

Biocompatible and Biodegradable 3D Printing from Bioplastics

Subjects: [Engineering](#), [Manufacturing](#)

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Three types of bioplastics are distinguished: Biodegradable bioplastics made from biobased materials. Examples include poly (lactic acid) (PLA), polyhydroxyalkanoates (PHAs), thermoplastic starch (TPS), and poly (butylene succinate) (PBS); Biodegradable bioplastics made from petrochemical resources. Examples include Poly (butylene adipate terephthalate) (PBAT) and polycaprolactone (PCL); Non-biodegradable or partially biodegradable bioplastics from biobased monomers and bioderived technical polymers. Examples are bioPE, bioPET, bioPP, and poly-trimethylene terephthalate (bioPTT).

biocomposites

biopolymers

additive manufacturing

fused filament fabrication

biodegradability

1. Introduction

Plastic materials are key in today's world, finding applications in a variety of industries, including consumer products, automotive, agriculture, aerospace, electronics and health care. While plastics continue to be popular due to their durability, adaptability, and cost-effectiveness, the continual depletion of petroleum resources to produce plastic materials and the environmental pollution caused by plastic disposal has aroused global concern. Efforts have been made globally to deal with the increasing amounts of plastic waste and to develop sustainable industrial materials [\[1\]\[2\]\[3\]\[4\]](#).

Bioplastics are a promising complement to conventional plastics since they are biocompatible and have the potential for biodegradability [\[5\]](#). Bioplastics are defined as plastics that are either derived from renewable natural resources (bioderived) and/or biodegradable [\[6\]\[7\]\[8\]\[9\]\[10\]](#). Such natural resources include starch, chitin, protein and cellulose [\[11\]](#). Biodegradable bioplastics provide disposal methods that reduce the quantity of plastic waste that ends up in the environment. Bioderived bioplastics, on the other hand, allow for a significant reduction in carbon footprint at the resource extraction stage. Biomass used for developing bioplastics is currently derived from starch constituents or cellulose.

The chemical structure of a bioplastic determines whether it is biodegradable; for example, a 100% bioderived bioplastic might not be biodegradable [\[12\]](#).

Despite the fact that bioplastics have been studied for almost a century, their widespread manufacturing is still in its early stages. The European Bioplastics Association reported a global bioplastic production of 2.11 MT in 2019, accounting for barely 0.6% of overall plastics production. This is expected to reach 2.89 MT by 2025 [\[10\]\[13\]](#). The major reasons for the slow adoption of bioplastics are their higher production costs and lower mechanical properties as compared to conventional plastics. Also, when plants are produced for bioplastics, there are additional environmental considerations, such as food competition and recycling problems [\[6\]\[12\]\[14\]](#). Nonetheless, because the world urgently requires a viable substitute for petroleum-based plastics, the bioplastics market is likely to develop significantly in the coming years, outpacing the petrochemical plastics market. Efforts have been made towards improving the mechanical properties of bioplastic materials to make them compete favorably with conventional plastics.

Many biocomposites now offer improved material properties; higher breathability, increased material strength [\[15\]](#), reduced thickness and improved optical properties, which all contribute to better material performance. These materials are naturally derived from proteins, lipids, aliphatic polyesters, or polysaccharides (obtained from agricultural products, livestock and fish farming) and are seen to be potential substitutes for conventional plastics due to their biocompatibility, safety and rate of biodegradation [\[16\]\[17\]](#). These polymers are quickly broken down by natural microorganisms in the presence of the proper temperature, moisture and oxygen without causing any environmental harm. Polysaccharides contain starch, cellulose, gum, alginate, chitosan, carrageenan, pectin, pullulan, or their derivatives [\[18\]](#). Starch constituents commonly used to produce bioplastic materials put pressure on food security. An alternative way to deal with this challenge is to make use of plant waste materials, such as agricultural by-products, in combination with recycled plastics to produce the bioplastics [\[19\]\[20\]\[21\]\[22\]](#).

Agricultural residues have recently emerged as a possible feedstock material in additive manufacturing processes [\[23\]\[24\]](#). Improved printing directionality [\[25\]\[26\]](#), enhanced mechanical properties [\[20\]\[27\]](#), and reduced warping [\[19\]\[22\]](#) have been reported with the use of agricultural residues. Additive manufacturing is a group of manufacturing techniques that has brought a paradigm shift in manufacturing in which objects are produced by materials joining layer upon layer. Recent developments in these techniques have made it possible to design and produce almost any product from a wide variety of materials with much flexibility, which was not possible with previous techniques. This group of techniques will facilitate the shift from the use of fossil-based materials to biocomposites as feedstocks, therefore, providing an opportunity to realize a more sustainable green economy [\[28\]\[29\]\[30\]](#).

