REVIEW PAPER



A review on poly lactic acid (PLA) as a biodegradable polymer

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Received: 9 November 2021 / Revised: 13 February 2022 / Accepted: 15 February 2022 / Published online: 6 March 2022

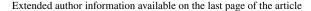
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Abstract

Biodegradable plastics are among the most promising materials to replace conventional petroleum-based plastics that have caused many adverse impacts on the environment, such as pollution (land, water, etc.) and global warming. Among a range of biodegradable plastics, poly lactic acid (PLA) is not only widely available but also safe to be decomposed after its usage without polluting the environment. PLA is also in parity with other conventional plastics such as PP, PET in terms of various properties suitable for industrial usage such as mechanical, physical, biocompatibility and processability. Thus, PLA has become the most used biopolymers in many industries such as agriculture, automotive and packaging by having these characteristics. Its higher demand has contributed to a stable increment in the global PLA market. In fact, over the years, the market for PLA has grown up and will keep on expanding in the future. Overall, the PLA-based bioplastic would be an excellent substitute for the existing conventional plastics in various applications, hence will serve to protect the environment not only from pollution but also work as a sustainable and economical product. This paper will review all the recent related works and literature on PLA as the biodegradable material regarding its properties, usability, productivity and substitute.

Keywords Poly lactic acid · Biodegradable · Polymer · Plastics · Composites · Market

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Introduction

Over the past decades, the growth of synthetic petroleum-based plastics has brought countless favors to the people and assisted in economic development. Back in the 1950s, the yearly plastic production was only 1.5 million tons, but in 2018, it increased to approximately 400 million tons. By 2050, it is expected that the annual output of global plastic will amount to 1800 million tons. However, despite their excellent features such as low cost, outstanding durability, processability and lightweight, most synthetic plastics have brief serviceable life before disposal, especially those for packaging and applied as a single-use item. So, low service life and dumping waste have brought serious environmental issues such as global warming and plastic pollution. Out of the 90 million tons of humanmade plastic, around 70 million tons are accumulated in the environment and will gradually break down into microplastics, creating significant health concerns. Global warming has also intensified due to the uncontrolled emission of greenhouse gases (GHGs) such as carbon dioxide (CO₂). The CO₂ concentration and global temperature in 2019 are over 400 ppm and had risen by 1 °C, respectively, which is a severe environmental issue. It is predicted that in 20 to 40 years, based on the current rate, the CO₂ concentration will go over 450 ppm. Global temperature will increase by 1.5 °C [1].

Strategies and plans have been made locally and globally in some respect to overcome these problems. For instance, the European Union (EU) vision was introduced in January 2018 to accomplish a more sustainable plastics industry by 2030. This strategy promotes plastic recycling and confers (bio) degradable plastics regarding their opportunities and risks. Thus, the demand for alternative materials has been increasing, especially biodegradable plastic/polymers. As a material, it is a viable option to improve and potentially solve the global issue of plastic waste hazards. It is because bio-based raw materials can absorb CO2 from the atmosphere and degrade much faster than traditional plastics, making it a suitable tool for reducing carbon emissions and plastic pollution [1, 2]. In addition, conventional plastics processing uses 65% more energy than processing bioplastic, releases 30–80% higher GHGs than bioplastic, and is unsustainable (due to environmental issues). Therefore, there is an urgent need to replace petroleum-based plastic with bio-based polymers [3].

Among the rigid bioplastics, one of the most commercially used and successful bioplastics is poly lactic acid (PLA), as it has good processability and mechanical properties. Compared to the other biodegradable products, PLA has exceptional durability, mechanical strength and transparency, making it one of the top choices of biodegradable materials to be used [1]. Apart from that, since lactides are formed by the agricultural byproducts' microbial fermentation (mainly the carbohydraterich substances), PLA is considered a suitable alternative to petrochemical-derived products [3]. With increasing awareness regarding environmental preservations, biodegradable materials, especially bio-based plastics, would replace and reduce the usage of conventional petroleum-based plastic soon. Therefore, this manuscript will give a proper understanding of PLA as a biodegradable polymer, the types and functionality of its applications in many part of works.



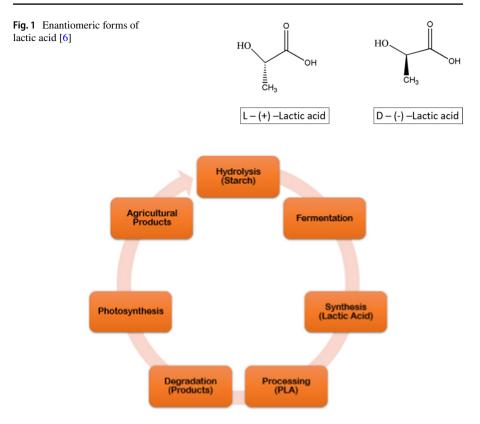


Fig. 2 PLA cycle in nature [4]

Poly lactic acid (PLA)

PLA is derived from lactic acid (LA), a linear aliphatic thermoplastic polyester [4]. It is recognized and classified as one of the aliphatic polyesters, similar to polyglycolic acid (PGA), and is commonly made from hydroxyl acids [5]. Due to the asymmetric carbon atom in its molecule, LA exists in two forms, L-LA and D-LA, as illustrated in Fig. 1. These two forms are mirror images of each other. They have identical physical and chemical properties in their pure forms. The only differences are that plane-pThe plane-polarized light direction of rotation that is generated by a chemical is indicated by the sign of (+) and (-) [1]. Polarized light rotates similarly in opposite directions, and that other asymmetric (chiral) reagents such as most enzymes in biological systems, have different reactions. The plane-polarized light path or like PETE polymer, but in term of temperature control condition, it has a significantly lower maximum continuous use of temperature [6]. Additionally, via chemical conversion, PLA can be repolymerized when it is recycled back to lactic acid [3]. Figure 2 below shows the PLA cycle in the nature.



The PLA properties

Depending on the ratio and distribution of L- and D-LA in the polymer chains, the thermal and mechanical properties of PLA would be justified. Due to -CH₃ side groups in the polymers, PLA is considered a hydrophobic element in nature. Thus, the steric shielding effect of the methyl side groups made it more resistant to hydrolysis compared to PGA [7]. L-lactide would produce semicrystalline polymer (PLLA), while poly (DL-lactide) (PDLLA) is an amorphous polymer [8].

Since the PLA is mainly made of L-LA blocks, the polymer is semicrystalline, and its structural regularity is high. At high temperatures, up to 37% of the crystalline regions would provide more mechanical strength. Glass transition temperature (Tg) is a significant characteristic of polymer materials, where it specifies the temperature above which amorphous regions of the PLA plastics would be changed rubbery or softened from their brittle nature. Tg for both semicrystalline and amorphous PLAs are between 50 and 70 °C [9]. The degradation half-life is dependent on the PLAs' stereochemistry, and the molecular weight typically ranges from six months to two years [4]. Additionally, as the degradation time differs, the mechanical properties would also be varied [8].

The poly (L-lactic acid) (PLLA) is a transparent, rigid polymer. It has elongation 85% to 105% at break. The tensile strength of PLLA is between 45 and 70 MPa. As for thermal properties, the melting point of PLLA is at 170–180 °C with a T_g of 53 °C [8]. However, due to the physical blending of the existing polymer with poly-D-lactide (PDLA), PLLA's melting and heat deflection temperature can be elevated by 40–50 °C and 60–190 °C, respectively [5]. Furthermore, PDLLA has no melting point but has T_g about 55 °C. These characteristics ensure PDLLA has lower tensile strength [8]. The tensile properties of neat PLA films with different D contents are listed in Table 1.

One important property for a semicrystalline thermoplastic is the recrystallization rate during cooling from the melting stage. Pure PLLA would spontaneously crystallize, but the crystallization rate would decrease with increased D-LA content in the solutions. Thus, less refined PLLA would generally contain a quasi-amorphous phase which showed inferior mechanical properties [9]. Production of commercial PLA typically uses polymer-grade L-LA, which includes over 98–99% L-LA and less than 1–2% of D-LA [10].

