Biointerface Research in Applied Chemistry
Open-Access Journal (ISSN: 2069-5837)

Review
Volume 15, Issue 1, 2025, 15
https://doi.org/10.33263/BRIAC151.015

Review of Biopolymer Composite Through 3D Printing Techniques

Aiman Haiqal Aizam Hamidie [1], Sharifah Adzila [1,*], Aisyah Razak [1], Najah Mat Isa [1]

1 Faculty of Mechanical and Manufacturing Engineering, Universiti Tun Hussein Onn Malaysia. 86400, Parit Raja, Johor, Malaysia
* Correspondence: adzila@uthm.edu.my (S.A.);

Scopus Author ID 43760889800
Received: 5.03.2024; Accepted: 15.12.2024; Published: 12.02.2025

Abstract: Since polymers are so important in our daily lives, 3D printing began with a polymer substrate for creating components. Aside from that, biopolymer composites have become popular due to their environmentally friendly, biodegradable, and biocompatible properties. Despite recent advances in 3D printing of biopolymer composites, this paper reviewed the printing technique of each biopolymer composite, which has a large number of printing techniques such as Fused Deposition Modelling (FDM), Selective Laser Sintering (SLS), Stereolithography (SLA), Ink Jet and others that can be used for biopolymer composite due to mechanical properties findings and application. These biopolymer composites are widely utilized in the biomedical field and are predicted to increase their benefits in this field, as well as FDM, which is the most regularly used 3D printing process.

Keywords: Biopolymer composite; 3D printing; FDM; SLS; SLA; Ink jet

1. Introduction

The method of quickly generating a system or component representation before its ultimate release or commercialization is referred to as rapid prototyping (RP) and is used to define the process in various industries. To put it another way, the goal is to produce something fast, and the result will either be a prototype or a basic model that will serve as the foundation for further models and, ultimately, the finished product. Additive manufacturing (AM) is the official name for what was once known as rapid prototyping and is often referred to as 3D Printing [1]. This technique has allowed a large deal of design flexibility, creating very complex components, products with a high degree of customization, and efficient waste reduction. The most recent one, known as Industry 4.0, is characterized by the adoption of practices that include the combination of advanced information technology and intelligent production systems. As a result, AM plays an essential part in the development of Industry 4.0 due to its various advantages, including the reduction of wasted time and materials, rapid prototyping, high efficiency, and decentralized production processes [2,3]. Methods of additive manufacturing (AM) have seen remarkable expansion and development in recent years. AM for polymers is an intriguing topic with significant potential for transformational and translational research in various sectors, including the biomedical, aerospace, and electronic industries. Extrusion of material, material jetting, vat polymerization, and powder bed fusion are some of the current technologies for polymer additive manufacturing [4]. Meanwhile, on the topic of the monetization of renewable bioresource materials, researchers have produced exemplary publications. Biopolymers may be derived from a wide variety of biobased raw materials, the most common of which are starch, cellulose, and biomass from wood. When compared to more traditional forms of plastic, they provide a considerable contribution to reducing the overall carbon impact [5]. Materials that comprise a blend of traditional ceramics, ceramics, or unexpected qualities that are not present in polymers are required to use modern technology in additive manufacturing (AM). The blended materials create new materials by combining two or more components, each of which has desirable attributes, in order to generate the traits and characteristics that are required of the finished product; an example of CaP was determined by their biocompatibility and bioactivity of the material [6-9]. The use of many materials in conjunction with one another to achieve the required qualities of each component may provide new possibilities and problems for additive manufacturing research [10].

2. 3D Printing Method Preparation

Due to their biodegradability and nontoxicity, the biopolymers family is an excellent candidate for fabricating implants and bio-devices that will come into direct contact with the human body. Because of this, there is a significant possibility that the biopolymers will be used in the 3D printing of biobased devices [11]. The method of 3D printing is one in which layers are added one on top of the other, and it can be used to create very complex and specialized devices that vary in size from the macro to the micro-scale. In recent years, additive manufacturing (AM) technology has rapidly advanced in various fields and industrial domains, including biomaterials, pharmaceutics, medical, aerospace, artworks, smart devices, and other areas [12]. These advancements have been made for both metal and polymer materials. The use of 3D printing technology in tissue engineering, which falls under the umbrella term of bioprinting, has led to a substantial number of noteworthy advances in biomedical implants for bone, blood vessels, tissue, and other anatomical components [13]. This research reviews the comprehensive information methods preparation and their applications.

2.1. Fused Deposition Modelling (FDM).

An FDM system, also known as Fused Filament Fabrication-FFF is an advanced technology that is widely used for printing polymeric materials such as polylactic acid (PLA), acrylic butadiene styrene (ABS), polyethylene terephthalate (PET), Polyhydroxyalkanoate (PHA) nylon, and so on. This technology was developed by Fused Filament Fabrication [14]. In FDM, the thermoplastic filaments are passed through one or more printing nozzles at a temperature higher than the melting point of the filament to build a three-dimensional object using a method known as layer-by-layer deposition. After being placed, the layer underneath it will instantly begin to harden and cool, allowing it to form a connection with the layer that will be put directly on top of it shortly thereafter [15-16]. The inexpensive cost of the printer and the ease with which it may be processed have led to FDM being the technology that is utilized the most out of all the others. However, determining the ideal parameters for polymer printing, such as nozzle temperature, printing speed, and any post-processing factors, may be difficult [17]. Figures 1 and 2 depict the illustration and mechanism of Fused Deposition Modelling.

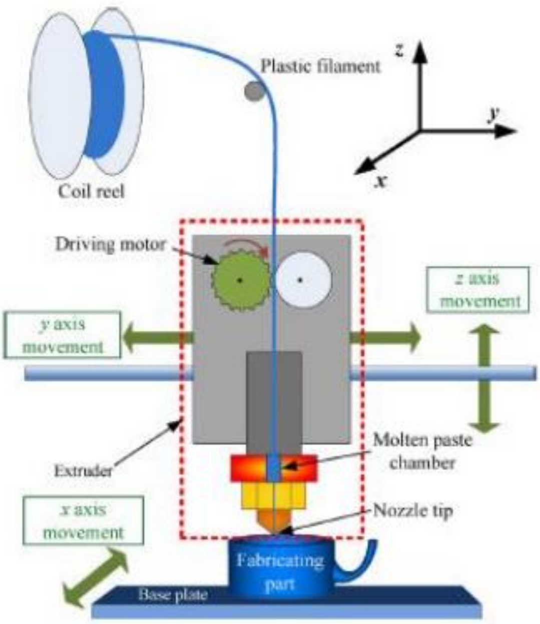

Figure 1. Illustration of fused deposition modeling. Reproduced from [18] with open access from IOP Conf. Ser.: Mater. Sci. Eng. Copyright 2021.

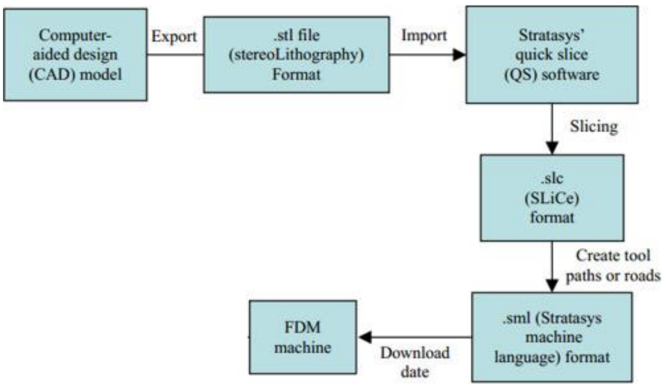

Figure 2. Flowchart of fused deposition modeling. Reproduced from [19] with Elsevier Publishing. Copyright 2020.

Table 1. Overview of biopolymer composites through fused deposition modeling.

References	Material	Printing Technique	Mechanical Properties	Application	Year
[20]	PHA/PF	FDM	Tensile Strength: 10-25MPa	Biomedical	2021
[21]	PHA/SSS	FDM	Tensile Strength: 10-25MPa Young Modulus: 900-1700MPa	Artificial Bone	2018
[22]	PHA/Lignin	FDM	Shear Viscosity: 150.8 Pa.s±15.4Pa.s	Wood Industry	2019
[23]	PHBH/NCF	FDM	Yielding Stress: 17-23MPa Elastic Modulus: 897-1014MPa	N/A	2019
[24]	PLA/PHB/NC	FDM	Storage Modulus: 3-6GPa	Biobased	2020
[25]	PHA/WF	FDM	Tensile Strength: 38.7MPa Flexural Strength: 77.3 MPa	Packaging Industry	2021
[26]	PHBV/RS	FDM	Max. Load: 2N	Piezoelectric Device	2021
[27]	PLA/PHB/Cellulose	FDM	Thermal Stability	Biobased Product	2019
[28]	PCL/Borate Glass	FDM	N/A	Tissue Engineering	2017
[29]	PLA/Graphite	FDM	Young's Modulus: 1866Mpa	Sensors	2020

