ISSN No:-2456-2165

Advanced 3D Printing Materials: A Short Review of Polymers and their Composites

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Abstract:- 3D printing technology uses a variety of materials to build several objects. It is also called rapid prototyping technology. It uses a 3D digital model to prepare the object through layer by the layer printing method. At present lots of polymeric materials are being used with a large number of applications in 3D-printed objects. But 3D printed objects made of pure polymer show inferior thermal, mechanical, electrical and other properties, therefore having limited applicability. The addition of filler material in a pure polymer matrix enhances the functionalities of these polymers. A review of 3D printable polymers and their composites is done in this review paper

Keywords:- *3D Printing Technology, Rapid Prototyping, Polymeric Materials, Polymer Matrix Composites (Pmcs), Customized Products.*

I. INTRODUCTION

3D printing technology is also known as solid-freeform (SFF) or additive manufacturing. With the help of this technology, we can produce complex geometries easily. It is very similar to normal printers, which directly print the objects **[1]**. 3D printing provides freedom of design, effective product development, customized product, production when needed, and advanced application in the medical field**[2]**. This technology creates objects with reduced waste and satisfactory geometric accuracy. But at the same time,3D printing has limited materials, restricted build size, needs post-processing, and produces part structures. With this technology, we do not need mass manufacturing and worldwide transportation of raw materials as well as finished products. This technology only needs local transportation of 3D printers and raw materials in nearby areas. In the Additive manufacturing process, we use 3D drawing tools like CAD/E Solid Works/UG Pro to create 3D models, which are to be printed. Then these models are exported to change.STL format. Then 3D slicing software performs the slicing operation on this model & generates a G-codes file, this file is finally transferred to a 3D printer. At the end of this process, the 3D printer finally prints the object by depositing the material layer by layer on the build platform**[1]**.

- *Types of Materials for 3d Printing*

3D printers use different types of materials to print the objects depending on the needs. For example polymer, metal, concrete, ceramic, etc. But polymeric materials have the largest application& most extensive molding techniques**[1]**.Polymers have a low melting point and are economic and flexible to be used in various processes. Parts made with polymers have a high strength-to-weight ratio. At the same time, the pure polymer-produced 3D printed parts have a limitation on mechanical properties and functionalities. Therefore polymer matrix reinforced composites (PMC) are getting wide attention for avariety of applications in scientific, engineering, and technological fields, due to their improved physical, thermal, mechanical, structural, and electrical superiority. Polymer matrix composites (PMCs) are being used widely in the whole world due to the availability of a wide range of natural and synthetic reinforcements. Fabrication of polymer matrix composites can be done by embedment of particles, fibers, nanomaterials, or ceramic reinforcements into different polymers' matrices**[3]**. These types of PMCs are discussed briefly as follows-

- *Particle-Reinforced Polymer Composites*

These are largelyemployed to improve the various properties of polymer matricesdue to their low cost. Particles can be readilyblended with polymers, either in liquid form (SLA and DLP) or in powder form (SLS), or can be extruded into 3D printable filaments for the FDM process.Depending upon the polymer matrix and filler particles used, the composites show improved thermal Properties (Thermal conductivity, coefficient of thermal expansion, etc.), enhanced mechanical properties (tensile and compressive strength, storage modulus, isotropic properties, friction coefficient, etc.), and electrical Properties (dielectric permittivity, electrical conductivity, etc.) **[2, 3]**.