So both physical and mechanical properties of PLA, such as tensile strength, hardness, stiffness, modulus and melting points, are influenced by the polymer crystallinity. The polymer is semicrystalline when PLLA is over 90%, while an amorphous polymer has lower amounts of PLLA (and therefore decreased optical purity). Furthermore, the solubilities of PLA in different types of solvents are: (1) *Completely soluble*: Dioxane, chloroform, methylene chloride, 1,1,2-trichloroethane, acetonitrile, dichloroacetic acid; (2) *Partially soluble*: Ethyl benzene, acetone, toluene, tetrahydrofuran (only when heated to boiling temperature); and (3) *Not soluble*: Water, alcohols, linear hydrocarbons [6]

Table 2 lists the mechanical properties of PLA-based polymers. The property values are expressed in ranges [6] since testing materials and procedures are



Table 1 Tensile properties of neat PLA films by several studies [4]

PLA	Processing	Tensile strength (MPa)	Elongation (%)
120 kDa	Extrusion Compression molding	55.1	3.8
1.1-1.7% D-content	Melt blending Compression molding	16.4	2.1
99 kDa	Extrusion	34.1	2.5
2002D® 4% D-content 235 kDa	Extrusion Injection molding	60.0	2.0
4042D® 6% D-content 130 kDa	Melt blending Compression molding	56.3	3.6
2002D® 4% D-content 235 kDa	Extrusion Blown molding	34.6	2.1
12% D-content 68 kDa	Compression molding	14.0	8.0
4042D® 6% D-content 130 kDa	Melt blending Compression molding	70.2	7.4
2000D®	Casting (Chloroform)	24.8	7.9
125 kDa	Extrusion Compression molding	55.4	2.6
120 kDa	Compression molding	62.7	6.1
12% D-content 160-220 kDa	Extrusion Compression molding	36.0	2.1
2002D® 4% D-content 235 kDa	Melt blending Compression molding	55.0	4.5
4032D®	Extrusion Injection molding	65.0	5.0
4042D® 6% D-content 130 kDa	Extrusion Injection molding	38.0	1.0

Table 2 Mechanical properties of PLA-based polymers [6]

1 1			
Property	PLA	PLLA	PDLLA
Density, ρ (g/cm ³)	1.21–1.25	1.24–1.30	1.25–1.27
Tensile strength, σ (MPa)	21–60	15.5-150	27.6-50
Elastic modulus, E (GPa)	0.35-0.5	2.7-4.14	1-3.45
Ultimate strain, ε (%)	2.5-6	3.0-10.0	2.0-10.0
Glass transition temperature, T _g (°C)	45-60	55-65	50-60
Melting temperature, T_m (°C)	150–162	170–200	Amorphous-no melting point

Table 3 Mechanical properties of PLA and petroleum-based polymers [11]

Property	PLA	PP	PET	Nylon
Density (g/cc)	1.25	0.9	1.4	1.2
Haze (%)	2.1	1-4	2–5	2-3
Tensile strength (MPa)	109.97	189.95	204.95	249.94
Tensile modulus (MPa)	3299.26	2399.46	3799.15	1824.59
Ultimate elongation (%)	160	110	140	125
Tear resistance (g/mm)	0.3810	0.1316	0.4572	0.3302

PET polyethylene terephthalate, PP polypropylene



being varied. Meanwhile, Table 3 shows the properties of PLA in comparison to other conventional petroleum-based plastics such as PET, PP and nylon.

PLA synthesis methods

There are three basic processes used to synthesize the PLA, i.e., formation of lactic acid through microbial fermentation, purification of lactic acid and preparation of its cyclic dimer, and ring-opening polymerization (ROP) of lactides or lactides polycondensation of the PLA monomer, namely LA [12]. However, the ring-opening polymerization process is the most widely used method of obtaining high molecular weight PLA [13]. Since the properties of PLA vary with isomer composition, temperature, and reaction time, the need to control the polymerization parameters is essential [14]. Figure 3 shows the chains of polymers being produced by two types of lactic acid, namely L- and D- lactic acid. Three types of stereo forms of poly have been produced, i.e., poly (l-lactide), poly (d-lactide), and poly (dl-lactide) [15]. Poly (dl-lactide) is also known as meso-dilactide, produced when L- and D- lactic acid are combined.

Initially, LA would be produced and is synthesized through fermentation or chemical synthesis. A different number of isomers would be produced depending on the methods used. For instance, when chemical synthesis is used, the racemic mixture (consists of both L (+)-lactic acid and D (-)-lactic acid) is produced with an equal amount of concentration. Meanwhile, a more significant number of either type of the LA isomer would be produced when the fermentation process is used. Generally, the microbial fermentation of renewable sources such as sago and cassava starch will yield optically pure L (+)- or D (-)- lactic acid with an appropriate selection of microorganisms [17]. After the formation of LA, it would undergo purification. Several techniques can be used for the purification, i.e., nanofiltration and electrodialysis, ion exchange resin, hybrid short path evaporation and reactive distillation [18]. The purified LA is then used to produce PLA.

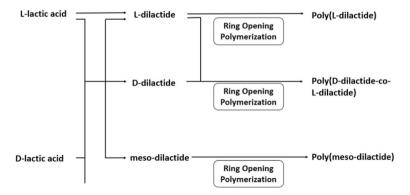


Fig. 3 Types of lactic acid and chains of polymers produced [16]



Ring-opening polymerization (ROP)

The first synthesis method is ring-opening polymerization (ROP). This method is catalyzed by organometal catalysts, the more successful conversion of lactide (the cyclic dimer of lactic acid) to PLA. Most PLA production processes use this method. In this method, the terminal end of the polymer chain acts as the reactive center. Thus longer polymer chain can be formed when the further cyclic monomers open their ring system. But initially, lactic acid is dehydrated at high temperatures and under vacuum conditions and poly-condensed into its oligomers. Then, via internal transesterification, it would be catalytically depolymerized into lactide. PLA with high molecular weight is obtained when the ring of lactide opens. Distillation or crystallization can be used to remove residual moisture, lactic acid and the meso-lactide from the optically pure D or L form lactide. Figure 4 shows the cyclic dimers for the ROP process.

Additionally, there is little moisture to be removed from the molten PLA resin [1, 18]. Short residence times, mild process conditions, absence of byproducts and high molecular weight are reasons to choose ROP as the preferred process for most industrial-scale production [6]. The schematic diagram of a typical ROP process is illustrated in Fig. 5.

Direct polycondensation polymerization

Through this method, the LA can be polymerized at reduced pressure and with a catalyst. Despite being an inexpensive route, this method is unfavorable as the obtained polymer had a low molecular weight (1000–5000 Da). A lower molecular weight may happen due to the highly viscous reaction of the mixture (slow moisture diffusion in the highly viscous polymeric melt); hence it is difficult to remove all the water produced from the lactic acid condensation. The PLA obtained properties such as molecular weight and characteristics may be limited by the residual water trapped in the PLA melt. But, incorporating coupling agents or adjuvant such as bis (tri-chloromethyl) in the process would help obtain a high molecular weight polymer [1, 18].

Fig. 4 Cyclic dimers for ROP process [6]



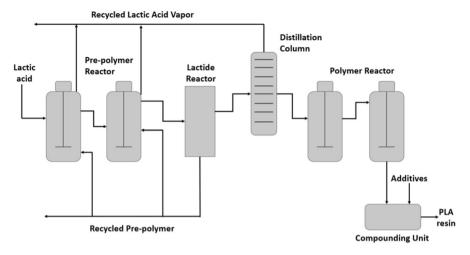


Fig. 5 Schematic diagram of typical ROP process [1]

Azeotropic condensation polymerization

Lastly, azeotropic condensation polymerization is also used. Through this method, the LA can directly contact polycondensate into a polymer with high molecular weight by using an organic solvent such as toluene, xylene or diphenyl ether. Distillations are then used for the azeotropic removal of water. However, this process is not useful as organic solvents generate impurities. The process becomes costly to remove moisture and post-processing [17]. Figure 6 illustrates the synthesis methods for PLA.