References	Material	Printing Technique	Mechanical Properties	Application	Year
			Compressive Strength: 38.6MPa		
[30]	PHB/PLA/HA	FDM	Flexural Modulus: 3.20GPa	Biomedical	2022
[31]	PLA/cHA	FDM	Compressive Strength: 2.12MPa	Bone scaffold	2020
[32]	PLGA/TCP	FDM	Compressive Strength: 3.7±0.2MPa Young's Modulus: 114.9±15.4MPa	Bone Scaffold	2022
[33]	PCL/HA	FDM	Tensile Strength: 20.25MPa Flexural Strength: 18.77MPa	Tissue Engineering	2019
[34]	PCL/β-TCP	FDM	Compressive Strength: 20MPa	In vitro	2021
[35]	HDPE/n-HAp	FDM	Plasma Treatment: 399.5eV	Bone Tissue	2022
[36]	PLA/HA/Ch	FDM	Flexural Testing: 120N	Tissue Engineering	2021
[37]	PEEK/cHAp	FDM	Tensile Strength: <50MPa Modulus of Elasticity: 7GPa	Bone Implant	2021
[38]	PCL/PLGA/β-TCP	FDM	Tensile Test: 20MPa	Bone Regeneration	2016
[39]	PLA/Flax Fibre	FDM	Tensile Strength: 10.20MPa	Sustainable Product	2019
[40]	sPEEK/HAP/rGO	FDM	Compressive Strength: 115-135MPa	Bone Implant	2022
[41]	PLA/Ch	FDM	Compressive Strength: 31MPa Tensile Strength: 29MPa Flexural Strength: 37MPa	Scaffold	2020
[42]	PLA/PVC/Wood Powder	FDM	Flexural Strength: 14.14MPa	Scaffold	2022
[43]	PLA/Wood	FDM	Tensile Strength: <35MPa Compressive Strength: <70MPa	Sustainable Filament	2021
[44]	PLA/β-TCP	FDM	Elastic Modulus: 65-72MPa	Scaffold	2021
[45]	PLA/LIPAs	FDM	N/A	Information Storage	2022
[46]	PLA/AV	FDM	N/A	Bone Tissue Engineering	2022
[47]	PLLA/HA/dECM	FDM	Flexural Strength: 150-200MPa	Bone Tissue Engineering	2022
[48]	PLA/CCNF	FDM	Tensile Strength: 23MPa	Dielectric Heat Sources	2020
[49]	PLA/Ti-Zr	FDM	Energy Measurement: 57.0711mW/m^2	Medical Implant	2022
[49]	PLA/Zn	FDM	Compressive Strength: 12-17MPa Elastic Modulus: 500-700MPa	Tissue Regeneration	2022
[50]	PLA/Keratin/Ch	FDM	Elastic Modulus: 1200-1250MPa	Medical Field	2020
[51]	PCL/Gelatin	FDM	Young's Modulus: 12.3±1.6MPa	Tissue Regeneration	2020
[52]	TPU/lignin	FDM	Tensile Strength: 10.87MPa	Biomedical Scaffold	2022
[53]	PLLA/Steel Particle	FDM	Effective Modulus: 0.8-1.5GPa	Tissue Engineering	2018
[54]	Chitin/IC/WC	FDM	N/A	Electrical Energy Delivery	2022
[55]	PLA/DDGS/ Plasticizer	FDM	Tensile Strength: 44.9MPa-42.7MPa Young's Modulus: 1565-1500MPa	Sustainable Filament	2021
[56]	PLA/EE	FDM	Tensile Strength: 183MPa	Eco-Friendly Product	2017
[57]	PLA/PP/NFC	FDM	Tensile Strength: 15.2MPa	Structural	2020

References	Material	Printing Technique	Mechanical Properties	Application	Year
			Compressive Strength: 8.5-15.5kN/m Flexural Strength: 142-162kN		
[58]	PLA/Clay	FDM	Modulus Strength: 3.27-3.66GPa Tensile Strength: 46.12-55.58MPa	Sustainable Filament	2018
[59]	PLA/SiC/C	FDM	Recovery Force: 1.75-3.08N	Shape Memory Polymer	2018
[60]	PCL/GNP	FDM	Compressive Strength: 35.3&17.4MPa	Myocardial Tissue Engineering	2022
[61]	PLA/Wood (Poplar Trees)	FDM	Tensile Strength: 50MPa	Renewable Material	2020
[62]	PLA/Rice Husk	FDM	Storage Modulus: 8.6-13.4MPa	Renewable Product	2022
[63]	PLA/WF (Aspen)	FDM	DTG Curved Peak: 330°C	Sustainable Filament	2020
[64]	PLA/AT	FDM	Tensile Strength: 2.6-3.1MPa Young's Modulus: 2.1-2.9GPa	Sustainable Material	2020
[65]	PLA/Ti	FDM	Tensile Strength: 28.3±0.2MPa 40.2±0.2MPa	Scaffold	2020
[66]	PLA/Olive Wood	FDM	E=1.4-2.1GPA Yield Stress: 30.7-40.9MPa	Renewable Material	2022
[67]	PLA/HPMC	FDM	Tensile Strength: 42±3.6MPa	Sustainable Filament	2020
[68]	PLA/Silicon	FDM	Tensile Strength: 60MPa	Sustainable Material	2017
[69]	PHBH/CNCs	FDM	E'=0.1~4.00GPa	Biodegradable Products	2020
[70]	PLA/Almond Shell	FDM	N/A	Sustainable Filament	2022
[71]	PLA/Lignin	FDM	N/A	Healthcare	2019
[72]	PBAT/Wood	FDM	Flexural Strength: 31-53MPa	Pellet	2018
[73]	PLA/Biogenic Carbon	FDM	N/A	Sustainable Filament	2018
[74]	PLA/Hemp	FDM	E'=2333-3130MPa 5.15-7.45MPa	Sustainable Filament	2019
[75]	PCL/ZnHA	FDM	Stiffness Modulus: 37.8MPa	Bone Tissue Engineering	2021
[75]	PCL/ZnHa-rGO	FDM	Stiffness Modulus: 49.5MPa	Bone Tissue Engineering	2021
[76]	PLGA/CQD	FDM	N/A	Scaffold	2022
[77]	PBAT/TiO_2	FDM	DMA: 3.0±0.2 & 4.3±0.2GPa	Composite Filament	2022
[78]	PCL/Porous Glass	FDM	Compressive Strength: 80±10-102±11MPa	Bone Defect	2019
[79]	PLA/SEBS/CNT	FDM	Tensile Strength: ~3000MPa	Industry	2021
[80]	PLA/Walnut Shell	FDM	Tensile Strength: 2.684MPa	Sustainable Filament	2022
[81]	BioPE/TMP	FDM	N/A	Sustainable Filament	2018
[82]	PHB/MWCNT	FDM	Tensile Strength: 30-35MPa	Sustainable Filament	2021
[83]	PLA/Wood Polysaccharides	FDM	Flexural Modulus: 3.2±0.3GPa	Industry	2018
[84]	PLA-PHB/Corncob	FDM	Tensile Strength: 23MPa Tensile Modulus: 1.4GPa	Sustainable Filament	2022
[85]	MBG/SA	FDM	N/A	Biomedical	2020

2.2. Selective Laser Sintering (SLS).

The additive manufacturing technique known as selective laser sintering (SLS) printing is a kind of powder bed fusion (PBF), which may be used in the production of polymeric components. Based on a previously established CAD model, the laser beam plays a significant part in this process since it is responsible for melting the polymer powder in a selected approach [86]. In this technique, the first layer, which is quite thin, is produced. After that, a new layer is created by simultaneously sintering and heating the material that is already on top of the layer that was created before. This process is done several times until the component has been produced in its entirety [87]. The SLS printing process has a lot of potential to be used in the production of porous scaffolds of a high grade and sophistication that include intricate patterns [88]. This method may also be used in the production of goods containing biopolymer composite such as polyglycolide (PGA), poly lactic-co-glycolic acid (PLGA), Polyether ether ketone (PEEK), etc., which makes them a very good option, and well-tolerated by immune systems and cells [89]. Therefore, 3D printing is a highly ideal and quick manufacturing process that may be employed for the so-called dissolving implants. This is because 3D printing is capable of producing incredibly complex shapes. Figures 3 and 4 show the illustration and mechanism of Selective Laser Sintering, and Table 2 shows its utilization on biopolymer.

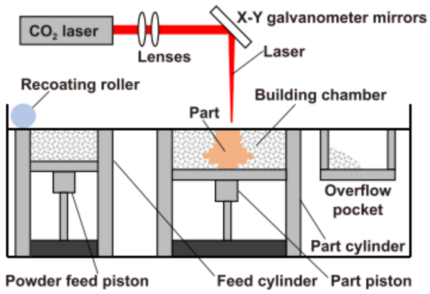

Figure 3. Illustration of selective laser sintering. Reproduced from [90] with open access from Int. J. Extrem. Manuf. Copyright 2022.

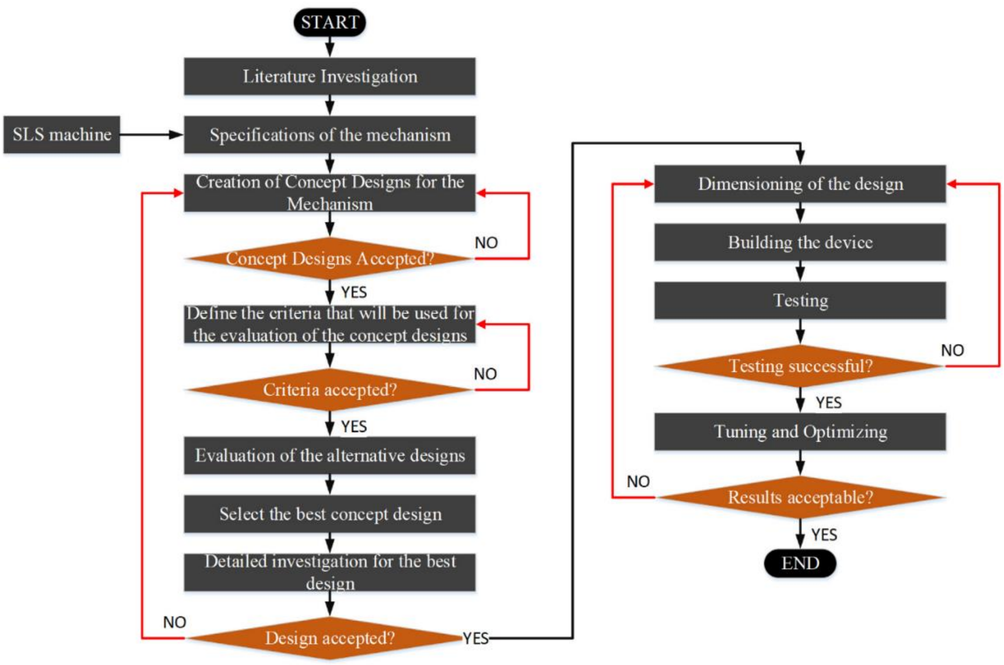

Figure 4. Flowchart of selective laser sintering. Reproduced from [91] with open access from Int. J. Manuf. Mater. Process. Copyright 2022.

Table 2. Overview of biopolymer composite through selective laser sintering.