- *Fiber-Reinforced Polymer Composites*

These reinforced fibers can also improve the properties of polymer matrix composite significantly. General 3D printing processes, which use fiber-reinforced polymer matrix composites are direct-write techniques and fused deposition modeling (FDM). In the FDM process, fibers and polymer pellets are well mixed in a mixer. Then this mixture is fed to the extruder, which converts it into filaments. For the direct writing process, fibers are blended with polymer paste, and then it is directly extruded (Printed). The result showed improved mechanical properties like tensile strength depending on the polymer matrix and fiber (short or continuous) used. Powder-based technologies like SLS, are not preferred, since preparing a smooth layer of fiber-powder mixture is not so easy. Carbon fiber (CFs), glass fiber, and some other natural and synthetic fibers are generally used as fiber reinforcements, to enhance the structural and mechanical properties of the 3D printed parts. Some other natural fibers are also being used nowadays. Void fraction and fiber orientation of composites plays an important role in deciding the properties of the final composite object **[2, 3]**.

- *Limitations To Fiber-Reinforced PMCs*

✓ Fiber contents in the compositeare asignificantfactor, which affects the various properties of materials. Nowadays, adding fiber contents of more than 40 wt. % (mass fraction)is not allowed, because the composites with more fiber contentclog the nozzle**[1]**.

✓ Further, the composites with higher fiber contentcreate difficultyincreating continuous filaments for fused deposition modeling (FDM) because the presence of high fiber content reduces toughness. So, the presence of low fiber content limits the properties of the resulting composites **[2]**.

✓ Difficulty observed in the addition of continuous fibers is anotherbarinthe 3D printing of fiber-based composites. The maximum research doneso far is based on thereinforcement of short fibers in the polymer matrix. But in recent years, various researchers reported continuous fiber-based composites used in 3D printing**[2]**

- *Ceramic Reinforced Polymer Composites*

Ceramic-reinforced polymeric composites area recent development in the engineering and technological fields. Common ceramics used for reinforced polymeric composites are Al_2O_3, SiC, TiO_2, glass fiber, and many more. The parts manufactured with Ceramic-reinforced polymeric composites show superior thermal properties (Thermal stability, high glass transition temperatures), and morphological and mechanical properties (Improved toughness, hardness, fatigue resistance, and wear resistance). These parts also show chemical inertness and corrosion resistance. The polymer ceramic composite has various applications in the field of 3D printing. It can be used in artificial bone fabrication, tissue engineering technology, and the dental industry. It can also be used for rapid tooling in the machining of thermoplastic materials **[3, 4]**

➢ *Typical Polymeric Materials and their Composites*

Several typical polymeric materials for 3D printing are generally categorized as – **Thermoplastics–Thermosetting.**

- *Thermoplastics*

Those polymers, which when heated above a certain temperature become soft and flexible and get solidified when cooled down, are called thermoplastics. These can be re-melted and re-shaped frequently. These thermoplastics are principally used in selective laser sintering (SLS) and fused deposition modeling (FDM) in the field of additive manufacturing (3D printing). Some of the thermoplastics commonly used for 3D printing are PLA, ABS, TPU, PCL, PEEK, Nylon, PVA, etc.**[1]**. Most of the types of filament used in fused deposition modeling (FDM)and powder used in Selective laser sintering (SLS) for 3D printing are made of thermoplastics. These filaments can be made of many different colors, transparent or glow-in-the-dark.

✓ *PLA (Polylactic Acid)*

It is a vegetable-based thermoplastic material. PLA (polylactic acid) generally uses cornstarch and sugarcane as raw materials. It is a widely used natural raw material for additive manufacturing. PLA is a completely biodegradable, environment-friendly, and biocompatible thermoplastic polymer. PLA has widespread use and is very famous in the 3D printing field. It has certain advantages like excellent physical, thermal, and mechanical properties for 3D printing, but more brittleness (than ABS), less flexibility (than nylon), and transparent materials. In 3D printing, FDM (fused deposition modeling) technology generally uses PLA along with ABS **[1]**.

ISSN No:-2456-2165

Qinet al. [5] blended PLA with polycarbonate, which increased the impact strength and crystallization rate of polylactic acid, at the same time degradability of PLA is not affected.

Song et al.[6] madea Plastic-wood composite (PLA-WF) composite, which is also a biodegradable composite. When WF (eucalyptus powder) percentage in the composite was increased upto 50 % by weight, then an increment in modulus and tensile strength was observed in comparison to pure polymer.