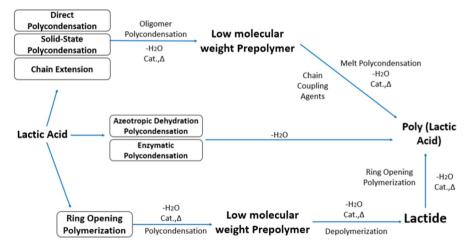


Fig. 6 Synthesis method for PLA [19]



Advantages and limitations of PLA

The main benefit of using PLA is its eco-friendliness. As the material can be derived from renewable resources such as corn, wheat or rice, it can be used without concern of its depletion. Furthermore, it is also biodegradable, recyclable and compostable, which indicates almost no environmental impact occurred during its disposal. The next advantage is PLA's biocompatibility properties. This property is especially advantageous for biomedical applications. There would be no toxic and carcinogenic effects of the material on the human body. When PLA degrades, it breaks down into H₂O and CO₂, not interfering with tissue healing [20, 21].

Besides this, in respect of processability, compared to other biopolymers such as PHA, PEG and PCL, PLA has better thermal processability. This can be achieved as varieties of processes can be applied or used to form the PLA into bioplastic. These processes include drying, film extrusion, cast film and sheet, blow molding and thermoforming. Lastly, the production of PLA-based bioplastics requires 25 to 55% less energy than conventional petroleum-based plastics and will be reduced more and could be less than 10% [21].

However, despite being considered one of the best materials to be used in the industries, PLA has some drawbacks. Firstly, it breaks with less than 10% elongation, showing it is very brittle and provides limitations to use in various applications [20, 21]. Hence, PLA has limited applications for products that need plastic deformation at higher stress levels, such as screws and fracture fixation plates in the biomedical field. Secondly, PLA also has a low degradation rate. The degradation rate of PLA is affected by its crystallinity, molecular weight and distribution, morphology properties, water diffusion rate into the polymers and stereoisomeric applications [21]. PLA would degrade through the backbone ester groups' hydrolysis, and the degradation rate is prolonged, 3 to 5 years of vivo lifetime. This low degradation is unusual in the biomedical field.

It can cause an inflammatory response from tissues to living hosts due to its strong hydrophobicity with a static water contact angle of approximately 80° and direct contact with biological fluids. It has been used as a soft cell-affinity tissue engineering material [20, 21]. Thus, polymers blending with other polymers, functionalization and nanofillers additions have been used [14]. Table 4 shows PLA has high gas permeability compared to other packaging plastics. It means that it has poor barrier properties and is hence unsuitable for some beverage bottle applications. Meanwhile, in Table 5, the main pros and cons of PLA are summarized.

PLA environmental footprint

Life cycle assessment (LCA) can evaluate the environmental footprint (EFP) of PLA. LCA is an important tool in identifying potential environmental load as it quantifies the load during the product life cycle using the "cradle-to-grave" concept [22]. The "Cradle-to-grave" concept is built upon the system's potential impact involving the production of the bio-based polymers, initiating from



Table 4	PLA, PGA and other
types of	plastics' gas and
moisture	e vapor permeability [1]

	Oxygen/ cc mil/ (100 in ² day atm)	Moisture vapor/g mil/ (100 in ² day)	CO ₂ / cc mil/ (100 in ² day atm)
PLA	38–42	18–22	183–200
PGA	0.036	0.5	0.19
PET	3–6	1-2.8	15-25
HDPE	130-185	0.3-0.4	400-700
PP	150-800	05-0.7	150-650
PA6	2–3	16–23	10-12
EVOH	2-2.6	1.4-6.5	N/A
HIPS	300-400	10	N/A
PVC	4–30	1–5	4-50

Table 5 PLA main pros and cons [6]

Advantages	Disadvantages
Eco-friendliness The production of PLA is from renewable sources (corn, wheat, rice). Furthermore, it is biodegradable, recyclable, and compostable. Its production absorbs CO ₂	Poor toughness PLA is a very brittle material, with a break elongation of < 10%. For those applications that require plastic deformation at high-stress levels, this may represent a limit
Biocompatibility The key product of PLA degradation, lactic acid, is non-toxic and the organism itself metabolizes it	Slow degradation rate By hydrolysis, PLA naturally degrades; its rate depends on several variables, such as crystallinity and molecular weight. Slow degradation of PLA results in a high lifetime of devices in vivo and poses problems with the disposal of commodities
Processability The thermal processability of PLA is greater than other biopolymers. Injection molding, film extrusion, blow molding, thermoforming, fiber spinning, and film-forming are among the methods of PLA processes	Hydrophobicity PLA static water contact angle value is about 80°, making PLA a relatively hydrophobic material, which results in low cell affinity and can lead to inflammatory response upon direct contact with biological fluids
Energy-saving PLA requires 25 – 55% less energy than petroleum-based polymers	Lack of reactive side-chain groups PLA is chemically inert, making surface functionalization and bulk modification challenging tasks

its raw materials to the production stage, including disposal and recovery processes, as shown in Fig. 7. Thus, the most appropriate and comprehensive disposal systems could be established by using LCA [23]. The framework of international standards such as ISO14040 and ISO 14044 is the guidelines used by the LCA studies [24]. The guidelines for performing detailed LCAs have been established by many organizations, including the Society for Environmental Toxicology and Chemistry (SETAC), Environmental Protection Agency (EPA) and the American National Standards Institute (ANSI) [25]. The crucial benchmarks such as greenhouse gases (GHG) emissions and non-renewable energy use are measured and compared with traditional polymers' data to obtain the PLA environmental performance. Table 6 shows the environmental footprint of



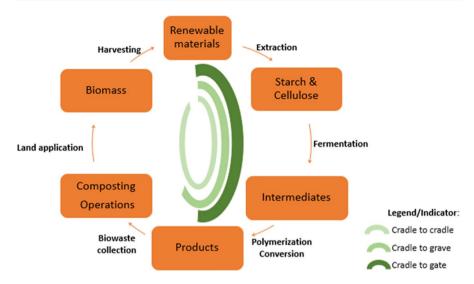


Fig. 7 "Cradle-to-grave" concept of bio-based polymers [24]

Table 6 Environmental footprint of 1 kg of PLA with consideration of the world as the geographical region [24]

Impact category	PLA
Climate change, kg CO ₂ eq	2.7907
Ozone depletion, kg CFC ⁻¹¹ eq	2.18E-0.7
Terrestrial acidification, kg SO ₂ eq	0.0218
Freshwater eutrophication, kg P eq	0.0004
Marine eutrophication, kg N eq	0.0065
Human toxicity, kg 1,4-DB eq	9.4123
Photochemical oxidant formation, kg NMVOC	0.0115
Particulate matter formation, kg PM ₁₀ eq	0.0063
Terrestrial ecotoxicity, kg 1,4-DB eq	0.0089
Freshwater ecotoxicity, kg 1,4-DB eq	0.0090
Marine ecotoxicity, kg 1,4-DB eq	3.5355
Ionizing radiation, kBq U235 eq	0.1398
Agricultural land occupation, m ² a	1.1321
Urban land occupation, m ² a	0.0674
Natural land transformation, m ²	0.0004
Water depletion, m ³	0.2726
Metal depletion, kg Fe eq	0.1538
Fossil depletion, kg oil eq	0.8246
Non-renewable energy, MJ primary	41.739

1 kg of PLA considering the world as the geographical region. In contrast, in Table 7, the important cradle-to-grave impact assessment categories for 1 ton of PLA. Additionally, Fig. 8 illustrates the life cycle of PLA.