Reference	Material	Printing Technique	Mechanical Properties	Application	Year
[92]	PHBV/CaP	SLS	Compressive Strength: 0.55MPa	Bone Scaffold	2010
[93]	PLGA/HA	SLS	Compressive Strength: 40MPa	Bone Regeneration	2021
[94]	PEEK/ZrO_2	SLS	Yield Stress: 5-17MPa	Scaffold	2019
[94]	PEEK/Al_2O_3	SLS	Yield Stress: 28-45MPa	Scaffold	2019
[94]	PEEK/TiO_2	SLS	Yield Stress: 5-8MPa	Scaffold	2019
[94]	PEEK/Cu	SLS	Yield Stress: 5MPa	Scaffold	2019

2.3. Stereolithography (SLA).

SLA technology is one of the pioneering technologies for 3D printing that uses a photochemical process to enable the manufacture of components or products. This approach may be considered one of the most versatile. An ultraviolet (UV) laser is used in this technology to polymerize the photocurable resin and crosslink the chemical monomers [95]. Therefore, the use of laser irradiation results in the formation of a patterned layer that is two-dimensional. Following the completion of the first phase, the printing platform will shift, allowing for the formation of a subsequent layer. SLA can fabricate the whole product via an iterative process that builds the product layer by layer [96]. SLA has made use of a variety of biopolymers. An aqueous poly(ethylene glycol) (PEG) hydrogel solution, for example, has been prepared, and study results demonstrate that the hydrogel may protect human skin fibroblast cells encapsulated in the solution from injury throughout the SLA process [97]. Figures 5 and 6 illustrate the illustration and mechanism of stereolithography and the utilization of biopolymer composite in Table 3.

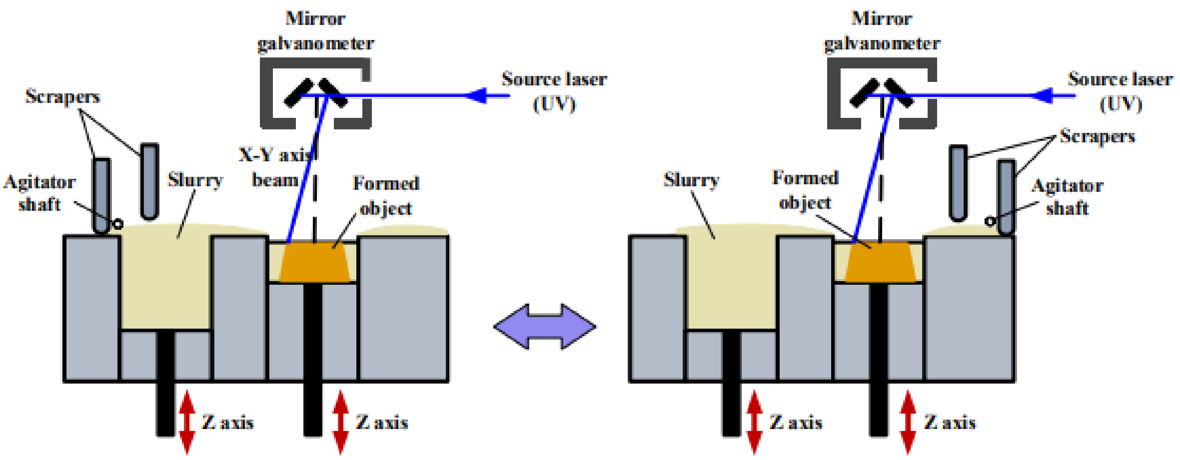

Figure 5. Illustration of stereolithography. Reproduced from [98] with open access from J. Phys.: Conf. Ser. Copyright 2021.

Table 3. Overview of biopolymer composite through stereolithography.

Reference	Material	Printing Technique	Mechanical Properties	Application	Year
[100]	P4HB/PHOH	SLA	N/A	Heart Valve	2002
[101]	45S5 Bioactive Glass	SLA	Tensile Strength: 37.9MPa	Scaffold	2021
[102]	PEGDA/AESO/nHA	SLA	Tensile Strength: 29.81±1.16MPa	Petroleum	2021
[103]	CMC/HA	SLA	N/A	Tissue Engineering	2018
[104]	PLLA/HA	SLA	Max. Strength: 16.6±0.9MPa 155.3±3.3MPa	Bone Scaffold	2022

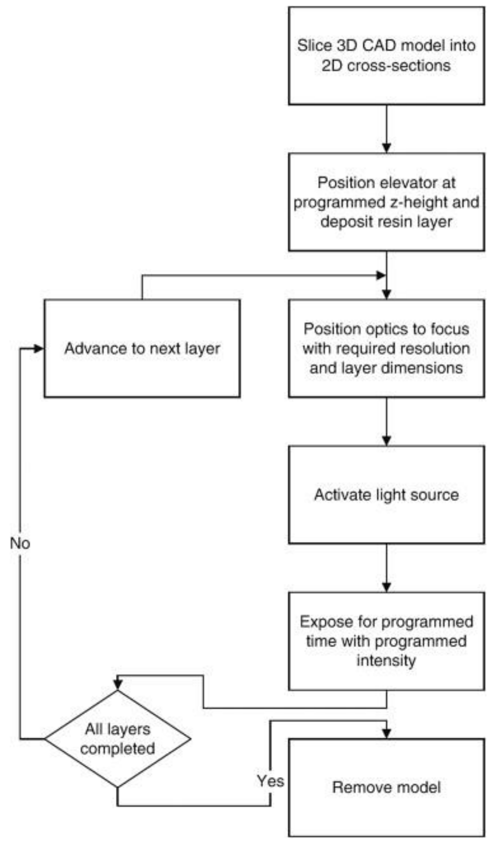

Figure 6. Flowchart of stereolithography. Reproduced from [99] with Elsevier Publishing. Copyright 2017.

2.4. Inkjet.

Inkjet three-dimensional (3D) printing is a quick, versatile, and cost-effective technique that allows the creation of basic and complex 3D items directly from Computer-Aided Design (CAD) data without the use of tooling. Inkjet 3D Printing is a subset of the Solid Freeform Fabrication family of manufacturing processes (SFF) [105]. The layer-by-layer deposition of 3D inkjet printing is comparable to other 3D printing technologies. Printing instructions are created from a 3D mechanical model of the product, exactly like in current fused deposition modeling (FDM) systems. This technique can deposit polymers, dielectric nanoparticles, and conductive nanoparticles, making it applicable to a wide range of materials [106].

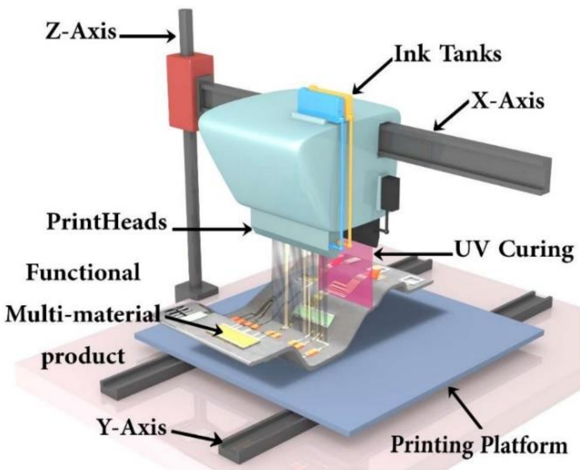

Figure 7. Illustration of inkjet printing. Reproduced from [107] with open access from Polymers—copyright 2022.

This approach uses an effective thermal field under the needle to improve ink viscosity, jetting morphological controllability, and printing structure solidification simultaneously. A steady and finer jet was produced under optimum circumstances, and various 3D structures were directly printed using this jet. Figures 7 and 8 depict illustrations and the mechanism of inkjet printing, while Table 4 shows the utilization of biopolymer.

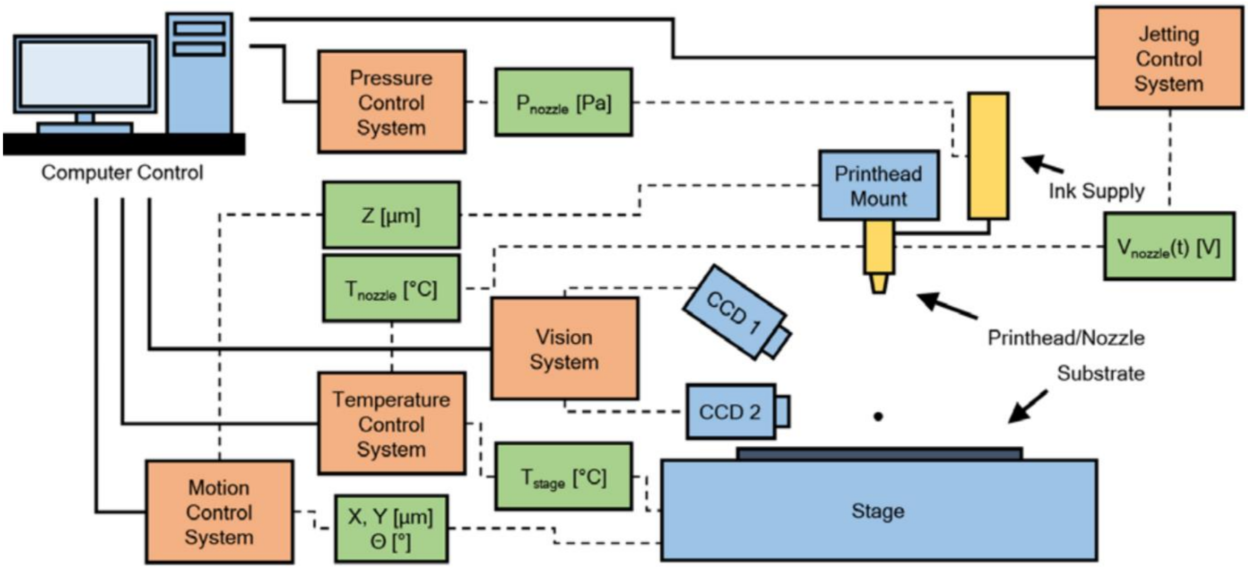

Figure 8. Mechanism of inkjet printing. Reproduced from [108] with open access from Polym. Polym. Compos. Copyright 2022.

Table 4. Overview of Biopolymer Composite Through Inkjet Printing.