2. Bioplastics with Biodegradability

Thermoplastics such as polyethylene terephthalate (PET), polyethylene (PE), polypropylene (PP) and polystyrene (PS) currently account for 60% of the total plastic demand [\[31\]](#). While these plastics are generally made from petrochemicals, there is an increasing demand for plastics derived from renewable resources as alternatives to petrochemically-derived plastics. These new alternative plastics are known as bioplastics, which are biobased, biodegradable or both [\[9\]\[32\]](#).

Three types of bioplastics are distinguished:

- Biodegradable bioplastics made from biobased materials. Examples include poly (lactic acid) (PLA), polyhydroxyalkanoates (PHAs), thermoplastic starch (TPS), and poly (butylene succinate) (PBS) [33];
- Biodegradable bioplastics made from petrochemical resources. Examples include Poly (butylene adipate terephthalate) (PBAT) and polycaprolactone (PCL) [34];
- Non-biodegradable or partially biodegradable bioplastics from biobased monomers and bioderived technical polymers. Examples are bioPE, bioPET, bioPP, and poly-trimethylene terephthalate (bioPTT) [35].

Biodegradable biobased plastics can be recycled or incinerated in the same way as conventional plastics; however, they are not extensively recycled since they are considered pollutants in the present recycling system [36]. They are primarily designed to degrade under precise conditions, most typically in a controlled setting in industrial composting plants [37]. Unless they are designed to biodegrade in a particular environment, such as soil or water, they will not degrade or will degrade very slowly in these conditions [11].

Table 1 shows some common bioplastics compositions, their properties and areas of application. The porosity, size, and carbon content of bioplastics alter as they degrade, affecting their ability to contain and retain water [38].

Table 1. Bioplastics composition, properties and applications.

No.	Bioplastic Composition	Properties	Application	Ref.
1	Poly(lactic acid) and poly (butylene adipate terephthalate)	<ul style="list-style-type: none"> - Dense matrix - Enhanced crystallinity - Reduced permeation to oxygen and moisture 	<ul style="list-style-type: none"> - Bakery packaging - Seafood packaging - Active packaging 	[39]
2	Chitosan, polyethylene glycol and methyl ether methacrylate	<ul style="list-style-type: none"> - Antioxidant - Antimicrobial nature - Enhanced mechanical properties 	<ul style="list-style-type: none"> - Active food packaging 	[40]
3	Thermoplastic starch	<ul style="list-style-type: none"> - Improved polymer compatibility - Reduced tensile strength - Antimicrobial nature 	<ul style="list-style-type: none"> - Fresh noodles 	[41]

No.	Bioplastic Composition	Properties	Application	Ref.
4	Cassava starch	<ul style="list-style-type: none"> - Optimized tensile strength and elastic modulus 	<ul style="list-style-type: none"> - Packaging 	[42]
5	Soya protein	<ul style="list-style-type: none"> - Super absorbent - water absorption capacity 	<ul style="list-style-type: none"> - Zinc release in soil 	[43]
6	Poly vinyl alcohol/poly (lactic acid)	<ul style="list-style-type: none"> - Higher encapsulation, efficiency and stability 	<ul style="list-style-type: none"> - Controlled release of fertilizers 	[44]
7	Gelatinized starch	<ul style="list-style-type: none"> - Higher swelling degree with delayed release of nutrients 	<ul style="list-style-type: none"> - Sustained release of micronutrients 	[45]
8	Seaweed extract and calcium alginate	<ul style="list-style-type: none"> - pH-responsive water holding capacity 	<ul style="list-style-type: none"> - Improved soil hydro physical and fertility 	[46]
9	Poly vinyl alcohol/poly (lactic acid)	<ul style="list-style-type: none"> - Compressibility - Mineralization behavior - Biocompatibility of the viscoelastic characteristic to natural cartilage 	<ul style="list-style-type: none"> - Tissue engineering 	[47]
10	Chitosan and biguanidine	<ul style="list-style-type: none"> - Sequential release behavior of the vascular endothelial 	<ul style="list-style-type: none"> - Injectable - Bone tissue engineering 	[48]
11	Starch/hydroxyapatite	<ul style="list-style-type: none"> - Low cost - Wide availability - biocompatibility 	<ul style="list-style-type: none"> - Biomedical device 	[49]

No.	Bioplastic Composition	Properties	Application	Ref.
12	Rice starch extract coated with iron oxide nanoparticles	<ul style="list-style-type: none"> - Biological active - Photo acoustic imaging 	<ul style="list-style-type: none"> - Chemo—photo thermal therapy 	[50], [51][52][53]

...s, intrinsic degradation responsible for degradation. It can be classified as chemical degradation, whereby agents such as water, oxygen, acids, alkalines and solvents are used for the breakdown of the biopolymer. Other types of biodegradation are physical degradation, atmospheric degradation and hydrolytic degradation, a unique form of chemical degradation [54].