Table 7 The relevant cradleto-grave impact assessment categories for 1 ton of PLA [26]

Impact category	Unit	Total
Global warming potential (GWP)	kg CO ₂ eq	501 (2334 ^a)
Water resource depletion (WS)	M ³ water eq	36.1
Marine eutrophication (EP)	kg N eq	13.3
Terrestrial eutrophication (EP)	molc N eq	33.8
Acidification (AP)	molc H+eq	18.2
Particulate matter (PM)	kg PM 2.5 eq	1.74
Land use (LU)	kg C deficit	17,441
Renewable energy use (REU)	GJ	60.4
Non-renewable energy use (NREU)	GJ	28.8

GWP includes direct land-use change, excludes biogenic emissions, and includes the CO₂ uptake by the bio-based material

^aTotal fossil GWP excluding the CO₂ uptake

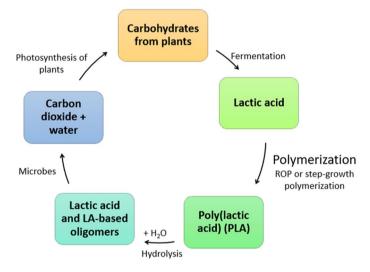


Fig. 8 Life cycle of PLA [27]

Biodegradable polymer/biopolymer

As defined by the International Union of Pure and Applied Chemistry (IUPAC), Biodegradable polymers are "polymers, susceptible to degradation by biological activity with the degradation accompanied by a lowering of its mass." Meanwhile, according to other definitions as standard CEN/TR 15,351: 2006, biodegradable material should be mineralized into the water, carbon dioxide and biomass during biodegradation. In a different environment, the humidity, temperature or concentrations of microorganisms vary, resulting in different biodegradation rates [2]. Polymer material with excellent performance is slowly biodegradable and exists in nature as part of the carbon cycle is considered the ideal biodegradable plastic [28].



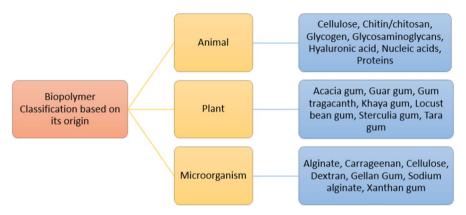


Fig. 9 Classification of biopolymer-based on its origin [30]

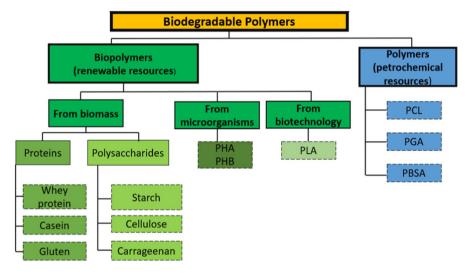


Fig. 10 Biodegradable polymers' classification based on two resources [29]

Sources of biopolymer

There are various natural resources from which biopolymers can be originated, such as cellulose, chitosan, starch and proteins from plants and animals, as shown in Fig. 9. Due to their green characteristics, these natural polymers are the desirable choices for petroleum-based and non-biodegradable or synthetic plastic packaging materials. Additionally, biodegradable polymers can also be classified depending on their synthesis processes and sources, as illustrated in Fig. 10. The classification is as follows: (1) Directly obtained from biomass: Polysaccharides, proteins, (2) Synthetic biopolymers from biomass: PLA; (3) Synthetic biopolymers from petrochemicals: poly (glycolic acid) (PGA), poly (butylene succinate-co-adipate)



(PBSA), polycaprolactone (PCL), and (4) Obtained from microbial fermentation: poly (hydroxybutyrate) (PHB), poly (hydroxy alkanoates) (PHA) [29]

Biopolymer production routes

Depending on the desired products and the materials/ precursors available, the production process for biopolymers can be classified into four different groups, namely renewable sources, bacterial synthesis method, chemical synthesis method and biopolymer blends. This classification is shown in Fig. 11, but the three principal routes in producing bioplastic are illustrated in Fig. 12. The bio-based polymers made from renewable sources can be used in various sources. For instance, plant-based precursors, cellulose esters, lignocellulose fibers, and PHA produced bio-based polymers. The physical properties of the final products are heavily influenced by the method of extraction. Generally, the preferred organic materials/ precursors contain a large amount of cellulose and other fibers as they help enhance the materials' mechanical strength [31]. In 2-step biomass conversion, the bio-based precursors would be produced in the first step through biochemical and/or chemical transformation. There would be two types of monomers, i.e., "novel" and "drop-in" monomers. The "drop-in" monomers are the bio-based replacement of the conventional monomers. The monomers produced would then be polymerized to complete the conversion [9].

Additionally, in the synthetic chemical method, the extraction of lignin and cellulose materials undergoes the treatment of agro-waste/food wastes with acids and alkalis. Hence, C=O and C-O-C, among the functional groups, are formed via the treatment processes. And their chemical bonds will influence the mechanical properties of the biopolymer. However, carboxylic acids, furan derivatives and ligninderived phenols, produced from the chemical process, inhibit the enzymatic activity essential in the fermentation phase. A specific species of white-rot fungi such as

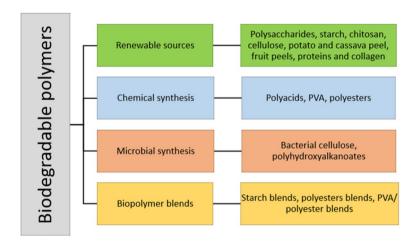
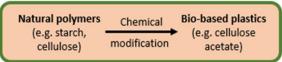


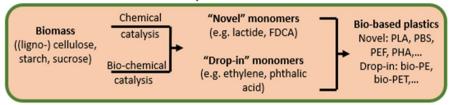
Fig. 11 Classification of production processes for biodegradable polymers [31]



I. Modification of natural polymers



II. 2-Step biomass conversion



III. Direct production in plants

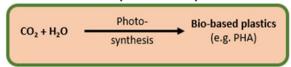


Fig. 12 Principal routes in producing bioplastic [9]

Ceriporiopsis subvermispora are added to reverse the harmful effect of acids and alkalis on the fermentation process [31]. So by the chemical modification of natural polymers such as cellulose and starch, bio-based plastic is obtained [9].

Additionally, through the interaction of microorganisms such as Gram-negative and Gram-positive bacteria in the presence of carbon-rich materials such as agrowastes, biodegradable polymers can also be produced. Limited availability of essential nutrients (e.g., phosphorus and nitrogen), pH changes, the type and composition of culture, and media are among the parameters that triggered the bacterial production of the polymers. In this route, a polymeric material would be produced, and without further modification, it can be used as plastic. PHA is one of the bio-based plastics that is produced via this route. This microbial synthesis method generates PHA, biomass, and exo-polysaccharides (EPS) at 3.63, 1.75 and 1.2 g/L, respectively. In addition, poly-b-hydroxybutyric acid (PHB) can also be produced through this synthesis method. Availability of a carbon-rich precursor that bacteria use as the food and energy source will influence the bacterial synthesis of the biopolymer. PHB is suitable for high strength applications compared to other microbial biopolymers as its mechanical properties are quite similar to petroleum-based biopolymers such as PP [9, 31].

Lastly, Minerv-PHATM, Bio-Onhave and Mater-Bi/Novamont are among the companies, incorporated the biopolymer blends method in their production of bio-based polymers. But bio-based polymers produced from biopolymer blends and



starch rely on techniques that had inadequate commercial utility. So, these companies' production capacity is deficient (97–560 kilotons). Despite their optimal performance, bioethanol-based biopolymer blends have also been reported to be not 100% biodegradable [31]. Hence, it can be concluded that this method is unfeasible in its current form.

Bioplastic carbon cycle and LCA

The carbon cycle occurs through its exchange into the earth. For a plant to grow, it absorbs carbon dioxide (CO_2) but releases it back into the earth when the plant biodegrades, thus creating a perfect closed-loop cycle. Figure 13 shows the carbon cycle of bioplastic, which is an environmentally friendly "cradle-to-grave" process. Although bioplastic has the same cycle time as traditional plastic, it is understood that about 35% of energy would be saved in the manufacturing process with bioplastic as it needs a lower processing temperature compared to the conventional one. The GHG emissions at the bioplastic production phase are also two-thirds lower than traditional plastic emissions during the production process [5]. Additionally, as agricultural products such as starch, cellulose and wood are being used as raw materials in the production of bioplastics, the environmental impact of plastics has been reduced, especially in terms of CO_2 release. Thus, the net balance of the CO_2 would be substantially reduced as the CO_2 produced during the production till the disposal process would be balanced by CO_2 intake during the plant growth cycle [32].