Reference	Material	Printing Technique	Mechanical Properties	Application	Year
[109]	SWCNT/Ch	Ink-Jet	N/A	Cardiac Surgery	2021
[110]	PVA/kC	Ink-Jet	Young's Modulus: 10-150MPa	Cartilage Reconstruction	2022
[111]	Algal Polysaccharides	Ink-Jet	N/A	Biomedical Field	2022
[112]	PLA/MBG	Ink-Jet	Compressive Strength: 91.5MPa	Bone Tissue Engineering	2022
[113]	GEIMA/Nanoclay	Ink-Jet	Shear Viscosity: 0.037Pa.s-0.039Pa.s	Sustainable Inks	2020
[114]	Polysaccharides/CaP	Ink-Jet	N/A	Tissue Engineering	2009
[115]	PBAT/SCNP	Ink-Jet	Elastic Modulus: ~45MPa	Biodegradable Product	2017
[116]	PCL/HA/Core-shell	Ink-Jet	Shear Viscosity: ~50Pa.s	Bone Regeneration	2021
[117]	PCL/MWCNT/PEO	Ink-Jet	N/A	Sustainable Material	2018
[118]	Sodium Alginate/CaP	Ink-Jet	Compressive Strength: 0.45-1.0MPa	Bone Tissue Engineering	2022
[119]	PCL/MBG	Ink-Jet	N/A	Bone Tissue Engineering	2022

2.5. Others.

In this review research, some minor printing techniques are non-prominent in AM. Table 5 shows an overview of some minor 3D printing techniques for biopolymer composite. E-jet printing has been used to create different 3D scaffolds with prescribed architecture. E-jet printing is a micro/nano-manufacturing technique that employs an electric field to induce fluid jet printing using micro/nano-scale nozzles [120]. Then, CAWS is a hybrid of AM and wet spinning that enables fine control of fiber deposition using layer-by-layer technology and the production of an exterior structure or interior architecture [121]. After that, the syringe pump uses bioprinting using the same mechanism layer-by-layer [122]. Direct ink writing (DIW) is the most flexible AM method since it is suited for more materials and may be used to produce complex 3D objects from practically any material, such as ceramic, polymer, metal, or composite (or multi-material) structures. To achieve form fidelity, the ink materials should have high viscoelasticity before extrusion, a good shear-thinning effect when passing through a tiny nozzle, and high viscoelasticity after deposition [123]. Extrudability, form stability, structuration, and interlayer bonding are all buildability criteria for an E3DP building material [124]. Using viscous biopolymer solutions, electrohydrodynamic printing (EHDP) can precisely manipulate the position, size, and morphology of micro-/nano-fibers and fabricate high-resolution scaffolds [125]. In Material Extrusion Additive Manufacturing (MEX), the feedstock containing powder and polymeric binders is heated until the filament softens and can be extruded through a printing nozzle [126].

Table 5. Overview of biopolymer composite through other 3D printing.

Reference	Material	Printing Technique	Mechanical Properties	Application	Year
[127]	PCL/PVP	E-Jet	Thermal Platform: 30°C - 55°C loss	Scaffold	2020
[128]	PHBHHx/PCL	CAWS	Yielding Stress: 0.18-0.36MPa Compressive Strength: 0.16-0.39MPa	3D Porous Scaffold	2017
[129]	PVA/HA/Ch	Syringe pump	Elastic Modulus: 91.14MPa	Hard Tissue Engineering	2019
[130]	PCL/BG	DIW	Compressive Strength: 2.10MPa	Scaffold	2022
[131]	Plant Oil-Based Biopolymer/HA	DIW	Young's Modulus: 11MPa	Biomedical	2019
[132]	BSC	E3DP	Von Mises Yield: $0.435<\phi<0.548$	Construction	2021
[133]	PLA/Wood Fibre	MEX	Tensile Strength: 28.4±1.0MPa	Sustainable Filament	2017
[134]	PCL/Zein	EHDP	Shear Viscosity: 115.6±4.4Pa.s	Medical	2022

3. Challenges and Future Trends

The utilization of biopolymer composite materials has opened numerous opportunities to develop bio-products in a variety of industries. Only a few studies have been done thus far, even though using biopolymer composites and AM technology can provide enormous flexibility in designing and constructing incredibly complex bioproducts like bone scaffolds. The major reason is the numerous challenges in the biopolymer composite 3D printing process [135]. Firstly, they belong to the category of difficult-to-print materials, and the manufactured products often exhibit severe warpages following the printing process. The other difficulty is that 3D-printed items have poor mechanical qualities, making them unsuitable for applications where material Strength is critical. Advances in material and fabrication technologies that improve the printability of the material will be essential to the future success of 3D printing of biomaterials. Even though many researchers have studied biopolymer composites, it is clear that they have a promising future. Furthermore, the additive manufacturing of biopolymer composite is most commonly utilized in the biomedical area, and due to its high biodegradability, it is predicted to be used more in biomedical devices in the future.

4. Summary

Biopolymer composites offer great features such as biodegradability, biocompatibility, and nontoxicity, making them an ideal choice for biobased components, particularly in manufacturing biomedical equipment. When form complexity and customization are critical considerations, 3D printing technology has opened new doors for future uses of these biomaterials. The current state of 3D printing technology and the applications of additively generated biopolymer composite components in scientific and industrial sectors were evaluated. In summary, the FDM printing method is more common than other printing technologies because of the widespread availability of low-cost equipment and ease of processing. As a result, extensive research is required to assess and enhance the processability and final performance.

Funding

This research was supported by the Ministry of Higher Education (MOHE) through the Fundamental Research Grant Scheme (FRGS/1/2019/TK05/UTHM/03/5) and Universiti Tun Hussein Onn Malaysia (UTHM) through GPPS (vot Q244)

Acknowledgments

This research was supported by the Ministry of Higher Education (MOHE) through the Fundamental Research Grant Scheme (FRGS/1/2019/TK05/UTHM/03/5) and Universiti Tun Hussein Onn Malaysia (UTHM) through GPPS (vot Q244). The authors wish to acknowledge their appreciation to the Faculty of Mechanical and Manufacturing Engineering (FKMP), Universiti Tun Hussein Onn Malaysia (UTHM), for using the facilities.

Conflicts of Interest

The authors declare no conflict of interest.

References

1. Rouf, S.; Malik, A.; Singh, N.; Raina, A.; Naveed, N.; Siddiqui, M.I.H.; Haq, M.I.U. Additive manufacturing technologies: Industrial and medical applications. *SUSOC* **2022**, *3*, 258-274. https://doi.org/10.1016/j.susoc.2022.05.001.
2. Mehrpouya, M.; Dehghanghadikolaei, A.; Fotovvati, B.; Vosooghnia, A.; Emamian, S.S.; Gisario, A. The potential of additive manufacturing in the smart factory industrial 4.0: A review. *Appl. Sci.* **2019**, *9*, 1-34. https://doi.org/10.3390/app9183865.
3. Mehrpouya, M.; Vosooghnia, A.; Dehghanghadikolaei, A.; Fotovvati, B. The benefits of additive manufacturing for sustainable design and production. In Sustainable manufacturing, 1st ed.; Gupta, K., Salonitis, K., Elsevier: Amsterdam, Netherlands, **2021**, *1*, pp. 22-59. https://doi.org/10.1016/B978-0-12-818115-7.00009-2.
4. Alammar, A.; Kois, J.C.; Revilla-León, M.; Att, W. Additive manufacturing technologies: Current status and future perspectives. *J Prosthodont* **2022**, *31*, 4-12. https://doi.org/10.1111/jopr.13477.
5. Najah, M.I.; Aisyah, R.; Adzila, S.; Haq, R. Mechanical properties of calcium phosphate reinforced polyhydroxyalkanoate (PHA) biocomposite. *J. Thermoplast. Compos. Mater.* **2022**, *0*, 1-18. https://doi.org/10.1177/08927057221128188.
6. Adzila, S.; Mustaffa, N. A.; Kanasan, N. Magnesium-doped calcium phosphate/sodium alginate biocomposite for bone implant application. *J. Aust. Ceram. Soc.* **2020**, *56*, 109-115, https://doi.org/10.1007/s41779-019-00417-4.