The existing methodologies for assessing the biodegradability of bioplastics can be classified into three categories based on where the bioplastic is disposed of [34][54]: (i) soils, (ii) compost, and (iii) aquatic systems.

2.2. Biodegradation in Soil

The two most common standard methodologies for testing the biodegradation of plastics in soil are ASTM D5988-18, “standardized test to find aerobic plastic biodegradation in soil” and ISO 17556:2019, “determining the absolute aerobic biodegradability of plastic materials in soil by measuring carbon dioxide production or oxygen demand in a respirometer” [55][56]. ASTM D5988-18 estimates the quantity of carbon dioxide produced by microorganisms as a function of exposure duration, hence assessing the degree of biodegradability with comparison to a reference material. Similarly, the ISO 17556:2019 method controls oxygen intake or carbon dioxide production to produce the best rate of plastic biodegradation in test soil. In both cases, the bioplastic sample is essentially buried in closed containers containing soil that has been previously prepared. The containers are then exposed to a temperature range that promotes mesophilic microbe development, together with optimal moisture and oxygen conditions. **Table 2** shows the different methodologies to assess biodegradability in soil for some developed biopolymers.

The most fundamental and extensively used biodegradability index is mass loss. This methodology entails the measurement of samples collected at various periods and drying them until they reach a constant weight [57][58][59]. The gel permeation chromatography used to determine molecular weight is another technique for detecting mass loss [60].

Scanning electron microscopy (SEM) morphology analysis can be used to analyze biodegradability in soil. This technique is for determining the influence of water intrusion in biopolymers, which is known as surface erosion. Furthermore, using this technique, the identification and evaluation of microorganisms growing on the surface can be studied during biodegradation. This methodology was used by Jana et al. [61], whereby SEM was used to observe the breakdown of PHA and PBS samples, as well as the formation of biofilms on material surfaces. On the contrary, microorganisms did not cover two slowly degradable polymers, that is, PBAT/PLA and ICL- PN. Their findings matched the CO₂ measurements and, more importantly, the microbiological assessment of the biodegradation process.

Table 2. Methodologies to assess biodegradability in soil.

Biopolymer	Soil Conditions	Methodology	Ref.
PBAT/Nanocellulose	Lab-controlled conditions	ASTM D5988	[62]
Cellulose-based	Lab-controlled conditions	ASTM D5988	[63]
PHBV/Olive Pomace	Lab-controlled conditions	ASTM D5988	[64]
Chitosan/Corn cob	Lab-controlled conditions	ASTM D5988	[65]
Polyurethane (PU)/Starch	Lab-controlled conditions	ASTM D5988 visual analysis, morphological and chemical characterization	[66]
PLA/Glycerol	Lab-controlled conditions	ISO 17556	[67]
Starch/Nanocellulose	Lab-controlled conditions	ISO 17556	[68]
PBS, PHA, PBAT/PLA	Lab-controlled conditions	Microorganism characterization (ISO 17556)	[61]
PBAT, starch, and additives mixture	Lab-controlled conditions	Ecotoxicological analysis (ISO 17556)	[61]
Compounds of PHB and natural fillers	Lab-controlled conditions	Mass loss	[59]
Starch/Nanocellulose	Outdoor conditions	Mass Loss	[57]
PVA/starch	Lab-controlled conditions	Mass loss, visual analysis, biofilm area, and soil characterization	[58]
PLA, PBS, PHB and PCL	Lab-controlled and outdoor conditions	Mass loss, microbial characterization and mechanical properties	[69]
Starch-based	Lab-controlled conditions	Mass loss, mechanical properties and morphological analysis	[70]
PBS/Sugarcane Fibre	Lab-controlled conditions	Mass loss, morphological analysis, and thermal characterization	[71]
PHA	Outdoor conditions	Mass loss, chemical and morphological analysis	[72]
PLA and PLA/starch	Outdoor conditions	Mass loss, thermal characterization, morphological and chemical analysis	[60]

Biopolymer	Soil Conditions	Methodology	Ref.
Mixtures of PVA/starch with natural fillers	Lab-controlled conditions	Mass loss, soil characterization, morphological and chemical analysis	et al. [70], [73] t ratios of city of the

biocomposites were found to have decreased by more than 50% within the first week and subsequently gradually worsened until the experiment's conclusion. Sabapathy et al. [72] used chemical analysis to test for biodegradation of PHA. Using Fourier transform infrared spectroscopy (FTIR), the researchers looked at the shift and intensity of specific infrared peaks to calculate the degree of degradation.