A material's sustainability and performance with the environment can be measured by using the LCA. For biopolymer, the LCA is similar to the LCA of biomass, as after degradation using enzymes and microbes, the biopolymers are converted into carbon and water. The biodegradation rate is calculated by the $\rm CO_2$ emitted during the analysis, divided by the theoretical $\rm CO_2$ present in the sample. Quantitative

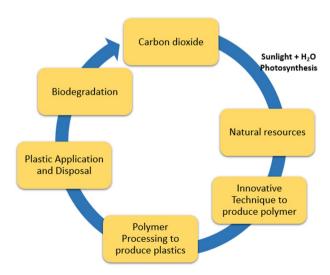


Fig. 13 Bioplastic carbon cycle [5]



evaluation of the potential biodegradability of biodegradable polymers in the natural environment can be done using $^{\rm Ed}K$. Starch and polyethylene are used as reference materials to define the $^{\rm Ed}K$ values of 100 and 0 correspondingly. ISO 14852 method is being used to determine those values by detecting the evolved ${\rm CO}_2$ as an analytical parameter [3].

Biopolymer waste management

The wastes that come with the application are unavoidable to separate initially, as biopolymeric products are commonly used in many industries or fields. Hence, the waste treatment of such products is important. The following several strategies could be used to deal with this waste issue.

Recycling

There are several ways by which the waste polymers can be recycled. The different stages in this process are collection, separation, processing/production and marketing [33]. The treatment method can be classified into three groups, i.e., mechanical, thermal and chemical generally. The recycling of polymers is at the beginning of the stages of the treatment. Still, only 14% of plastic packaging is collected globally for recycling, and the amount of exchanged waste plastics is less than 5% of the yearly worldwide production of new plastic [34].

In mechanical recycling, the waste plastic solids are separated, melted, extruded and re-granulation before manufacturing new products [33]. Figure 14 shows the steps involved in mechanical recycling. The monotype polymers are usually more suitable for this form of recycling than multilayer plastics, but materials such as temperature-sensitive polymers, composites and thermosets cannot be treated mechanically [34]. Since this process is simple in terms of the technologies used, it is the most prevalent method used for waste recycling. This approach is equally suitable

Fig. 14 Steps involved in mechanical recycling [38]





and feasible for manufacturing both glass fiber and carbon fiber reinforced polymer composites. In addition, it also has less environmental impact than the other form of recycling in terms of energy consumption [35]. However, it is a simple and easy technique but creates problems in degrading and heterogeneity solid plastic waste. This happens due to the chemical reaction triggered by the addition of polymerization and polycondensation. Theoretically, these are reversible processes, and photooxidation might happen with energy or heat supply. In addition, the development of oxidized compounds and harsh climatic conditions might cause linear and branching of the plastic polymer chain [36]. The value of the materials is also significantly decreased with grinding [35].

The next one is thermal recycling. In this technique, the wastes will go through a pyrolysis process, heated at a temperature of 300–700 °C without an oxygen environment. After the pyrolysis process, the synthetic gas or oil and char will remain used for industrial application. For example, the oil or synthetic gas obtained can be used for energy recovery, and char can be used as fertilizer in agriculture. But the recovered fibers could be reused in a limited capacity as they could be significantly damaged depending on the heating temperature. Synthetic gases, oils and solids can be obtained in different ratios depending on the heating temperature of the process [35]. This method is ideal for processing complex polymer waste [37]. Pyrolysis is useful for processing hydrocarbons and reusing plastic waste. It also reduces land-filling and CO_2 emissions, making the products faster and utilizing them to generate electricity and heat [36].

Lastly, chemical recycling could be defined as converting plastic materials or waste into smaller molecules of liquids or gases that could be used to make new petrochemical products and plastics [36]. Monomers or oligomers can be obtained from the polymers through a chemical reaction, and reproduction may occur [38]. This recycling method is very beneficial for sustainable industries as low waste production and high product yield. The recent techniques for this recycling to generate monomer units are smelting by the furnace, direct chemical treatment, and degradation of polyethylene terephthalate and nylon. Additionally, it is possible to process both heterogeneous and polluted polymers by using this chemical recycling [36].

Mechanical recycling uses the simplest technological process among these recycling methods and produces the least valuable final product. Whereas using chemical recycling, the best quality products could be obtained. However, as this chemical recycling process is still developing, it cannot be used commercially in full force [35].

Degradation and composting

Degradation will affect many properties of the materials related to its functional characteristics directly or indirectly, as it is a complex nonlinear time-dependent process. The mechanical properties of a material are heavily affected by the last stage of the degradation process. So the degree of degradation must be checked and monitored to ensure the properties of materials, and degradation makes the material more brittle. Several types of degradation processes are photoinduced, thermal, ultrasonic, mechanical, hydrolytic, biological and chemical degradation. However, degradation



does not occur to all polymeric materials as petrochemicals-based plastics cannot be broken down by natural processes and needs a unique method to degrade [39].

Composting is another waste disposal method. The main goal for composting is to degrade the materials into suitable products. There are several types of industrial-scale composting, and they are aerated static pile composting (aeration-controlled perforated piping), in-vessel composting (bioreactors, i.e., air flow and temperature-controlled tanks), and windrow composting (waste are piled in long rows). Though the compost controlled technique is widely used, it is inefficient as no improvement could be made naturally [34].

Incineration with energy recovery

The waste materials' energy produced from the incineration (burning process) would be used in electricity and heat generation [24]. This incarnation process is an efficient technique, especially for processing plastic waste such as electronic and electrical waste (highly mixed plastic wastes) [36]. Consequently, this method is useful for volume-reduction of the wastes and is effective as the alternative method for waste-to-energy conversion. Using this method, the dependence on fossil fuels and other energy sources would be significantly reduced [24].

Airborne particulates and greenhouse gas (GHG) emissions are the products of this process. In municipal solid waste (MSW) incinerators, CO₂ emissions have been increased. Thus, to avoid the toxic materials being released from the incineration activities, a suitable filter trapping system should be installed [39].

Landfills/landfilling

5 to 25wt% of total waste in landfills are plastics, and half of solid plastics produced per year are wasted on a global scale [34]. Globally, landfills' space is becoming a hurdle issue for most countries, despite being the traditional approach in plastic waste management [36]. This waste management option is unpreferable because of its environmental impacts as health hazards, vegetation damage, ground water pollution, etc., due to the formation of gas and leachate in the landfills. Degradation is almost impossible to occur in landfills naturally. An initial step of chemical hydrolysis is required to biodegrade [24]. In addition, GHGs emissions would also occur when organic breakdowns take place in landfills. Another con of this method is the irretrievable loss of resources and lands since post-closure landfill sites cannot be reused due to engineering and/ or health risk issues. Hence, landfilling is ruled out as a reliable solution for a medium to long-term waste management system [39].

Environmental effects of biodegradable polymer

As bioplastics are mostly made up of biomass or renewable resources, the dependency on fossil resource usage can be reduced by replacing the fossil resources step-by-step with alternative resources [40]. The global issues revolving around plastic waste can be tackled by using biodegradable polymers as alternatives and replacing



most non-degradable polymers [2]. Besides that conventional plastics degradation generally requires decades, but 180 days or less for most bioplastics is needed to biodegrade. Apart from that, after being used as the bioplastic's raw materials, the polymer can be reused for energy generation or recovery, contributing to increased resource efficiency [40, 41]. In addition, as the conversion of biowaste (for certain biodegradable plastics) into methane occurs at the anaerobic digesters, the produced methane can be used to run generators for energy production [42]. Fabrication of biodiesel from recycled bioplastics are also possible in the future. The composted biodegradable polymers would then become soil nutrients to be absorbed back into the earth [5].

Another environmental benefit of bioplastic is the decrease of its carbon footprint and greenhouse gasses (GHG) emissions [40]. Also, up to 35% of energy could be saved due to its lower processing temperature requirement [5]. Agricultural products such as cellulose, starch and sugar have been used as bioplastic raw materials and have fewer plastics environmental impact, particularly in terms of CO₂ release [32]. Additionally, the carbon footprint of biodegradable polymers is lesser than conventional plastics as one metric ton of biodegradable plastics produces CO₂, between 0.8 and 3.2 metric tons lesser than conventional plastic[5]. Table 8 lists the impact of some polyester-based (bio) degradable polymers' impact on the environment.

PLA as biodegradable polymers

PLA is the most popular commercially used bio-based plastic due to its better product functionalities among polymers with comparable characteristics. Its inherent biodegradability made it possible to offer multiple end-of-life options, such as anaerobic digestion and industrial composting. These properties are very helpful in preventing organic waste from ending up in landfills or incineration [26]. PLA is a versatile material and could replace traditional plastic such as polystyrene and polypropylene [42].