7. Razak, A.; Isa, N.M; Adzila, S. Synthesis of calcium phosphate extracted from eggshell waste through precipitation method. *Biointerface Res. Appl. Chem* **2021**, *11*, 15058-1506, https://doi.org/10.33263/BRIAC116.1505815067.
8. Razak, S.A., Isa, N.A.A.M. and Bakar, S.A.S.A., 2022. Review on eggshell waste in tissue engineering application. *Int. J. Integr. Eng.* **2022**, *14*, 64-80. https://doi.org/10.30880/ijie.2022.14.04.007.
9. Najah, M.I.; Razak, A.; Che Shukur Nekmat, N.A.; Adzila, S.; Othman, R; Nordin, N. Characterization of calcium carbonate extracted from eggshell waste at various calcination temperature. *IJETER* **2020**, *8*, 6725-6731, http://www.warse.org/IJETER/static/pdf/file/ijeter168102020.pdf.
10. Ngo, T.D.; Kashani, A.; Imbalzano, G.; Nguyen, K.T.; Hui, D. Additive manufacturing (3D printing): A review of materials, methods, applications and challenges. *Compos. B. Eng.* **2018**, *143*, 172-196. https://doi.org/10.1016/j.compositesb.2018.02.012.
11. Nanda, S.; Patra, B.R.; Patel, R.; Bakos, J.; Dalai, A.K. Innovations in applications and prospects of bioplastics and biopolymers: a review. *Environ. Chem. Lett* **2022**, *20*, 379-395. https://doi.org/10.1007/s10311-021-01334-4.
12. Shahrubudin, N.; Koshy, P.; Alipal, J.; Kadir, M.H.A.; Lee, T.C. Challenges of 3D printing technology for manufacturing biomedical products: A case study of Malaysian manufacturing firms. *Heliyon* **2020**, *6*, 1-14. https://doi.org/10.1016/j.heliyon.2020.e03734.
13. Chia, H.N.; Wu, B.M. (2015). Recent advances in 3D printing of biomaterials. *J. Biol. Eng.* **2015**, *9*, 1-14. https://doi.org/10.1186/s13036-015-0001-4.
14. Chiulan, I.; Frone, A.N.; Brandabur, C.; Panaitescu, D.M. Recent advances in 3D printing of aliphatic polyesters. *Bioengineering* **2017**, *5*, 1-18. https://doi.org/10.3390/bioengineering5010002.
15. Mehrpouya, M.; Gisario, A.; Azizi, A.; Barletta, M. Investigation on shape recovery of 3D printed honeycomb sandwich structure. *Polym. Adv. Technol.* **2020**, *31*, 3361-3365. https://doi.org/10.1002/pat.5020.
16. Mehrpouya, M.; Azizi, A.; Janbaz, S.; Gisario, A. Investigation on the functionality of thermoresponsive origami structures. *Adv. Eng. Mater.* **2020**, *22*, 1-9. https://doi.org/10.1002/adem.202000296.
17. Hsueh, M.H.; Lai, C.J.; Liu, K.Y.; Chung, C.F.; Wang, S.H.; Pan, C.Y.; Zeng, Y.S. Effects of printing temperature and filling percentage on the mechanical behavior of fused deposition molding technology components for 3D printing. *Polymers* **2021**, *13*, 1-14. https://doi.org/10.3390/polym13172910.
18. Blaj, M., & Oancea, G. Fused deposition modeling process: a literature review. *IOP Conference Series: Materials Science and Engineering*, **2021**, *1009*(1), 12006. https://doi.org/10.1088/1757-899X/1009/1/012006.
19. Gao, X.; Yu, N.; Li, J. Influence of printing parameters and filament quality on structure and properties of polymer composite components used in the fields of automotive. In Structure and properties of additive manufactured polymer components, 1st ed.; Friedrich, K., Walter, R., Soutis, C., Advani, S.G., Fiedler, B., Woodhead Publishing: Sawston, United Kingdom, **2020**, *1*, pp. 303-330. https://doi.org/10.1016/B978-0-12-819535-2.00010-7.
20. Jo, J.; Kim, H.; Jeong, S.Y.; Park, C.; Hwang, H.S.; Koo, B. Changes in mechanical properties of polyhydroxyalkanoate with double silanized cellulose nanocrystals using different organosiloxanes. *Nanomaterials* **2021**, *11*, 1-17. https://doi.org/10.3390/nano11061542.
21. Wu, C.S. Characterization, functionality and application of siliceous sponge spicules additive-based manufacturing biopolymer composites. *Addit. Manuf.* **2018**, *22*, 13-20. https://doi.org/10.1016/j.addma.2018.04.034.
22. Vaidya, A.A.; Collet, C.; Gaugler, M.; Lloyd-Jones, G. Integrating softwood biorefinery lignin into polyhydroxybutyrate composites and application in 3D printing. *Mater. Today Commun* **2019**, *19*, 286-296. https://doi.org/10.1016/j.mtcomm.2019.02.008.
23. Valentini, F.; Dorigato, A.; Rigotti, D.; Pegoretti, A. Polyhydroxyalkanoates/fibrillated nanocellulose composites for additive manufacturing. *J. Polym. Environ.* **2019**, *27*, 1333-1341. https://doi.org/10.1007/s10924-019-01429-8.
24. Frone, A.N.; Batalu, D.; Chiulan, I.; Oprea, M.; Gabor, A.R.; Nicolae, C.A.; Panaitescu, D.M. Morpho-structural, thermal and mechanical properties of PLA/PHB/Cellulose biodegradable nanocomposites obtained by compression molding, extrusion, and 3d printing. *Nanomaterials* **2019**, *10*, 1-20. https://doi.org/10.3390/nano10010051.
25. Tian, J.; Zhang, R.; Wu, Y.; Xue, P. Additive manufacturing of wood flour/polyhydroxyalkanoates (PHA) fully bio-based composites based on micro-screw extrusion system. *Mater. Des.* **2021**, *199*, 1-14. https://doi.org/10.1016/j.matdes.2020.109418.

26. Bon, S.B.; Chiesa, I.; Morselli, D.; Degli Esposti, M.; Fabbri, P.; De Maria, C.; Valentini, L. Printable smart 3D architectures of regenerated silk on poly (3-hydroxybutyrate-co-3-hydroxyvalerate). *Mater. Des.* **2021**, *201*, 1-9. https://doi.org/10.1016/j.matdes.2021.109492.
27. Frone, A.N.; Batalu, D.; Chiulan, I.; Oprea, M.; Gabor, A.R.; Nicolae, C. A.; Panaitescu, D.M. Morpho-structural, thermal and mechanical properties of PLA/PHB/Cellulose biodegradable nanocomposites obtained by compression molding, extrusion, and 3d printing. *Nanomaterials* **2019**, *10*, 1-20. https://doi.org/10.3390/nano10010051.
28. Kolan, K.; Liu, Y.; Baldridge, J.; Murphy, C.; Semon, J.; Day, D.; Leu, M. Solvent based 3D printing of biopolymer/bioactive glass composite and hydrogel for tissue engineering applications. *Procedia CIRP* **2017**, *65*, 38-43. https://doi.org/10.1016/j.procir.2017.04.022.
29. Przekop, R.E.; Kujawa, M.; Pawlak, W.; Dobrosielska, M.; Sztorch, B.; Wieleba, W. Graphite modified polylactide (PLA) for 3D printed (FDM/FFF) sliding elements. *Polymers* **2020**, *12*, 1-22. https://doi.org/10.3390/POLYM12061250.
30. Kanabenja, W.; Passarapark, K.; Subchokpool, T.; Nawaaukkaratharnant, N.; Román, A.J.; Osswald, T.A.; Potiyaraj, P. 3D printing filaments from plasticized Polyhydroxybutyrate/Polylactic acid blends reinforced with hydroxyapatite. *Addit. Manuf.* **2022**, *59*, 1-9. https://doi.org/10.1016/j.addma.2022.103130.
31. Oladapo, B.I.; Ismail, S.O.; Zahedi, M.; Khan, A.; Usman, H. 3D printing and morphological characterisation of polymeric composite scaffolds. *Eng. Struct.* **2020**, *216*, 1-11. https://doi.org/10.1016/j.engstruct.2020.110752.
32. Sun, F.; Sun, X.; Wang, H.; Li, C.; Zhao, Y.; Tian, J.;Lin, Y. Application of 3D-printed, PLGA-based scaffolds in bone tissue engineering. *Int. J. Mol. Sci.* **2022**, *23*, 1-15. https://doi.org/10.3390/ijms23105831.
33. Jiao, Z.; Luo, B.; Xiang, S.; Ma, H.; Yu, Y.; Yang, W. 3D printing of HA/PCL composite tissue engineering scaffolds. *Adv. Ind. Eng. Polym. Res.* **2019**, *2*, 196-202. https://doi.org/10.1016/j.aiepr.2019.09.003.
34. Wang, Q.; Ye, W.; Ma, Z.; Xie, W.; Zhong, L.; Wang, Y.; Rong, Q. 3D printed PCL/β-TCP cross-scale scaffold with high-precision fiber for providing cell growth and forming bones in the pores. *Mater. Sci. Eng. C* **2021**, *127*, 1-7. https://doi.org/10.1016/j.msec.2021.112197.
35. Park, H.C.; Ryu, J.; Jung, S.; Park, H.J.; Oh, H.K.; Kook, M. S. Effect of hydroxyapatite nanoparticles and nitrogen plasma treatment on osteoblast biological behaviors of 3D-printed HDPE scaffold for bone tissue regeneration applications. *Materials* **2022**, *15*, 1-13. https://doi.org/10.3390/ma15030827.
36. Wang, W.; Zhang, B.; Li, M.; Li, J.; Zhang, C.; Han, Y.; Zhang, X. 3D printing of PLA/n-HA composite scaffolds with customized mechanical properties and biological functions for bone tissue engineering. *Compos. Part B Eng.* **2021**, *224*, 1-12. https://doi.org/10.1016/j.compositesb.2021.109192.
37. Oladapo, B.I.; Zahedi, S.A.; Ismail, S.O.; Omigbodun, F.T.; Bowoto, O.K.; Olawumi, M.A.; Muhammad, M. A. 3D printing of PEEK–cHAp scaffold for medical bone implant. *Bio-Design Manuf.* **2021**, *4*, 44-59. https://doi.org/10.1007/s42242-020-00098-0.
38. Won, J.Y.; Park, C.Y.; Bae, J.H.; Ahn, G.; Kim, C.; Lim, D.H.; Huh, J.B. Evaluation of 3D printed PCL/PLGA/β-TCP versus collagen membranes for guided bone regeneration in a beagle implant model. *Biomed. Mater.* **2016**, *11*, 1-16. https://doi.org/10.1088/1748-6041/11/5/055013.
39. Le Duigou, A.; Barbé, A., Guillou, E.; Castro, M. 3D printing of continuous flax fiber reinforced biocomposites for structural applications. *Mater. Des.* **2019**, *180*, 1-8. https://doi.org/10.1016/j.matdes.2019.107884.
40. Baligidad, S.M.; Kumar, G.C.; Maharudresh, A.C.; Lekshmi, I.C.; Rajasree, S.; Pillai, R. Investigation on strain rate sensitivity of 3D printed sPEEK-HAP/rGO composites. *J. Manuf. Process.* **2022**, *79*, 789-802. https://doi.org/10.1016/j.jmapro.2022.05.025.
41. Singh, S.; Singh, G.; Prakash, C.; Ramakrishna, S.; Lamberti, L; Pruncu, C.I. 3D printed biodegradable composites: An insight into mechanical properties of PLA/chitosan scaffold. *Polym. Test.* **2020**, *89*, 1-13. https://doi.org/10.1016/j.polymertesting.2020.106722.
42. Kumar, S.; Singh, R.; Singh, T.P.; Batish, A. Mechanical and morphological properties correlation of PLA-PVC-Wood Powder-Fe3O4 composite matrix for 3D printing. In Additive, subtractive, and hybrid technologies: Recent innovations in manufacturing, 1st ed.; Prakash, C., Singh, S., Ramakrishna, S., Springer International Publishing: Berlin, Germany, **2022**, 1, pp. 51-58. https://doi.org/10.1007/978-3-030-99569-0_4.
43. Narlıoğlu, N.; Salan, T.; Alma, M.H. Properties of 3D-Printed wood sawdust-reinforced PLA composites. *BioResources* **2021**, *16*, 5467-5480. https://doi.org/10.15376/biores.16.3.5467-5480.
44. Salamanca, E.; Tsao, T.C.; Hseuh, H.W.; Wu, Y.F.; Choy, C.S.; Lin, C.K.; Pan, Y.H.; Teng, N.C.; Huang, M.C.; Lin, S.M.; Chang, W.J. Fabrication of Polylactic Acid/β-Tricalcium Phosphate FDM 3D Printing fiber

to enhance osteoblastic-like cell performance. *Front. Mater.* **2021**, *8*, 1-10. https://doi.org/10.3389/fmats.2021.683706.

