of industries, which includes fashion industry, automotive and aerospace, biomedical, and construction. Indeed, SLA has proven as a fast, stable, mask-less, and layer-by-layer additive process, with significant ability of building truly 3D, high-aspect-ratio, and light-weight microscale and mesoscale structures.[108] Similar to SLS, SLA too has noticeable scope for the biomedical applications as it can utilize both natural and synthetic polymers.[75] However, retaining the degradability and mechanical properties is of utmost importance for available polymers. Kalsoom et al.[41] and Parandoush and Lin[42] highlighted the advanced materials and applications of SLA. At the same time, a significant volume of literature cites the intrinsic problems of the SLA technology.[109-113]

2.4 | Laminated object manufacturing

The least explored and popular type of 3DP system is known as laminated object manufacturing (LOM), which majorly used for the production of prototypes with the help of paper, polymeric films and foils, and metal laminates as feedstock.[114-116] Further, the use of different combination of aforementioned feedstock helps attaining better mechanical properties.[82] LOM-based printing is divided into three major steps, including adhesion joining, clamping, and ultrasonic welding.[82] Thus, this approach represents a compromise between additive and subtractive manufacturing strategies in the building of three dimensional structures. Like FDM, in LOM too, fabrication of fiber-reinforced composite filaments and laminates is required as a prestep before printing.[43]
Inkjet printing is different from the other 3DP technologies in not just a way that it has high resolution but also because of the fact that it allows drop-on-demand of the various types of the feedstock such as hydrogels, bio-inks, liquid polymers, metallic solutions, and ceramics. Apart from the conductive interfaces, which have been practiced with inkjet printing most widely, this printing technology has also been used for building biomedical devices, sensors, electrical devices, etc. Especially during the last years, the fabrication of narrow conductive tracks by methods of inkjet printing has been investigated extensively. Further, researchers investigated the production of light emitting substances and electrically active devices by using inkjet printing.

As discussed above, there are a wide range of industrial applications acting as a catalyst for the concerned community in order to provide their continuous and blooming research efforts. Table 2 highlights the current industrial or scientific applications of 3DP-based PMCs.

<table>
<thead>
<tr>
<th>Application</th>
<th>Material System</th>
<th>3DP Used</th>
<th>Key Outcomes</th>
<th>Ref</th>
</tr>
</thead>
<tbody>
<tr>
<td>Smart devices</td>
<td>TangoBlack+ and shape memory polymer fibers</td>
<td>FDM</td>
<td>In view of the benefits of easy fabrication and the controllable multi-shape memory effect, the printed shape memory polymer structures have great potential in biomedical domain</td>
<td>122</td>
</tr>
<tr>
<td></td>
<td>Polyurethane acrylate oligomers, reactive diluents, photoinitiator, rheology modifier and the alumina platelets.</td>
<td>Direct ink-writing</td>
<td>The 5D design space covered by the multi-material magnetically assisted 3D printing platform gave lights to the manufacturing of functional heterogeneous materials</td>
<td>123</td>
</tr>
<tr>
<td></td>
<td>Polyelethylene glycol diacrylate and Barium Titanate Nanoparticle</td>
<td>3D optical printing</td>
<td>The composites with a 10% mass loading of the chemically modified barium titanate nanoparticle showed piezoelectric coefficients of ~40 pC/N</td>
<td>124</td>
</tr>
<tr>
<td></td>
<td>Polymorph and carbon black</td>
<td>FDM</td>
<td>The developed material and unmodified FDM system was found suitable printing electronic sensors able to sense mechanical flexing and capacitance changes</td>
<td>125</td>
</tr>
<tr>
<td>Energy</td>
<td>ABS/Graphene</td>
<td>3D printer HOF1-X1</td>
<td>The composites' linear thermal coefficient was lower than 75 ppm °C⁻¹ from room to glass transition temperature, making it crucial to build minute thermal stress</td>
<td>126</td>
</tr>
<tr>
<td>Biomedical</td>
<td>PLGA polymer with ß-tri-calcium phosphate</td>
<td>TheriForm</td>
<td>The developed scaffolds had higher percentages of new bone area compared with unfilled control defects</td>
<td>127</td>
</tr>
<tr>
<td></td>
<td>PLA, wax, and polysulphonamide</td>
<td>3D printer Model Maker II</td>
<td>The PLA scaffolds developed with complex internal architectures mimicked human trabecular bone were produced</td>
<td>128</td>
</tr>
<tr>
<td>Automobile</td>
<td>Nylon and carbon fiber</td>
<td>SLS</td>
<td>The printing presented a complete product with suitable tolerances</td>
<td>155</td>
</tr>
<tr>
<td>Aerospace</td>
<td>ABS/PLA, and short carbon microfibers</td>
<td>FDM</td>
<td>Filaments embedded with short carbon micro fibers showed better print capabilities for aerospace industry</td>
<td>156</td>
</tr>
<tr>
<td>Textile</td>
<td>Conductive polymers</td>
<td>FDM, SLS, and SLA</td>
<td>The fashion industry has received numerous benefits by the fruitful utilities of 3DPs</td>
<td>157</td>
</tr>
<tr>
<td>Food</td>
<td>Different types of natural polymers</td>
<td>FDM, SLS, Powder bed binder jetting, and inkjet printing</td>
<td>The review paper highlighted that there are a wide range of natural edible polymers which can be printed through various classes of 3DPs</td>
<td>158</td>
</tr>
<tr>
<td>Structural</td>
<td>Nickel coated with carbon and alumina fibers</td>
<td>SLA</td>
<td>Flexural strength of aligned samples with 1% by wt of carbon fiber loading was improved by 90%</td>
<td>159</td>
</tr>
</tbody>
</table>
SUMMARY AND FUTURE ROAD MAPS