Table 8 Positive impact of polyester-based (bio) degradable polymers toward environment [41]

Polymer	Positive impact on sustainable environment
PHA	Renewable natural resources-based polymer; produced by bacteria as storage material; living organisms made it commercially by using biochemical processes; non-toxic, biodegradable polymers, converted to the same metabolites as living organisms: water and carbon dioxide; reduce the usage of fossil energy ("old carbon")
PHB	Under appropriate conditions (in the presence of PhaZ7 depolymerase from <i>Paucimonas lemoignei</i>), the synthetic polymer biodegrades, forming monomer, dimer, and trimer
PLA	Synthesis from renewable monomer; polymers that easily hydrolytically degrade; lesser greenhouse gases release and less consumption of non-renewable energy compared to traditional polymers
Polyamides	Possible synthesis from renewable monomers. Bio-based polyamide and poly (ester amide) s thermoplastics of valuable properties



Production methods

PLA processing methods are well-established polymer-manufacturing techniques adopted from commercial polymers manufacturing techniques. But various control and application must be followed to get the full benefit of biopolymer [24, 43]. The common methods used to manufacture biopolymers are hot-melt extrusion, injection molding, blow molding and thermoforming, and the details of these techniques are as follows:

Hot-melt extrusion (HME)

In the HME process, heat converts plastic raw materials into melted form before turning them into a uniform shape and density product by forcing them through a die. This process is continuous and widely used in the production of polymer products with uniform shape and density. This method has been used in many diverse industrial fields, especially the food processing and plastic manufacturing industry. High viscosity is essential in the extrusion process to work it properly, unlike the casting process. The heat transfer and solidification would only occur if the melts could hold their shape long enough to exit the die [44, 45].

The equipment for HME commonly consists of an extruder and its auxiliary equipment, equipment for the downstream processing, and other monitoring tools used for evaluating the product and quality. Temperature, pressure, feeding rate and screw speed are controlled when polymeric substances are forced through the die. This melted polymer can obtain different products with various shapes and sizes, such as sheets, plastic bags and pipes. Figure 15. shows the schematic diagram of an extrusion molding machine. The extruder consists of one or two rotating screws (corotating or counter-rotating) poisoned centrally inside the stationary cylindrical barrel. The product's shape from the extrusion process is set by the end-plate die, connected to the barrel's end [44, 46].

Additionally, suppose the extrusion process is divided into four sections to complete its activities. In that case, it could be described as (1) Raw materials are

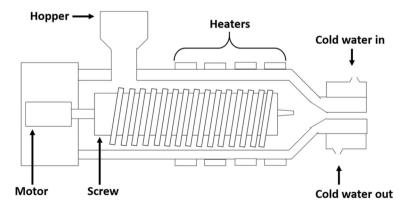


Fig. 15 Schematic diagram of extrusion molding machine [47]



through a hopper, (2) Mixing, grinding, particle size reduction, venting and kneading, (3) Flow through the die, and (4) Extrusion is done before being processed further downstream [44].

There are two types of extruders available for this process, namely single-screw and twin-screw extruders. The most used extrusion system globally is the single-screw extruder, as it is simpler and cheaper. Inside the barrel, the screw will work for feeding, melting, de-volatilizing and pumping. Depending on the designed manufacturing process, the single-screw extruder can either flood or starve fed. This type of extruder can generate thousands of pounds of pressure during the melting and mixing process, which helps mold the material in the desired shape [46].

The twin-screw extruder is similar to the single-screw type. Still, its operation is more complex as the flow mechanisms are depended on the type of operation (corotating vs counter-rotating) and screw design. The advantages of the twin-screw type are as (1) the material feeding and dispersion capacities are more accessible, (2) less tendency of overheating (3) and the transit times are shorter [44, 45].

Injection molding

One of the most effective methods for large-scale manufacturing of objects from thermoplastics is injection molding. This process does not require any additional finishing generally. In this process, initially, the thermoplastic polymer will be heated above its melting point. Then, the melt will be injected into the mold, where the product of the desired shape will be formed. The most important mechanisms in this process are pressure flow and heat transfer [48].

Figure 16 shows the schematic diagram of an injection molding apparatus. The process starts with the feeding of plastic pellets into the screw from the feed hopper, and the plastic that comes out from the screw is fully melted, desired and

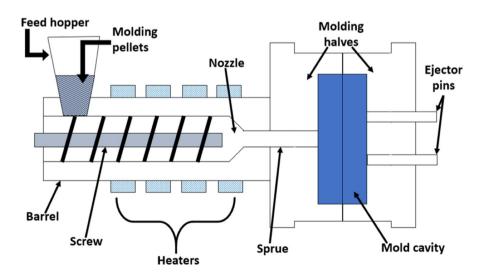


Fig. 16 Schematic diagram of an injection molding apparatus [49]



Fig. 17 Plastic injection molding cycle [51]

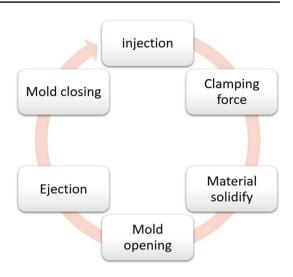


Table 9 Step-by-step operation of the injection molding process [45]

Step	Injection unit	Mold	Clamp unit
1. Mold Closing	The shot is prepared, screw rotating, check valve is open	Closed	Closed
2. Injection	Screw, nonrotating, pushes forward, check valve is closed	Closed, mold is filling	Closed
3. Hold On	The screw pushes forward, check valve is closed	Closed, packing continues, the polymer is solidifying	Closed
4. Shot Preparation and Cooling	The screw retracts and begins rotating, check valve is open	Closed, the polymer is solidified, cooling	Closed
5. Ejection	The screw continues rotating, check valve is open	Opened, the part ejected	Opened

dried by high pressure. The melt is filled up on a mold, and extra melts are also added to compensate for shrinkage. Two halves are clamped together to make up the mold cavity. One of the two halves of the mold is opened to eject the component when the component has been cooled off. Release agents might be applied in the molds to avoid the stickiness of the molded object. Typically, if good practice is maintained throughout the production phase, components made via this method do not require finishing. Hence, large quantities of small components manufacturing are possible at reasonable cost [49, 50]. Figure 17 illustrates the plastic injection molding cycle. As this is a cyclic operation and the repeated process cycle, parts are produced continuously but periodically [45]. Meanwhile, in Table 9, the steps of the injection molding cycle process are summarized.



Blow molding

The hollow three-dimensional products, especially bottles for the food packaging industry, usually use the blow molding technique. Basically, in this method, "parison" (a thermoplastic molten tube) is inflated or blown in the mold cavity's shape, and the hollow object is formed. After blowing, the parison possesses the mold shape and maintains that shape upon leaving the mold. In addition, there are three main thermoplastic processes blow molding. They are as blow molding (used to produce biaxially oriented jars and bottles that have greater strength, clarity and barrier properties), extrusion blow molding (PP bottles with hot-filling capability and good contact clarity), and injection blow molding (relatively small bottles and widemouth jars). Among these three processes, extrusion is the most commonly used process, followed by stretch blow molding and injection blow molding [50, 52, 53].

Blow molding has three main steps regardless of the type of materials used. Firstly, the parison is made up using a controlled quantity of melt. Then, a force is applied to the material to follow the mold. At the last step, to retain the shape, the molded object is cooled down. Depending on the requirement of the final product, either injection molding-based process or extrusion-based process has been used. As the extrusion blow molding process is the cheapest among the two techniques, containers and vessels that do not require exceptional precision and high properties are produced using this method.

Meanwhile, more demanding container applications (e.g., carbonated beverage bottles) are produced using injection blow molding. This injection molding also offers better mechanical and barrier properties [45]. Different blow molding processes with their characteristics are listed in Table 10.