45. Skonta, A.; Gkantzou, E.; Spyrou, K.; Spyrou, S.; Polydera, A.; Gournis, D.; Stamatis, H. 3D Printed Polylactic Acid (PLA) well plates for enzyme inhibition studies: The case of pancreatic lipase. *Catal. Res.* **2022**, *2*, 1-19. https://doi.org/10.21926/cr.2203032.
46. Donate, R.; Alemán-Domínguez, M.E.; Monzón, M.; Yu, J.; Rodríguez-Esparragón, F; Liu, C. Evaluation of Aloe vera coated polylactic acid scaffolds for bone tissue engineering. *Appl. Sci.* **2020**, *10*, 1-15. https://doi.org/10.3390/app10072576.
47. Hwangbo, H.; Lee, J.; Kim, G. Mechanically and biologically enhanced 3D-printed HA/PLLA/dECM biocomposites for bone tissue engineering. *Int. J. Biol. Macromol.* **2022**, *218*, 9-21. https://doi.org/10.1016/j.ijbiomac.2022.07.040.
48. Alam, F.; Shukla, V.R.; Varadarajan, K.M.; Kumar, S. Microarchitected 3D printed polylactic acid (PLA) nanocomposite scaffolds for biomedical applications. *J. Mech. Behav. Biomed. Mater.* **2020**, *103*, 1-9. https://doi.org/10.1016/j.jmbbm.2019.103576.
49. Pascual-González, C.**;** de la Vega, J.**;** Thompson, C.**;** Fernández-Blázquez, J.P.**;** Herráez-Molinero, D.**;** Biurrun, N.**;** Lizarralde, I.**;** Del Rio, J.S.**;** Gonzalez, C.**;** Llorca, J. Processing and mechanical properties of novel biodegradable poly-lactic acid/Zn 3D printed scaffolds for application in tissue regeneration. *J. Mech. Behav. Biomed. Mater.* **2022**, *132*, 1-10. https://doi.org/10.1016/j.jmbbm.2022.105290.
50. Rojas-Martínez, L.E.; Flores-Hernandez, C.G.; López-Marín, L.M.; Martinez-Hernandez, A.L.; Thorat, S.B.; Vasquez, C.R.; Del Rio-Castillo, A.E.; Velasco-Santos, C. 3D printing of PLA composites scaffolds reinforced with keratin and chitosan: Effect of geometry and structure. *Eur. Polym. J.* **2020**, *141*, 1-10. https://doi.org/10.1016/j.eurpolymj.2020.110088
51. Cakmak, A.M.; Unal, S.; Sahin, A.; Oktar, F.N.; Sengor, M.; Ekren, N.; Gunduz, O.; Kalaskar, D.M. 3D printed polycaprolactone/gelatin/bacterial cellulose/hydroxyapatite composite scaffold for bone tissue engineering. *Polymers* **2020**, *12*, 1-14. https://doi.org/10.3390/polym12091962.
52. Zhou, X.; Ren, Z.; Sun, H.; Bi, H.; Gu, T.; Xu, M. 3D printing with high content of lignin enabled by introducing polyurethane. *Int. J. Biol. Macromol.* **2022**, *221*, 1209-1217. https://doi.org/10.1016/j.ijbiomac.2022.09.076.
53. Mozafari, H.; Dong, P.; Hadidi, H.; Sealy, M.P.; Gu, L. Mechanical characterizations of 3D-printed PLLA/steel particle composites. *Materials* **2018**, *12*, 1-11. https://doi.org/10.3390/MA12010001.
54. Gu, S.; Ji, H.; Liang, K.; Ji, Y. Chitin nanocrystal based aqueous inks for 3D printing via direct ink writing. *J. Elastomers Plast.* **2022**, *54*, 922-936. https://doi.org/10.1177/00952443221102589.
55. Bajwa, D.; Eichers, M.; Shojaeiarani, J.; Kallmeyer, A. Influence of biobased plasticizers on 3D printed polylactic acid composites filled with sustainable biofiller. *Ind. Crops Prod.* **2021**, *173*, 1-8. https://doi.org/10.1016/j.indcrop.2021.114132.
56. Khakestani, M.; Jafari, S.H.; Zahedi, P.; Bagheri, R.; Hajiaghaee, R. Physical, morphological, and biological studies on PLA/n HA composite nanofibrous webs containing Equisetum arvense herbal extract for bone tissue engineering. *J. Appl. Polym. Sci.* **2017**, *134*, 1-10. https://doi.org/10.1002/app.45343.
57. Peric, M.; Putz, R.; Paulik, C. 3D-Printed PLA Filaments Reinforced with Nanofibrillated Cellulose. *J. Renew. Mater.* **2020**, *8*, 759-772. https://doi.org/10.32604/jrm.2020.09284.
58. Coppola, B.; Cappetti, N.; Di Maio, L.; Scarfato, P.; Incarnato, L. (2018). 3D printing of PLA/clay nanocomposites: Influence of printing temperature on printed samples properties. *Materials* **2018**, *11*, 1-17. https://doi.org/10.3390/ma11101947.
59. Liu, W.; Wu, N.; Pochiraju, K. Shape recovery characteristics of SiC/C/PLA composite filaments and 3D printed parts. *Compos. Part A Appl. Sci. Manuf.* **2018**, *108*, 1-11. https://doi.org/10.1016/j.compositesa.2018.02.017.
60. Masarra, N.A.; Batistella, M.; Quantin, J.C.; Regazzi, A.; Pucci, M.F.; El Hage, R; Lopez-Cuesta, J.M. Fabrication of PLA/PCL/Graphene nanoplatelet (GNP) electrically conductive circuit using the Fused Filament Fabrication (FFF) 3D printing technique. *Materials* **2022**, *15*, 1-27. https://doi.org/10.3390/ma15030762.
61. Bhagia, S.; Lowden, R.R.; Erdman III, D.; Rodriguez Jr, M.; Haga, B.A.; Solano, I.R.M.; Gallego, N.C.; Pu, Y.; Muchero, W.; Kunc, V; Ragauskas, A.J. Tensile properties of 3D-printed wood-filled PLA materials using poplar trees. *Appl. Mater. Today* **2020**, *21*, 1-9. https://doi.org/10.1016/j.apmt.2020.100832.

62. Sun, Y.; Wang, Y.; Mu, W.; Zheng, Z.; Yang, B.; Wang, J.; Zhang, R.; Zhou, K.; Chen, L.; Ying, J.; Liu, X. Mechanical properties of 3D printed micro-nano rice husk/polylactic acid filaments. *J. Appl. Polym. Sci.* **2022**, *139*, 1-13. https://doi.org/10.1002/app.52619.
63. Estakhrianhaghighi, E.; Mirabolghasemi, A.; Zhang, Y.; Lessard, L; Akbarzadeh, A. 3D-printed wood-fiber reinforced architected cellular composites. Adv. Eng. Mater. **2020**, *22*, 1-14. https://doi.org/10.1002/adem.202000565.
64. Liao, J.; Brosse, N.; Pizzi, A.; Hoppe, S.; Zhou, X.; Du, G. Characterization and 3D printability of poly (lactic acid)/acetylated tannin composites. *Ind. Crops Prod.* **2020**, *149*, 1-9. https://doi.org/10.1016/j.indcrop.2020.112320.
65. Kumar, S.; Singh, R.; Singh, M.; Singh, T.P.; Batish, A. Multi material 3D printing of PLA-PA6/TiO_2 polymeric matrix: Flexural, wear and morphological properties. *J. Thermoplast. Compos. Mater.* **2022**, *35*, 2105-2124. https://doi.org/10.1177/0892705720953193.
66. Fico, D.; Rizzo, D.; De Carolis, V.; Montagna, F.; Palumbo, E.; Corcione, C.E. Development and characterization of sustainable PLA/Olive wood waste composites for rehabilitation applications using Fused Filament Fabrication (FFF). *J. Build. Eng.* **2022**, *56*, 1-15. https://doi.org/10.1016/j.jobe.2022.104673.
67. Jiang, G.; Yang, T.; Xu, J.; Tao, D.; Luo, C.; Wang, C.; Dong, Q.; Wang, Y.; 2020. Investigation into hydroxypropyl-methylcellulose-reinforced polylactide composites for fused deposition modeling. *Ind. Crops Prod.* **2020**, *146*, 1-8. https://doi.org/10.1016/j.indcrop.2020.112174.
68. Hamidi, A.; Jain, S.; Tadesse, Y. 3D printing PLA and silicone elastomer structures with sugar solution support material. *EAPAD* **2017,** *10163*, 159-166. https://doi.org/10.1117/12.2258689.
69. Giubilini, A.; Siqueira, G.; Clemens, F.J.; Sciancalepore, C.; Messori, M.; Nyström, G.; Bondioli, F. 3D-printing nanocellulose-poly (3-hydroxybutyrate-co-3-hydroxyhexanoate) biodegradable composites by fused deposition modeling. *ACS Sustain. Chem. Eng.* **2020**, *8*, 10292-10302. https://doi.org/10.1021/acssuschemeng.0c03385.
70. Veeman, D.; Palaniyappan, S. Process optimisation on the compressive strength property for the 3D printing of PLA/almond shell composite. *J. Thermoplast. Compos. Mater.* **2022**, *0*, 1-15. https://doi.org/10.1177/08927057221092327.
71. Obielodan, J.; Vergenz, K.; Aqil, D.; Wu, J; Ellistrem, L.M.; 2019. Characterization of PLA/lignin biocomposites for 3D printing. *Solid Free. Fabr. 2019 Proc. 30th Annu. Int. Solid Free. Fabr. Symp. - An Addit. Manuf. Conf. SFF* **2019**, *2019*, 998-1007. http://dx.doi.org/10.26153/tsw/17335.
72. Singamneni, S.; Smith, D.; LeGuen, M.J.; Truong, D.; 2018. Extrusion 3D printing of polybutyrate-adipate-terephthalate-polymer composites in the pellet form. *Polymers* **2018**, *10*, 922-935. https://doi.org/10.3390/polym10080922.
73. Ertane, E.G.; Dorner-Reisel, A.; Baran, O.; Welzel, T.; Matner, V.; Svoboda, S. Processing and wear behaviour of 3D printed PLA reinforced with biogenic carbon. *Adv. Tribol.* **2018**, *2018*, 1-12. https://doi.org/10.1155/2018/1763182.
74. Xiao, X.; Chevali, V.S.; Song, P.; He, D.; Wang, H. Polylactide/hemp hurd biocomposites as sustainable 3D printing feedstock. *Compos. Sci. Technol.* **2019**, *184*, 1-8. https://doi.org/10.1016/j.compscitech.2019.107887.
75. Maleki-Ghaleh, H.; Siadati, M.H.; Fallah, A.; Zarrabi, A.; Afghah, F.; Koc, B.; Abdolahinia, E.D.; Omidi, Y.; Barar, J.; Akbari-Fakhrabadi, A.; Beygi-Khosrowshahi, Y. Effect of zinc-doped hydroxyapatite/graphene nanocomposite on the physicochemical properties and osteogenesis differentiation of 3D-printed polycaprolactone scaffolds for bone tissue engineering. *Chem. Eng. J.* **2021**, *426*, 1-16. https://doi.org/10.1016/j.cej.2021.131321.
76. Nasrin, A.; Hassan, M.; Mirabet, M.M.; Windhab, N.; Gomes, V.G. 3D-printed bioresorbable poly (lactic-co-glycolic acid) and quantum-dot nanocomposites: Scaffolds for enhanced bone mineralization and inbuilt co-monitoring. *J. Biomed. Mater. Res. Part A* **2022**, *110*, 916-927. https://doi.org/10.1002/jbm.a.37340.
77. Sciancalepore, C.; Togliatti, E.; Marozzi, M.; Rizzi, F.M.A.; Pugliese, D.; Cavazza, A.; Pitirollo, O.; Grimaldi, M.; Milanese, D. Flexible PBAT-Based Composite Filaments for Tunable FDM 3D Printing. *ACS Appl. Bio Mater.* **2022**, *5*, 3219-3229. https://doi.org/10.1021/acsabm.2c00203.
78. Zhang, W.; Ullah, I.; Shi, L.; Zhang, Y.; Ou, H.; Zhou, J.; Ullah, M.W.; Zhang, X.; Li, W. Fabrication and characterization of porous polycaprolactone scaffold via extrusion-based cryogenic 3D printing for tissue engineering. *Mater. Des.* **2019**, *180*, 1-10. https://doi.org/10.1016/j.matdes.2019.107946.
79. Ádám, B.; Weltsch, Z. Thermal and mechanical assessment of PLA-SEBS and PLA-SEBS-CNT biopolymer blends for 3D Printing. *Appl. Sci.* **2021**, *11*, 1-18. https://doi.org/10.3390/app11136218.