Chizari et al. [7] Mixed carbon nanotubes (CNT) 40 % by weight in PLA to make nanocomposite. Which increased the electrical conductivity of the composite. Further studies found that this composite was successfully 3D printed in smart sensors in textiles.

Nanya et al. [8] manufactured continuous carbon-fiber (surface processing with methylene dichloride solution) reinforced PLA composite, using FDM and the result showed an enhancement in bending strength (164 %) and tensile strength (13.8 %) as compared to unprocessed fibers (Fig 1).

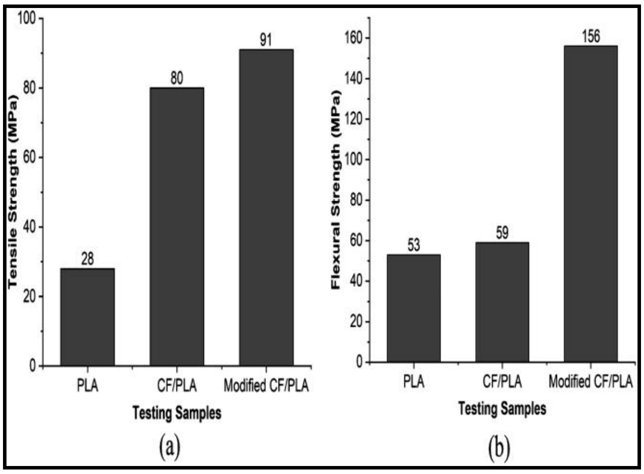

Fig 1 (a) Tensile Strength and (b) Flexural Strength of pure PLA, Carbon Fiber/PLA, and Modified Carbon Fiber/PLA Samples. Ref. [8]

Taoet al. [9]Made PLA-wood powder composite and extruded it in the form of wire, which was further used in the 3D printing process (FDM). Thermal properties, tensile strength, surface morphology, and crystallinity of composites were checked. It was found that the initial thermal stability of the composite decreased a little bit and the initial flexural strength of the composite increased.

Evgeni et al.[10]blended different compositions of multi-walled carbon nanotubes (MWCNT) and graphene nanoplates (GNP) in the polylactic acid substrate, which improved the electrical and thermal properties of polymer matrix composites (PMC). The well-diffused nanofillers enhanced the thermal and electrical properties of the composite. Composite can be extruded in the form of wire, which was further used in the 3D printing process (FDM) with enhanced electrical and thermal properties.

Table 1 PLA-based Composites

Investigator Name	Filler Material	Improved Properties
Qin et al. [5]	PC (Polycarbonate)	Increased impact strength and crystallization rate.
Song et al.[6]	WF (eucalyptus powder) upto 50 % by weight	Increased modulus and tensile strength.
Chizari et al. [7]	CNT (carbon nanotube) upto 50 % by weight	Increased electrical conductivity.
Nanya et al. [8]	Continuous carbon-fiber	Enhancement in bending (164 %) strength and tensile strength (13.8 %).
Tao et al. [9]	Wood powder	Increased initial flexural strength.
Evgeni et al. [10]	Multi-walled carbon nanotubes and graphene nanoplates	Improved thermal and electrical properties.

✓ *ABS (Acrylonitrile-Butadiene-Styrene)*

ABS is made by combining 03 monomers: acrylonitrile, butadiene, and styrene. It is suitable for additive manufacturing due to the following properties:

- ABS-manufactured objects have dimensional accuracy with a fine surface finishand good impact resistance characteristics.It has a higher melting point than PLA and extrudes with less friction. It is also suitable for product functional versatilities**[1]**.
- Each monomer of ABS resin is important because of its chemical structure. Acrylonitrile adds to chemical and heat resistanceand also provides surface hardness to the ABS resin. Butadienegives impact resistance, whereas styrene provides rigidity, strength,and processability **[1]**.
- However, the molten ABS has a bad odor, therefore ABS is generally not preferred as a printing material.