Thermoforming

The following PLA processing is thermoforming. This process is commonly used to form parts that need simple features, such as packaging containers. Materials would be heated to a pliable temperature, and this malleable plastic possesses the

Table 10	Comparison extrusion,	, injection molding	and blow m	olding techniq	ues [54]
	_				

	Extrusion	Injection molding	Blow molding
Type of process	Continuous	Cyclic	Cyclic
Type of product	Extruded profile	Complex but not hollow	Hollow
Design complexity	Low	High	Medium
Forming mechanism	Viscous flow	Viscous flow	Inflation
Forming pressure	Medium	High	Low
Cooling mechanism	Two-sided convection or conduction	Two-sided conduction	One-sided conduction
Cooling rate	Medium	High	Low
Dimensional variation	Medium	Low	High
Tooling cost	Low	High	Medium



final shape [57]. The flowchart of the production process of a thermoforming part is shown in Fig. 18. By using infrared (IR) lamps, the initial heating of the PLA film would be done. Next, thermoforming of PLA sheets pass through aluminum molds to form the products [24].

Applications of PLA as biodegradable polymers

Compared to other aliphatic polyesters, PLA has shown many excellent properties such as high mechanical strength and modulus, biodegradability, biocompatibility and easy processing. The increment of PLA application is also related to enhancing its properties as heat-resistance modification, copolymerization and blending modification [23]. The different industries that used the PLA are as follows:

Medical/biomedical industry

The biomedical industries are among the sectors that incorporate PLA bioplastic in their products. Several interesting properties have made PLA an ideal material to be used for biomedical applications. One of them is its hydrolysis mechanism, where PLA naturally degrades in situ. Thus, additional surgeries are not needed to take out the planted device. Hence the patient's recovery rate can be improved, and the health system costs are also minimized. PLA's natural biocompatibility also helps to reduce the attainment of the critical immune response. As the degradation products are made up of lactic acids and short oligomers, these familiar materials can be metabolized by the body [6]. However, there might be some difficulty faced by pure PLA to fulfill all the needed requirements in this field. Hence, over the years, nanocomposites based on PLA have been extensively investigated as alternative materials. PLA-based nanocomposites are available with copolymers and nanocomposites as matrices [55]. Improvement in nanomaterials with its proper application, nanomedicine is an emerging field of medical treatment nowadays. These nanomaterials,

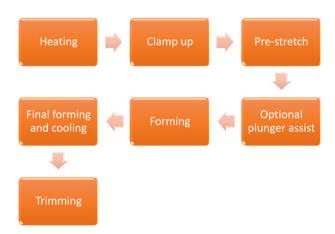


Fig. 18 Flowchart of the production process of a thermoforming part [24]



according to the FDA guideline, 2017, ranging from 1 to 1000 nm, is similar to molecules of biological interest such as proteins and viruses. As polymer nanoparticles can be absorbed by cells, thus opened many new potentials for the development of biomedicine, such as image contrast agents for diagnostic purposes, new media for the targeted drugs and vaccines delivery systems [6].

The properties of PLA are expected to be improved due to the surface and small-scale effect of nanomaterials. Hence, PLA-based nanocomposites are preferable to materials made from pure PLA in biomedical applications such as substitution and repairment of synthetic bone, drug delivery systems and tissue engineering. Poly (lactic-glycolic acid) (PLGA) has drawn the most attention compared to other PLA-based copolymers for clinical use. For the substitution and repairment of synthetic bone in orthopedic, oral, and craniofacial surgeries, plates, screws, and pins made of polymers have been used. PLA and PLGA are the two materials that have been extensively studied and used to produce porous scaffolds and repair damaged bones. Due to the PLA materials superior degradation, no need for more operations and implant removal can also be prevented, subsequently lessening the patient's pain. Apart from this, stress block can be avoided, and operational risk can be reduced using PLA. And compared to PLA-based nanocomposites, pure PLA materials lack bone-bonding force. Their efficacy to regenerate and degradation behavior are lacking as compared to PLA-based nanocomposites [55].

An emerging area in human health care is tissue engineering, which deals with the formation and regeneration of tissue and organ. In tissue engineering, scaffolds play a decisive role as they act as cell adhesion and growth carriers. Blood vessels, bone regeneration and the neural system are among the biomedical branches that use scaffolds to help with the treatment. Due to its good biocompatibility, PLA materials are extensively being used as scaffolds. PDLLA and PLGA are among the most widely used biopolymers in tissue engineering due to their wide range of melting and glass transition temperatures. In addition, the aligned nanofibrous PLLA scaffold has excellent capability in neural tissue engineering. In osteogenesis, PLLA has proven to be exceptionally useful in stimulating positive results. Gutierrez- Sanchez and colleagues had functionalized PLA scaffolds through adsorption of a tripeptide moiety that facilitated cell adhesion [55, 56]. The overview of PLA biomedical applications is listed in Table 11.

Packaging/food packaging

The next industrial application is the packaging field. Originated from lactic acid, PLA is a thermoplastic, biodegradable aliphatic polyester with plenty of potential for packaging application. The lactic acid monomer has two optical isomers, and the ratio between these two would determine the qualities of PLA as the packaging materials. For instance, high melting and crystallinity points would be obtained when 100% L-PLA monomers are used. Meanwhile, when 90/10% D/L copolymers are used, it will produce polymerizable melt above its Tg, satisfying the bulk packaging conditions. As a packaging material, PLA is a good alternative and replacement for conventional plastic such as low-density polyethylene (LDPE), high-density polyethylene (HDPE), polystyrene (PS) and polyethylene terephthalate (PET).



Table 11 The biomedical applications of PLA [6]

Field	Application
Orthopedic	Peripheral nerve and spinal cord injury regeneration
	Bioabsorbable screws
	Meniscus repair
	Guided bone regeneration
Cardiac	Chest wall reconstruction
	Stent
Dentistry	Guided tissue regeneration
	Biocompatibility space fillers
Plastic surgery	Suture
	Reconstructive surgery
	Dermal fillers
	Skin draft
General surgery	Hemia mesh
Gynecology	Stress incontinence mesh
Radiology	Theranostic imaging
Oncology	Nanoparticles for drug delivery

Good water solubility resistance, high molecular weight, good processability and biodegradability are attractive properties that make PLA an excellent packaging material. In terms of tensile strength modulus, flavor and odor barrier, PLA has similar polyethylene and PET or flexible PVC properties. Apart from that PLA has temperature stability and processability of PS, and polyethylene's printability and grease resistance. Additionally, for commercialization on a large scale, PLA is among the earliest bio-based polymer, and it can be shaped into injection molded objects, films, and coatings. Containers, films, and coating for paper and paper boards are among the forms of processed PLA. PLA can be recycled back to LA before undergoing repolymerization [3, 57].

In a study conducted by the Technological University of Denmark, it was observed that when PLA packages were used for butter, yoghurt, cheeses and margarine, the packages had excellent moisture barrier, mechanical protection, and protection fats, light, and gases. Additionally, during the biodegradation process, the lactic acid migration to the products is so minimal that it can be considered null. Hence, it was concluded that PLA-based food packaging is ideal for packing foods with short shelf life or high breathing, such as bakery, fruits and vegetables [11]. Apart from being used in food packaging, PLA has also been used in bottles for juices and water, but the market for this application is not as broad as the food packaging industry. This is because PLA has a low barrier toward CO2 and displays insufficient creep behavior, making the PLA application in the bottles productions limited to non-carbonated beverages [24].

Hence, due to the limitation in terms of barrier and mechanical performance, the usage of PLA for commercial packaging applications has many challenges. For example, compared to conventional plastics that are petroleum-based such as



PET, PLA is rigid, brittle. It has low gas barrier properties and is relatively sensitive to heat deformation. So, it is difficult to heat seal by this material. However, several ways can be implemented to resolve these difficulties, such as tailoring polymer processing, polymer blending and including compounds such as nucleating agents and plasticizers. Usage of nano and micro-composites and PLA coating with high barrier materials can also address some challenges. Gas and water vapor barrier properties are seen to be improved when PLA are coated with a thin layer of poly (ethylene oxide) or poly (\varepsilon-caprolactone) (PCL), and these modifications do not affect the PLA films' visual appearance [24, 56]. In Table 12, the list of applications of PLA as packaging materials and the company produced are being displayed [60].