2.3. Biodegradation in Compost

Composting is an aerobic form of solid waste treatment where biodegradable materials are organically broken down into humus. In the presence of microorganisms and under controlled conditions, humus is a beneficial nutrient source for increasing soil productivity and agricultural yield. This method is very helpful in resolving the disposal issue and reducing greenhouse gas emissions since compost is a healthy organic substrate that can be returned to the ecosystem [74][75].

Most methodologies of biodegradation of biopolymers in aerobic composting conditions have more standardizations than in soils due to the fact that these tests are typically carried out in laboratories. The common standards are shown in **Table 3**. The most often used standards are ISO 14855-1:2012 [76] and ASTM D5338 -15 [77]. Other standards used for testing biopolymer biodegradability in compost include the ASTM D6400-21 [78], ISO 17556 [56], ISO 20200 [79], and ISO 17088:2021 [80].

Table 3. Methodologies to assess biodegradation in compost.

Biopolymer	Origin of Compost	Methodology	Ref.
PLA, PLA/Nanocellulose (Gum)	Food waste	ASTM D5338, chemical/morphology analysis, mass loss, thermal/microorganism characterization	[74]
PVA, PVA/Nanocellulose (CNF)	MSW	ASTM D5338, mass loss, morphology, visual and chemical analysis	[75]
PHA-based	MSW	ASTM D5338 and ISO 20200	[81]
PVA/Starch	MSW	ISO 14855-1, ISO 20200, thermal/microorganism characterization	[82]
PLA, PLA/TAC and PLA/PHB/TAC	MSW	CO ₂ concentration, mass loss, chemical analysis, thermal characterization	[83]
Nano-reinforced PLA	MSW	ISO 16929, mass loss and microorganism characterization	[84]
PLA and PLA/Silica	Compost fermented with biomass	Modified ISO 17556	[85]

Kalita et al. [74] investigated the aerobic biodegradation of PLA biocomposite films, which were made by modifying PLA with the addition of fillers like chitosan and gum. Biodegradation was studied using a variety of analytical approaches, including differential scanning calorimetry (DSC), molecular weight analysis, microbial colony count and contact angle analysis. The findings indicated that modified PLA films are biodegradable, as well as the applicability of the methodologies for studying biopolymer biodegradation.

2.4. Biodegradation in Aquatic Systems

The catastrophic pollution situation that is currently afflicting aquatic systems is primarily due to plastic waste. This pollutant begins its flow in wastewater treatment effluents, which then make their way to fresh inland waters such as lakes and rivers, before continuing on to the oceans, where it settles and continues to break down into micro- and nano-plastics. The growth, behavior, development, reproduction, and life-span of marine and freshwater species may be impacted by the created debris [86][87]. **Table 4** highlights the most common biodegradability standards in aquatic systems that have been developed by ISO and ASTM.

Table 4. Methodologies to assess biodegradation in aquatic systems.

Standard	Approach
ISO 19679:2020	“Plastics—Determination of the aerobic biodegradation of non-floating plastic items at the seawater/sediment interface—Analyses amount of evolved CO ₂ ”
ISO 18830:2016	“Plastics—Determination of the aerobic biodegradation of non-floating plastic items at the seawater/sediment interface—Measures the demand for O ₂ in a closed respirometer”
ISO 14853:2016	“Plastics—Determination of the ultimate anaerobic biodegradation of plastic items in an aqueous system—Technique by measurement of biogas generation”
ASTM D7991-15	“Standard Test Procedure for Measuring Aerobic Biodegradation of Plastics Buried in Sandy Marine Sediment in a Controlled Laboratory”
ASTM D7473-12	“Standard Test Procedure for Weight Attrition of Plastic Materials in the Marine Environment by Open System Aquarium Incubations”
ASTM D6691-17	“Standard Test Procedure for Determining Aerobic Biodegradation of Plastic Materials in the Marine Environment by a Natural Sea Water Inoculum or Defined Microbial Consortium”

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