Zhong W et al. [11] blended carbon nano-fiber and glass fiber with ABS for fused deposition modeling-based additive manufacturing. The result showed that the machining and mechanical properties (Modulus and Tensile strength) of the 3D-printed objectswere enhanced.

Nikzad M et al. [12-13]usedcopper and iron powder to blend with ABS toimprove its properties of ABS. When this composite is used in fused deposition-based additive manufacturing. It was found that the mechanical, electrical, and thermodynamic properties of ABS (as raw material) have been considerably enhanced. With high iron content ABS/Fe composite, theenergy storage modulus, rigidity,and compression strength of 3D printedobjects are significantly enhanced.

ISSN No:-2456-2165

Tekinalp et al. [14]used short carbon fiber (upto 40 % by wt.) of 0.2 - 0.4 mm to embed in ABS resin. Resultsdemonstrated that the modulus and tensile strength of 3D manufactured objects were enhanced by 700% and 115% respectively. But objects made by fused deposition modeling have high porosity. So the further scope of research is, to improve the compatibility and adhesion of two phases through the chemical treatment of reinforced fibers(Fig 2)

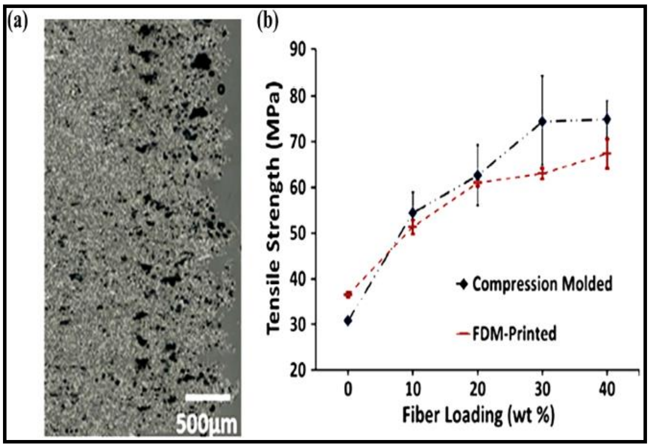

Fig 2 (a) Micrograph of the Polished Surface of the Printed ABS/30 Wt. % Carbon Fiber Composite
(b) Effect of Fiber Content on Tensile Strength of the Printed ABS/Carbon Fiber Composite. Ref.[14].

Wei et al. [15] added graphene tothe ABS matrix, which improved thermal stability and enhanced thermal conductivity.

Quillt et al. [16] made ABS /BN (boron nitride) composites, which enhanced electrical insulation by injection molding and thermal conductivity by FDM. Further research demonstrated that with the increase in BN percentage, the flexural strength and toughness decreased.

Table 2 ABS-based Composites

Investigator Name	Filler Material	Improved Properties
Zhong W et al. [11]	Carbon nano-fiber and glass fiber	Improved modulus and Tensile strength.
Nikzad M et al. [12-13]	Copper and iron powder	Enhanced energy storage modulus, rigidity, and compression strength.
Tekinalp et al. [14]	Short carbon fiber (upto 40 % by wt.)	Increased modulus by 700 % and tensile strength by 115 %.
Wei et al. [15]	Graphene	Enhanced thermal stability and thermal conductivity.
Quillt et al. [16]	BN (Boron nitride)	Increased flexural strength and toughness.

✓ *TPU (Thermoplastic Polyurethane)*

TPU shows a wide range of properties such as resistance to abrasion, grease, and oil. It also has elasticity and transparency. It also has corrosion resistance, excellent wear resistance, and radiation resistance. It has wide applicability in additive manufacturing[1].

Akbarian et al. [17] reinforced aramid fiber (short)in a polyurethane matrix and observedthat these fibersshowfracture and axial splittingit is embeddedin the polyurethane matrix. Aramid fibers are natural fibers and are cheap from aneconomic point of view.