Agriculture

PLA has also been used in the agriculture field. The incorporation of plastic for agricultural applications is called plasticulture and began in the 1950s to improve and increase production. There are several good reasons for the usage of plastic in agriculture, which are: (1) Soil protection from erosion and protection of plants from insects, birds and weed with mulch, (2) The utilization of drip irrigation tubing, and (3) shielding the greenhouses' tunnels.

Formerly, non-renewable, traditional plastics were used as the primary material and option in this industry. Still, as environmental effects continued, biodegradable plastics such as PLA, PHAs and PBAT evolved as the substitute. But pure PLA is not substantial for this industry since PLA's poor mechanical and thermal properties had limited the usage of homopolymer PLA in the plasticulture industry. Thus, commercialized PLA-based mulch films are made by blending PLA with other biodegradable polyesters. Typically, plasticizers are used to make commercialized PLA-based mulch films, and they are biodegradable as LA derivates or oligomers have been formed. However, plasticulture is still a developing field. Many areas need to be improved as the high cost of the polymers, the usage of bioplastics in agriculture is relatively new, and its execution is not matured yet [24].

Table 12 PLA as packaging materials and its applications [57]

Packaging application	Biopolymer	Company
Coffee and tea	Cardboard cups coated with PLA	KLM
Beverages	PLA cups	Mosburger (Japan)
Fresh salad	PLA bowls	McDonald's
Fresh cut fruits and vegetables	Rigid PLA trays and packs	Asda (retailer)
Potato chips	PLA bags	PepsiCo's Frito-Lay
Yoghurt	PLA jars	Stonyfield (Danone)
Bread	Paper bags with PLA window	Delhaize (retailer)



Automotive industry

Additionally, the automotive industry had also shown interest over the years in including materials that were natural-based in their parts. These materials demonstrate excellent performance and contribute to improving fuel efficiency as vehicles weight might be reduced. Reduction of the greenhouse gas (GHG) emissions into the atmosphere are also attained [58]. 23% of the total global carbon emissions are related to automotive and 80% of environmental pollution exhibited from automotive's total emission. The fuel efficiency would improve up to 7% with every 10% of the vehicle's entire weight reduction, and around 20 kg of CO₂ would be reduced from the environment with every 1 kg saved on the vehicle's weight. So twofold benefits are obtained when using biocomposite materials in this industry; increased fuel efficiency by lowering the total vehicle weight and decreased CO₂ emission on the environment [59]. Additionally, the approximate reduction of 40–55% vehicle auto-body weight could be achieved when nanocomposite materials are used instead of steel and aluminum auto-body. Mass decrease in automotive applications is essential for future electrical vehicles to optimize the ratio of the vehicle's weight to the battery's capacity [27].

Some of the existing bioplastics on the market are befitting to be used in the automotive sector, with PLLA and its composites. Excellent biodegradability/ recyclability and attractive mechanical and physical properties are a few of the key points which make PLLA a superb choice for this industry. However, the PLA has still lacked in brittleness and thermal stability. However, by using suitable modifiers and additives, such problems can be overcome. Many automobile manufacturers such as Ford, Mazda, Toyota and Hyundai Motors had incorporated bio-based PLLA blends to manufacture various components. PLLA could also be used as interior automotive parts. BIOFRONT using PLA could also manufacture door, tread plates and car dashboards [58].

PLA and bioplastic market overview

Now, NatureWorks, based in the USA, are the world's primary industrial-scale PLA production plant. It was set up in 2002 with a PLA line capacity of 70,000 metric tons. It expanded with 150,000 tons capacity in 2015. PLA plant in Thailand under the Total and Corbion's joint venture company is the second largest one with the production of 75,000 tons PLA. Meanwhile, in China, a 5000-ton PLA line was established by Hisun and added a 10,000-ton line in 2017.

Additionally, in 2018, several lactide-to-PLA fiber lines were built by Hengtian with a capacity of 10,000 tons, while COFCO installed a lactide-to-PLA plant with a total of 10,000 tons in the same year in China. Next, Synbra has set up a 5000-ton line to manufacture Expandable PLA (BioFoam TM) [1]. The current critical industrial of PLA projects has been listed in Table 13.

It is projected that by 2025, the world biodegradable plastic market will achieve \$6.73 billion from \$3.02 billion in 2018, more than double. The increasing demand



Table 13 Major PLA projects with an estimation of their capacities in 2019–2020 [1]

Main PLA projects in 2018–2020	Location of operation	KT
NatureWorks	USA	150
Total Corbion PLA JV	Thailand	75
Hisun	China	45
BBCA and Galactic	China	40
COFCO	China	10
Hengtian	China	10
SuPLA	China	10
TongJieLiang	China	10
Synbra	Netherlands	5
TianRen	China	3
Futerro (under Galactic)	Belgium	1
Jiangxi KeYuan	China	1
Sulzer	Switzerland	< 1
Pyramid (under Udhe)	Germany	< 1
Other announced projects (e.g., XinNing, HongDa, TongBang, YouCheng)	China	N/A

for biodegradable polymers in emerging economies like Brazil, China and India is the main driver for this dramatic increment. PHA and PLA are stated to be the major contributors to the expansion of bio-based biodegradable plastics. Market shares of 1.2% and 13.9% are occupied by PHA and PLA, accordingly. Both materials are expected to see an increase in global production, with PLA production increment from 293,290 tons in 2019 to 317,000 tons in 2024, equivalent to 8% of increased production [56]. Figure 19 shows the world production capacities of biodegradable

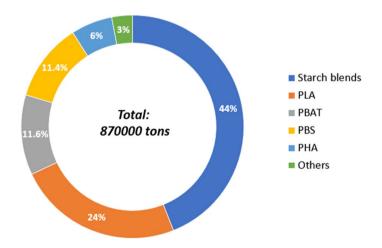


Fig. 19 World production capacities of biodegradable plastics in 2017 [2]



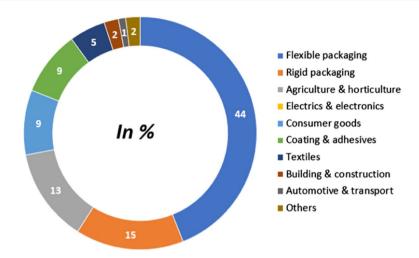


Fig. 20 Biodegradable plastic (by market segment) in 2018 [42]

Table 14 List of companies that manufacture PLA-based biodegradable films [3]

PLA-based (trade name/company/country)	PLA-/starch-based (trade name/company/country)
Ingeo (NatureWorks, USA)	Ecovio ® (NatureWorks, USA)
PURAC (PURAC Co., Thailand)	Bio-Flex (BASF, Germany)
BIOFRONT (Teijin, Japan)	Plantic (Plantic Co., USA)
HiSun (Revoda, Canada)	Biolice (Limagrain, France)
Pyramid (Tate and Lyle, Denmark)	Compole (Japan corn starch, Japan)

plastics in 2017. It can be understood that starch blends still dominate the global capacity for biodegradable polymers produced on an industrial scale by 44%, followed by PLA at 24%, PBS and PBAT totalled 23%, PHAs at 6% and others at 3% [2].

Meanwhile, Fig. 20 shows the biodegradable plastics (by market segment) in 2018. This figure indicates that biodegradable plastics are suitable for packaging applications since about 60% of the market in 2018 is accounted for flexible and rigid packaging [42]. Table 14 shows the list of companies that manufacture PLA-based biodegradable films.

Conclusion

Biopolymers and bioplastics have significant importance in research and development because of their eco-friendly alternative to conventional petroleum-based plastics. PLA is one popular bioplastic as it possesses excellent mechanical, structural, thermal and physical properties. Its production versatility with extrusion, injection



molding and thermoforming have made it rather attractive for manufacturing varieties of products, including in the biomedical, packaging and automotive industries. As these industries kept on growing, both PLA and bioplastic production markets would also increase. Following the combination of material's engineering properties and business potential, significant research and development could be carried out on the industrial applications of PLA, for example in biodegradable food packaging, biocompatible materials for medicine and lightweight automotive parts. However, it should be noted that the development of bioplastics is not isolated from academic researchers and the scientific community. So PLA is successfully used as the perfect substitute for conventional plastics and protects the environment as it is biodegradable.

Acknowledgements The authors would like to acknowledge Universiti Malaysia Sarawak (UNIMAS) for their financial and logistic support.

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