From the cited literature, it has been observed that the scope of 3DP and PMCs for various scientific and engineering applications is huge. It has been seen that the 3DP technologies have successfully addressed the industrial applications, such as automobile, aerospace, structural, energy, biomedical, electrical, and smart devices. Further, for developing PMCs through 3DP technologies, suitable feedstock systems are not usually readily available. Therefore, literature has witnessed the in-house developed PMC systems; the practical utility of the 3DP systems has enhanced. However, such exercises demand intensive optimization of the materials' chemistries and processing states, which are found to be highly effective as concerned to the final outcomes. Nowadays, various conventional manufacturing technologies for the PMCs are being replaced with the 3DP owing to the availability of the better process control and feasibility of precise positioning of the reinforcements in later. Some of the recent publications even explored the feasibility of using reinforced thermoset polymers for demanding applications. However, use of a viscous feedstock, because of the presence of the fillers, is a big challenge as such material systems often demand parametric optimizations or replacement of the certain parts, accordingly. Moreover, it is difficult to conclude that whether the applications, as highlighted in the main text of the review, will receive commercialization certifications or will always be confined with the research and development laboratories. For instance, most of the biomedical devices and tools developed are yet to receive Food and Drug Administration (USA). Table 3 shows the research opportunities and challenges for 3DP of PMCs.

In order to establish the combined outcomes of PMCs and 3DP systems, it is a must to establish the repeatability and consistency of the manufactured parts. However, it is certainly difficult to predict how the available capacities of the printers will cover the broad range nano to macro. Despite the development of highly advanced applications through 3DP of PMCs, for example, energy storage devices, biomedical constructs, and electrical and electronic devices, it is always necessary to conduct a comparative analysis with the standard products on the basis of sustainability, energy, life, and efficacy. Quite a number of studies related to smart product manufacturing are still seeking for field applications as well as product lifecycle management. Moreover, the 3DP-based PMCs through a tight coupling among material development, process development, process control, testing, and certification of products produced by 3DP will lay the foundation of valuable progresses of many other emerging technologies. Industries and business enterprises should also welcome the academically developed 3DP-based PMCs with open arms as without the collaboration of academia industries, this technology cannot be flourished. The interdisciplinary research must be used in a variety of research activities to revolutionize 3DP-based manufacturing and inspection methods in order to overcome existing

<table>
<thead>
<tr>
<th>S No</th>
<th>Opportunities</th>
<th>Challenges</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>System up-gradation: The commercially available 3DP systems are generally compatible with pure polymers and are tuned accordingly. Further, different types of 3DP setups could be merged together to strengthen the merits.</td>
<td>The tuning of the 3DP systems in accordance of developed PMCs needs replacement of the elemental components which itself is time consuming task.</td>
</tr>
<tr>
<td>2</td>
<td>Feedstock development: There has been a huge scope for the development of the customized feedstock for the different types of printing systems in order to enable the resulting product suitable for end-user application.</td>
<td>This is an interdisciplinary task, which needs the combined efforts of the materials scientists, manufacturing experts, and statisticians.</td>
</tr>
<tr>
<td>3</td>
<td>Print production: This involves the application of the different optimization and statistical approaches in order to investigate the best process condition for the efficient fabrication of the prints.</td>
<td>The main challenge here is to find out the most significant parameters as any biased selection may result in undesirable outcomes.</td>
</tr>
<tr>
<td>4</td>
<td>Testing and evaluation: The opportunity available here in terms of evaluating the printed products in realistic environments, from where the performance of the products could be simulated very close to the realistic conditions.</td>
<td>Indeed, the creation of the closely simulating test environment is very difficult. It is the fact that most of the test has been performed using standardized specimens, which may not replicate the complexity of the actual geometries.</td>
</tr>
<tr>
<td>5</td>
<td>Certification: The products after successful testing should be certified in order to widen the scope of commercialization prospects. The academic contribution in this will make the process smooth.</td>
<td>Because of the variations in the standards in different countries, it is very difficult to make the commercial start-up successful.</td>
</tr>
</tbody>
</table>
manufacturing challenges and improve industry competitiveness. Furthermore, the hybridization of the various types of 3DP systems can overcome the exiting printing issues related to the materials and parameters. The combination of different printing principles will enable the production of the efficient multimaterial architectures.

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CONFLICT OF INTEREST

There exists no conflict of interest.

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FURTHER READINGS


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