80. Song, X.; He, W.; Yang, S.; Huang, G.; Yang, T. Fused deposition modeling of poly (lactic acid)/walnut shell biocomposite filaments—Surface treatment and properties. *Appl. Sci.* **2019**, *9*, 1-17. http://dx.doi.org/10.3390/app9224892.
81. Filgueira, D.; Holmen, S.; Melbø, J.K.; Moldes, D.; Echtermeyer, A.T.; Chinga-Carrasco, G. 3D printable filaments made of biobased polyethylene biocomposites. *Polymers* **2018**, *10*, 1-15. https://doi.org/10.3390/polym10030314.
82. Dan, L.; Cheng, Q.; Narain, R.; Krause, B.; Pötschke, P.; Elias, A. Three-dimensional printed and biocompatible conductive composites comprised of polyhydroxybutyrate and multiwalled carbon nanotubes. *Ind. Eng. Chem. Res.* **2021**, *60*, 885-897. https://doi.org/10.1021/acs.iecr.0c04753.
83. Xu, W.; Provinch, A.; Uppstu, P.; Kronlund, D.; Moritzd, N.; Hemming, J.; Preis, M.; Sandler, N.; Stefan, W.; Xu, C.; Three dimensional printing of polylactic acid (PLA)-wood polysaccharide composite. *Turun yliopisto* **2017,** *2017*, 1-35, https://doi.org/10.1016/j.carbpol.2018.01.069.
84. Ohaeri, O.; Cree, D. Development and Characterization of PHB-PLA/Corncob Composite for Fused Filament Fabrication. *J. Compos. Sci.* **2022**, *6*, 1-22. https://doi.org/10.3390/jcs6090249.
85. Zhu, M.; He, X.; Xin, C.; Zhu, Y.; Liu, Z. 3D printing of an integrated triphasic MBG-alginate scaffold with enhanced interface bonding for hard tissue applications. *J. Mater. Sci. Mater. Med.* **2020**, *31*, 1-8. https://doi.org/10.1007/s10856-020-06459-6.
86. Kruth, J.P.; Wang, X.; Laoui, T.; Froyen, L., 2003. Lasers and materials in selective laser sintering. *Assem. Autom.* **2003**, *23*, 357-371. https://doi.org/10.1108/01445150310698652.
87. Riza, S.H.; Masood, S.H.; Rashid, R.A.R.; Chandra, S., 2020. Selective laser sintering in biomedical manufacturing. In Metallic biomaterials processing and medical device manufacturing, 1st ed.; Wen, C., Woodhead Publishing: Sawston, United Kingdom, **2020**; *1*, pp. 193-233. https://doi.org/10.1016/B978-0-08-102965-7.00006-0.
88. Yang, Y.; Wang, G.; Liang, H.; Gao, C.; Peng, S.; Shen, L.; Shuai, C. Additive manufacturing of bone scaffolds. *Int. J. bioprinting* **2019**, *5*, 1-25. https://doi.org/10.18063/IJB.v5i1.148.
89. Chen, G.Q.; Zhang, J. 2018. Microbial polyhydroxyalkanoates as medical implant biomaterials. *Artif. cells, nanomedicine, Biotechnol.* **2018**, *46*, 1-18. https://doi.org/10.1080/21691401.2017.1371185.
90. Han, W., Kong, L., & Xu, M. Advances in selective laser sintering of polymers. *International Journal of Extreme Manufacturing*, **2022**, *4*(4), 42002. https://doi.org/10.1088/2631-7990/ac9096.
91. Psarommatis, F.; Vosniakos, G.C. Systematic development of a powder deposition system for an open selective laser sintering machine using analytic hierarchy process. *J. Manuf. Mater. Process.* **2022**, *6*, 1-24. https://doi.org/10.3390/jmmp6010022.
92. Duan, B.; Wang, M., 2010. Customized Ca–P/PHBV nanocomposite scaffolds for bone tissue engineering: design, fabrication, surface modification and sustained release of growth factor. *J. R. Soc. Interface* **2010**, *7*, 615-629. https://doi.org/10.1098/rsif.2010.0127.focus.
93. Babilotte, J.; Martin, B.; Guduric, V.; Bareille, R.; Agniel, R.; Roques, S.; Héroguez, V.; Dussauze, M.; Gaudon, M.; Le Nihouannen, D.; Catros, S. Development and characterization of a PLGA-HA composite material to fabricate 3D-printed scaffolds for bone tissue engineering. *Mater. Sci. Eng. C* **2021**, *118*, 1-13. https://doi.org/10.1016/j.msec.2020.111334.
94. Chen, S.G.; Yang, J.; Jia, Y.G.; Lu, B.; Ren, L. TiO_2 and PEEK reinforced 3D printing PMMA composite resin for dental denture base applications. *Nanomaterials* **2019**, *9*, 1-18. https://doi.org/10.3390/nano9071049.
95. Vahabi, H.; Laoutid, F.; Mehrpouya, M.; Saeb, M.R.; Dubois, P. Flame retardant polymer materials: An update and the future for 3D printing developments. *Mater. Sci. Eng. R* **2021**, *144*, 1-24. https://doi.org/10.1016/j.mser.2020.100604.
96. Huang, J.; Qin, Q.; Wang, J. A review of stereolithography: Processes and systems. *Processes* **2020**, *8*, 1-16. https://doi.org/10.3390/pr8091138.
97. Stevens, B.; Yang, Y.; Mohandas, A.; Stucker, B.; Nguyen, K.T. A review of materials, fabrication methods, and strategies used to enhance bone regeneration in engineered bone tissues. *J. Biomed. Mater. Res. - Part B Appl. Biomater* **2008**, *85*, 573-582. https://doi.org/10.1002/jbm.b.30962.
98. Chen, H., Chen, B., Li, J., Yan, H., Cheng, L., Chen, J., & Zeng, Y. The influence of SLA process parameters on the property of alumina. *Journal of Physics: Conference Series*, **2021**, *1965*(1), 12112. https://doi.org/10.1088/1742-6596/1965/1/012112.
99. Ozbolat, I. T. Laser-Based bioprinting with minor contributions by Hemanth Gudapati, The Pennsylvania State University. *3D Bioprinting* **2017**, *6*, 165-197. https://doi.org/10.1016/b978-0-12-803010-3.00006-8.