El-shekel et al. [18-19]blended Kenaf fiber and Cocoa Pod Husk Fibers with TPU to strengthenit. The result showedenhanced bending and tensile properties.

Kim et al. [20]discovered a 3D multi-axial force sensor madethroughfused deposition modeling, whose sensing part was manufactured of carbon/TPU nanocomposite structure partwas made ofTPU as filament.

Wang et al. [21] prepared a nanoporous PMMA(Polymethyl methacrylate)/TPU composite. Itshows enhanced mechanical properties with improvedthermal conductivity.It has wide applicability in additive manufacturing.

Chen et al. [22]made TPU/GO/PLA nanocomposite elastomer. This biocompatible composite is a good tissue engineering material. Ithas unique excellent electro-thermal properties and antibacterial properties.

Table 3 TPU-based Composites

Investigator Name	Filler Material	Improved Properties
Akbarian et al. [17]	Aramid (short nylon) fiber	Exceptional tensile strength and thermal stability.
El-shekeil et al. [18-19]	Kenaf fiber and Cocoa Pod Husk Fibers	Enhanced bending and tensile strength.
Kim et al. [20]	Carbon nanoparticles	Improved electrical properties used in making 3D sensors.
Wang et al. [21]	Nanoporous PMMA (Poly methyl methacrylate)	enhanced mechanical properties with improved thermal conductivity.
Chen et al. [22]	GO/PLA nano-materials	excellent electro-thermal properties and antibacterial properties.

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✓ *PCL (Polycaprolactone)*

PCL is used in additive manufacturing (3D printing)because of its low melting point (lower than ABS). It is a biodegradable, biocompatible, and non-toxic material and safe in handling. Itis also biodegradable by hydrolysis, which makes it an excellent material for medical research and applications. **[1]**.

Visser et al. [23] developed an additive manufacturing technology, called melt electrospinning writing (an emerging technique that uses polymer melts for controlled deposition of the electrospun fiber). These fibers when mixed with methyl acrylamidemakea high-strength scaffold.

Lee et al. [24] blended PCL with chitosan (CS)(obtained by electro-spinning)and made PCL/CS chains by additive manufacturing. This composite is used successfully to produce artificial blood vessels.

Qu, et al. [25] prepared nHA (nano-hydroxyapatite 30% (wt.))/PCLnanocomposite. This composite is then used to make bone scaffolds throughelectro-hydrodynamic printing. This scaffoldhas a diameter (8. of 85±12)µm, verynear to the size ofliving cells. Due to good biocompatibility,these scaffolds open the door for tissue regeneration

Table 4 PCL-based Composites

Investigator Name	Filler Material	Improved Properties
Visser et al. [23]	Methyl acrylamide	Improved strength used for making scaffolds.
Lee et al. [24]	Electro spun chitosan (CS)	High strength and flexibility are used for manufacturing artificial blood vessels.
Qu, et al. [25]	nHA (nano-hydroxyapatite 30% (wt.))	Good biocompatibility is used in making bone scaffolds and tissue regeneration.

✓ *Peek (Polyetheretherketone)*

In the field of 3D printing, PEEK is known for its high performance. PEEK is a highly crystalline material and undergoes significant dimensional shrinkage. The parts manufactured by PEEK show low warping and better dimensional accuracy. Since PEEK is extruded at a quite high temperature, therefore it shows better adhesion with the previously printed layer. PEEK has high strength and shows chemical inertness.

✓ *Polyamides (Nylon)*

Due to its strong self-bonding between layers and flexibility Nylon filament isnow vigorously being used in the 3D-printing industry. It is extruded between 240 - 270 degrees Celsius. Nylon is not dissolved in acetone, like ABS and PLA.Nylon materials can be dyed in different colors using common clothing dyes, which then can be used for nylon fabrics. These can be opaque or transparent too.

Chung et al.[26] added a glass bead in nylon – 11, which solved the problem of low tensile strength and storage modulus.

Athreya et al.[27] added carbon black with nylon-12 to improve the electrical properties and enhance the flexural modulus of the composite.

Van Der et al. [28]made a composite by reinforcing continuous carbonfiber (CF) intoa nylon polymer matrix for 3D printing. Studies showed an improvement in the tensile strength of the printed objects.

Hui et al. [29]prepared nHA (nanohydroxyapatite) coated with nylon-12 powder, which showed the improved functional and structural behavior of the composite. The 3D printed objects using selective laser sintering (SLS). When nHA was added upto 0.5 wt. %, a significantimprovement in modulus and tensile strength was observed.

Boparai K et al. [30] added Al and Al_2O_3 to Nylon – 6, which improved the wear-resistant property.

Table 5 Polyamides-based Composites

Investigator Name	Filler Material	Improved Properties
Chung et al. [26]	Glass bead	Enhanced tensile strength and storage modulus.
Athreya et al. [27]	Carbon black	Improved electrical properties and flexural modulus.
Van Der et al. [28]	Continuous carbon-fiber (CF) (34.5 vol %)	Improvement in tensile strength by 446 %.
Hui et al. [29]	nHA (nanohydroxyapatite) up to 0.5 wt. %	Improvement in modulus and tensile strength.
Baraopi K et al. [30]	Al and Al_2O_3	Improved wear resistance

✓ *Polycarbonates (PC)*

These thermoplastic polymers have a carbonate group in their chemical structure. These can easily be molded and worked. These are tough, strong materials and upto a certain degree act as a thermal insulators. Some polycarbonates are optically transparent. These are lightweight and can be easily cut and shaped. 3D printed parts manufactured by polycarbonates have high impact strength, high adhesion properties, and low hardness and are transparent.

Shemelya et al. [31] prepared Tungsten/PC composite, which improved the tensile strength, impact resistance, and dielectric permittivity.

✓ *PVA (Polyvinyl Alcohol)*

It is used as a support material in 3D printing because it has good solubility in water and also showsa biodegradable property.It is extruded between 180 - 200 degrees Celsius. PVA is electrically conductive, therefore it is used to print electrical circuits into fabricated products.Poly (vinyl alcohol) (PVA) and its composites have gained significant attention as cutting-edge materials for 3D printing owing to their distinctive properties. PVA is an intelligent and stimuli-responsive polymer that has already exhibited exceptional mechanical, thermal, and chemical strength in various applications such as medicine and the environment. Recently, researchers have evinced an inclination towards exploiting PVA in 3D printing technologies due to its remarkable flowability, stimuli-responsivity, extrudability, biocompatibility, biodegradability, cost-effectiveness, and other exceptional attributes. These compelling benefits render PVA and its composites an alluring choice for a broad spectrum of applications, ranging from drug delivery systems and tissue engineering to the production of biodegradable packaging materials and construction materials. Hence, PVA and its composites have emerged as an increasingly popular material for 3D printing, paving the way for innovative and developmental opportunities across various domains.

✓ *HIPS (High-Impact Polystyrene)*

It is a modified form of normal polystyrene. HIPS is a low-cost, amorphous thermoplastic material. It is used as a support material in 3D printing because it has good solubility in limonene (a biologically derived solvent made from citrus plants). Itis used in packing material and food containers. HIPS has similar properties to ABS but does not dissolve in acetone.