100. Sodian, R.; Loebe, M.; Hein, A.; Martin, D.P.; Hoerstrup, S.P.; Potapov, E.V.; Hausmann, H.; Lueth, T.; Hetzer, R. Application of stereolithography for scaffold fabrication for tissue engineered heart valves. *ASAIO J* **2002** *48*, 12-16. https://doi.org/10.1097/00002480-200201000-00004.
101. Aráoz, B.; Karakaya, E.; González Wusener, A.; Detsch, R.; Bizzotto, J.; Gueron, G.; Boccaccini, A.R.; Hermida, É.B. 3D printed poly (hydroxybutyrate-co-hydroxyvalerate)-45S5 bioactive glass composite resorbable scaffolds suitable for bone regeneration. *J. Mater. Res.* **2021**, *36*, 4000-4012. https://doi.org/10.1557/s43578-021-00272-9.
102. Mondal, D.; Haghpanah, Z.; Huxman, C.J.; Tanter, S.; Sun, D.; Gorbet, M.; Willett, T.L. mSLA-based 3D printing of acrylated epoxidized soybean oil-nano-hydroxyapatite composites for bone repair. *Mater. Sci. Eng. C* **2021**, *130*, 1-11. https://doi.org/10.1016/j.msec.2021.112456.
103. Sayed, M.; El-Maghraby, H.F.; Bondioli, F.; Naga, S.M.; 2018. 3D carboxymethyl cellulose/hydroxyapatite (CMC/HA) scaffold composites based on recycled eggshell. *J. Appl. Pharm. Sci.* **2018**, *8*, 23-30. https://doi.org/10.7324/JAPS.2018.8304.
104. Gang, F.; Ye, W.; Ma, C.; Wang, W.; Xiao, Y.; Liu, C.; Sun, X. 3D printing of PLLA/biomineral composite bone tissue engineering scaffolds. *Materials* **2022**, *15*, p.4280. https://doi.org/10.3390/ma15124280.
105. Napadensky, E. Inkjet 3D printing. The chemistry of inkjet inks. In The chemistry of inkjet inks, 1st ed.; Magdassi, S., World Scientific: Singapore, **2009**, *1*, pp.255-268. https://doi.org/10.1142/9789812818225.
106. Kumar, N.; Jain, P.K.; Tandon, P.; Pandey, P.M. Experimental investigations on suitability of polypropylene (PP) and ethylene vinyl acetate (EVA) in additive manufacturing. *Mater. Today: Proc.* **2018**, *5*, 4118-4127. https://doi.org/10.1016/j.matpr.2017.11.672.
107. Elkaseer, A.; Schneider, S.; Deng, Y.; Scholz, S.G. Effect of Process Parameters on the Performance of Drop-On-Demand 3D Inkjet Printing: Geometrical-Based Modeling and Experimental Validation. *Polymers* **2022**, *14*, 2557. https://doi.org/10.3390/polym14132557.
108. Safari, F.; Kami, A.; Abedini, V., 2022. 3D printing of continuous fiber reinforced composites: A review of the processing, pre-and post-processing effects on mechanical properties. *Polym. Polym. Compos.* **2022**, *30*, 1-26. https://doi.org/10.1177/09673911221098734.
109. Alagarsamy, K.N.; Mathan, S.; Yan, W.; Rafieerad, A.; Sekaran, S.; Manego, H.; Dhingra, S. Carbon nanomaterials for cardiovascular theranostics: Promises and challenges. *Bioact. Mater.* **2021**, *6*, 2261-2280. https://doi.org/10.1016/j.bioactmat.2020.12.030.
110. Muscolino, E.; Di Stefano, A.B.; Trapani, M.; Sabatino, M.A.; Giacomazza, D.; Alessi, S.; Cammarata, E.; Moschella, F.; Cordova, A.; Toia, F.; Dispenza, C. κ-Carrageenan and PVA blends as bioinks to 3D print scaffolds for cartilage reconstruction. *Int. J. Biol. Macromol.* **2022**, *222*, 1861-1875. https://doi.org/10.1016/j.ijbiomac.2022.09.275.
111. Mandal, S.; Nagi, G.K.; Corcoran, A.A.; Agrawal, R.; Dubey, M.; Hunt, R.W. Algal polysaccharides for 3D printing: A review. *Carbohydr. Polym.* **2022**, *300*, 1-18. https://doi.org/10.1016/j.carbpol.2022.120267.
112. Pant, S.; Thomas, S.; Loganathan, S.; Valapa, R.B. 3D bioprinted poly (lactic acid)/mesoporous bioactive glass based biomimetic scaffold with rapid apatite crystallization and in-vitro Cytocompatability for bone tissue engineering. *Int. J. Biol. Macromol.* **2022**, *217*, 979-997. https://doi.org/10.1016/j.ijbiomac.2022.07.202.
113. Cidonio, G.; Glinka, M.; Kim, Y.H.; Kanczler, J.M.; Lanham, S.A.; Ahlfeld, T.; Lode, A.; Dawson, J.I.; Gelinsky, M.; Oreffo, R.O. Nanoclay-based 3D printed scaffolds promote vascular ingrowth ex vivo and generate bone mineral tissue in vitro and in vivo. *Biofabrication* **2020**, *12*, 1-16. https://doi.org/10.1088/1758-5090/ab8753.
114. Grijpstra, J.; Gerwig, G.J.; Wösten, H.; Kamerling, J.P.; de Cock, H. Production of extracellular polysaccharides by CAP mutants of Cryptococcus neoformans. *Eukaryot. Cell* **2009**, *8*, 1165-1173. https://doi.org/10.1128/EC.00013-09.
115. Singamneni, S.; Smith, D.; LeGuen, M.J.; Truong, D. Extrusion 3D printing of polybutyrate-adipate-terephthalate-polymer composites in the pellet form. *Polymers* **2018**, *10*, 1-13. https://doi.org/10.3390/polym10080922.
116. Liu, C.; Wang, Z.; Wei, X.; Chen, B.; Luo, Y. 3D printed hydrogel/PCL core/shell fiber scaffolds with NIR-triggered drug release for cancer therapy and wound healing. *Acta Biomater.* **2021**, *131*, 314-325. https://doi.org/10.1016/j.actbio.2021.07.011.
117. Huang, B.; Vyas, C.; Roberts, I.; Poutrel, Q.A.; Chiang, W.H.; Blaker, J.J.; Huang, Z.; Bártolo, P. Fabrication and characterisation of 3D printed MWCNT composite porous scaffolds for bone regeneration. *Mater. Sci. Eng. C* **2019**, *98*, 266-278. https://doi.org/10.1016/j.msec.2018.12.100.

118. Wattanaanek, N.; Suttapreyasri, S.; Samruajbenjakun, B. 3D printing of calcium phosphate/calcium sulfate with alginate/cellulose-based scaffolds for bone regeneration: multilayer fabrication and characterization. *J. Funct. Biomater.* **2022**, *13*, 1-16. https://doi.org/10.3390/jfb13020047.
119. Feng, X.; Wu, Y.; Bao, F.; Chen, X.; Gong, J. Comparison of 3D-printed mesoporous calcium silicate/polycaprolactone and mesoporous Bioacive glass/polycaprolactone scaffolds for bone regeneration. *Microporous Mesoporous Mater.* **2019**, *278*, 348-353. https://doi.org/10.1016/j.micromeso.2019.01.007.
120. Yang, Y.; Qiao, X.; Huang, R.; Chen, H.; Shi, X.; Wang, J.; Tan, W.; Tan, Z. E-jet 3D printed drug delivery implants to inhibit growth and metastasis of orthotopic breast cancer. *Biomaterials* **2020**, *230*, 1-13. https://doi.org/10.1016/j.biomaterials.2019.119618.
121. Neves, S.C.; Mota, C.; Longoni, A.; Barrias, C.C.; Granja, P.L.; Moroni, L. Additive manufactured polymeric 3D scaffolds with tailored surface topography influence mesenchymal stromal cells activity. *Biofabrication* **2016**, *8*, 1-16. https://doi.org/10.1088/1758-5090/8/2/025012.
122. Wang, Z.; Xiang, L.; Lin, F.; Tang, Y.; Cui, W. 3D bioprinting of emulating homeostasis regulation for regenerative medicine applications. *J. Control. Release* **2023**, *353*, 147-165. https://doi.org/10.1016/j.jconrel.2022.11.035.
123. Yang, G.; Sun, Y.; Li, M.; Ou, K.; Fang, J.; Fu, Q. Direct-ink-writing (DIW) 3D printing functional composite materials based on supra-molecular interaction. *Compos. Sci. Technol.* **2021**, *215*, 1-10. https://doi.org/10.1016/j.compscitech.2021.109013.
124. Le, T.T.; Austin, S.A.; Lim, S.; Buswell, R.A.; Gibb, A.G.; Thorpe, T. Mix design and fresh properties for high-performance printing concrete. *Mater. Struct.* **2012**, *45*, 1221-1232. https://doi.org/10.1617/s11527-012-9828-z.
125. Sun, J.; Jing, L.; Fan, X.; Gao, X.; Liang, Y.C. Electrohydrodynamic printing process monitoring by microscopic image identification. *Int. J. Bioprinting* **2019**, *5*, 1-2. https://doi.org/10.18063/ijb.v5i1.164.
126. Vidakis, N.; Petousis, M.; Mountakis, N.; David, C.N.; Sagris, D.; Das, S.C. Thermomechanical response of thermoplastic polyurethane used in MEX additive manufacturing over repetitive mechanical recycling courses. *Polym. Degrad. Stab.* **2023**, *207*, 1-16. https://doi.org/10.1016/j.polymdegradstab.2022.110232.
127. Li, K.; Wang, D.; Zhao, K.; Song, K.; Liang, J. Electrohydrodynamic jet 3D printing of PCL/PVP composite scaffold for cell culture. *Talanta* **2020**, *211*, 1-11. https://doi.org/10.1016/j.talanta.2020.120750.
128. Puppi, D.; Morelli, A.; Chiellini, F. Additive manufacturing of poly (3-hydroxybutyrate-co-3-hydroxyhexanoate)/poly (ε-caprolactone) blend scaffolds for tissue engineering. *Bioeng.* **2017**, *4*, 1-17. https://doi.org/10.3390/bioengineering4020049.
129. Ergul, N.M.; Unal, S.; Kartal, I.; Kalkandelen, C.; Ekren, N.; Kilic, O.; Chi-Chang, L.; Gunduz, O. 3D printing of chitosan/poly (vinyl alcohol) hydrogel containing synthesized hydroxyapatite scaffolds for hard-tissue engineering. *Polym. Test.* **2019**, *79*, 1-10. https://doi.org/10.1016/j.polymertesting.2019.106006.
130. Ilyas, K., Akhtar, M.A., Ammar, E.B.; Boccaccini, A.R. Surface Modification of 3D-Printed PCL/BG composite scaffolds via mussel-inspired polydopamine and effective antibacterial coatings for biomedical applications. *Mater.* **2022**, *15*, 1-19. https://doi.org/10.3390/ma15238289.
131. Bahmani, A.; Comeau, P.A.; Montesano, J.; Willett, T.L. Extrudable hydroxyapatite/plant oil-based biopolymer nanocomposites for biomedical applications: Mechanical testing and modeling. *Mater. Des.***2019**, *174*, 1-11. https://doi.org/10.1016/j.matdes.2019.107790.
132. Biggerstaff, A.; Fuller, G.; Lepech, M.; Loftus, D. Determining the yield stress of a Biopolymer-bound soil composite for extrusion-based 3D printing applications. Constr. Build. Mater. **2021**, *305*, 1-8. https://doi.org/10.1016/j.conbuildmat.2021.124730.
133. Kariz, M.; Sernek, M.; Obućina, M.; Kuzman, M.K. Effect of wood content in FDM filament on properties of 3D printed parts. *Mater. Today Commun.* **2018**, *14*, 135-140. https://doi.org/10.1016/j.mtcomm.2017.12.016.
134. Subuki, I.; Nasir, K.N.A.; Ramlee, N.A. A Review on the effect of zein in scaffold for bone tissue engineering. *Pertanika J. Sci. Technol.* **2022**, *30*, 2805-2829. https://doi.org/10.47836/pjst.30.4.28.
135. Díez-Pascual, A.M. Synthesis and applications of biopolymer composites. *Int. J. Mol. Sci.* **2019**, *20*, 1-7. https://doi.org/10.3390/ijms20092321.