- *Thermosetting*

Those polymers, which cannot be re-melted by applying heat once they are solidified are known as thermosetting. These thermosetting decompose at a higher temperature. Thermosetting resins are cured by the photopolymerization process i.e. by applying ultraviolet (UV) light. 3D printing processes generally used to cure thermosetting are SLA (Stereolithography) and DLP (Digital light processing). Commonly used thermosetting resins for 3D printing are acrylate resin and epoxy resin. Some other resins are3D-systems Accura Ceramax, DSM Somos PerFORM, Liqcreate Strong-X, Carbon3D CE221 131, Formlabs Grey Pro,and many more(Developed according to the need of application like standard resin, durable resin, rubber-like resin, high-temperature resin, castable resin, dental and medical resin, and ceramic filled resin, etc.).The mechanical properties of these polymers are dependent on photo-initiator concentration, monomer composition, and curing conditions. The amount of cross-linking alters the toughness, thermal stability, strength of these polymers, and other properties.**[32].**

✓ *Acrylate Resin*

Bio-based Acrylate photo-polymer resins are a mixture of multifunctional monomers, oligomers, a photo-initiator, and an optical absorber. All formulations demonstrated adequate viscosity. The absorber controls the depth of penetration of the incident light and so the amount of polymerization. The stiffness and thermal stability of 3D-printed objects increase with the increase in the double-bond concentration inside the acrylate resin. Good surface finish and high resolution can be obtained in 3D-printed objects due to their high viscosity. Good cross-linking density and non-homogeneity make acrylate resin-based parts brittle and tough **[32]**.

✓ *Epoxy Resin*

It is a recyclable, inexpensive material. Epoxy resin is easy to add colors and objects into resin. It is also a tough and thermally reactive material.

Hector et al. [33] blended epoxy resin with a multi-walled carbon nanotube (10 wt. %), which improved the tensile strength of a composite by 7.5 % as compared to pure polymer.

Compton et al. [34] produced a composite by reinforcing the epoxy resin matrix with short carbon fibers and SiC whiskers. The result showed high stiffness and toughness along with improved Young's modulus of the 3D printed objects.

Table 6 Epoxy Resin-based Composites

Investigator Name	**Filler Material**	**Improved Properties**
Hector et al. [33]	Multi-walled carbon nanotube (10 wt. %)	Improved the tensile strength by 7.5 %.
Compton et al. [34]	Short carbon fibers and SiC whiskers (35 wt. %)	High stiffness and toughness along with improved Young's modulus.

II. CONCLUSION

- The limited availability of diverse and readily printable materials, including thermoplastic polymers and their composites, thermosetting photopolymers, and powder-formed materials, restricts the application of 3D printing technology in the industry.

- There is a pressing need to develop a wider range of printable materials that are not only cost-effective but also environmentally friendly, to meet the increasing demand for diverse applications in the 3D printing industry.

- The low mechanical strength of most printed composites is a significant limitation, resulting in parts that fail to meet functional requirements, which poses challenges for the production of high-performance end-use products.

- The presence of voids in printed parts is one of the main reasons for their low mechanical strength, necessitating post-processing methods that increase time and cost, and hinder the efficiency of the 3D printing process.

- Inconsistent properties of printed parts, such as uneven distribution of material and varying mechanical properties, pose challenges in achieving consistent and uniform quality of printed objects, requiring further research and development.

- Despite the development of several 3D printers capable of processing different types of materials, there is a need for continued innovation in machine design and capabilities to enable the printing of a broader range of materials with higher precision and speed.

- The cost of 3D printers remains a significant barrier to their widespread adoption in industrial applications, necessitating efforts to make them more affordable and accessible to a wider range of users.

- Poly(vinyl alcohol) (PVA) and its composites have emerged as promising materials for 3D printing due to their unique properties, such as flowability, stimuli-responsivity, extrudability, biocompatibility, biodegradability, and cost-effectiveness.

- PVA and its composites offer compelling benefits for a broad spectrum of applications owing to its promising mechanical and chemical strength, including drug delivery systems, tissue engineering, biodegradable packaging materials, and construction materials, making them an attractive choice for 3D printing applications.

- Despite the current limitations, ongoing research and development efforts are driving innovation in 3D printable materials, machines, and processes, paving the way for future advancements and opportunities in various domains, and unlocking the full potential of 3D printing technology in the industry.

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