



A review on biodegradable composites based on poly (lactic acid) with various bio fibers

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Abstract

Green, or biodegradable, composite materials are gaining attraction because they are less harmful to the environment and have the potential to eventually replace traditional composite materials. PLA-based green composites have drawn more and more attention from researchers due to the growing ecological risk and environmental consciousness. Because of its excellent mechanical and thermal qualities, eco-friendliness, biodegradability, and antibacterial qualities, PLA has emerged as the most promising matrix material for sustainable bio composites. These composites have the potential to be more appealing than conventional petroleum-based composites, which are toxic and nonbiodegradable. Due to their cost-effectiveness and lightweight nature, composite materials are widely used in a wide range of applications in the structural, automotive, aerospace, and other household sectors. However, the recycling process of traditional fiber-reinforced plastics frequently poses environmental challenges. The mechanical characteristics of bio composites may be greatly affected by the kind of fiber employed in the fiber/matrix adhesion. Furthermore, the current state of PLA 3D printing with natural fiber reinforcement was outlined, along with its potential in 4D printing for uses of stimuli-responsive polymers. The purpose of this study is to highlight the progress made and investigations completed on bio-composites based on natural fibers and polylactic acid (PLA) during the last decade. The synthesis, biodegradation, properties, methods and possible uses of PLA-derived bio composite materials are all summarized in this article.

Keywords Bio fiber · Polylactic acid · Bio composites · 3D printing · Applications

Abbreviation

PLA	Poly lactic acid
ALK	Alkali treated fiber
TPS	Thermoplastic starch
GMA	Grafted maleic anhydride
ENR	Epoxidized natural rubber
FFF	Fused filament fabrication

Introduction

In an effort to encourage sustainable development, research into the use of natural fibers rather than synthetic ones, such as glass fibers, as reinforcing agents for composite applications is expanding. Natural fibers have several advantages over synthetic ones, including lower density, complete biodegradability, and recyclable nature (Bledzki et al. 1996). Natural fibers also offer comparable strengths and stiffness to synthetic fibers while coming from renewable resources, wearing down processing equipment less than synthetic fibers (Wambua et al. 2003). Two or more components with various characteristics that remain separate and distinct within a single unity are referred to as composites. Nanocomposites have demonstrated special qualities and a range of uses in the energy, electronics, environmental, and other fields. Green composites made of renewable resources have been developed subsequently (Naik et al. 2022; Duo et al. 2021; Hassan et al. 2021).

Green composite materials are being developed and manufactured to environmentally friendly and ethically

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replace traditional components in the automotive, aerospace, sporting goods, construction, and marine industries. Green composites are goods developed from sustainable forestry and agriculture feedstock, such as residues and byproduct crops. Depending on their origin, natural fibers can be classed as mineral, lignocellulosic or animal (Das et al. 2019). An increasing number of people are interested in polymer composites made of renewable and biodegradable plant-based resources, primarily from forests, due to worries about environmental damage and the incapacity to meet the demand for more adaptable, environmentally acceptable materials. These composites are sometimes called "green," and there are many different industrial uses for them. Green composites could be a good alternative to polymeric materials derived from petroleum because they are less harmful to the environment (Rahman et al. 2023). Over the past ten years, researchers have been interested in developing biodegradable materials and a great number of biopolymers have been developed and are currently being used in a variety of industries. Composites are widely used in industries such as transportation, automotive and aviation due to their remarkable properties and versatility (Amjad et al. 2021). Fiber-reinforced polymer composites have rapidly captured the commercial market in recent years. The reinforced polymer composite has various advantages, including the ability to conserve depleting sources of conventional materials such as metals and their alloys. Green composites have greatly expanded their applicability in numerous disciplines of engineering by replacing many engineering components comprised of synthetic fiber-reinforced composites. Green composites have received significant attention in both educational and manufacturing settings due to its appealing qualities such as low density, high specific strength, recyclability, economics and environmental friendliness. Green composites can be made with a range of natural and synthetic biodegradable polymers. Many reports and research articles have been published (Zini and Mariastella 2011).

Plant fibers provide high specific strength are renewable, sustainable and eco-efficient, making them desirable materials for use in industry. Their potential to absorb carbon dioxide indicates that they could be useful in reducing pollution in the environment (Yusoff and Takagi 2016). Natural fibers are commonly utilized as PLA reinforcement to improve mechanical and thermal performance while keeping its biodegradability. Several publications, for example, deal with fiber selection, geometry, optimum percentage and placement in the bio-polymeric matrix, and compatibility. Several fibers, including flax and kenaf, have been mixed with PLA to optimize performance for specific user applications (Gurunathan et al. 2015; Moliner et al. 2020).

Numerous investigations are being conducted with the goal of creating completely biodegradable composite constructions using a mix of PLA and natural fibers (Nazrin et al. 2020). Given that both PLA and natural fibers come from renewable resources and decompose and compost easily, composites made from these two types of materials are environmentally friendly and recyclable. According to Harussani et al. (2020), natural fiber-reinforced composites may be readily inclined of by incineration, landfill or by treatment (green) of pyrolysis; hence the bio-composites offer substantial benefits owing to their lower production and waste disposal treatment costs. Additionally, biopolymers may be effectively used in a variety of composite fabrication techniques, such as compression molding, extrusion and injection molding, although less study has been done on composites made from reprocessed raw substances with matrixes. Natural polymers are suitable natural materials as they are not one-time-use products. Composites reinforced with natural fiber will replace petroleum-based polymer composites due to their biodegradability and recyclable nature (Nazrin et al. 2020). Polylactide, also known as polylactic acid (PLA), is currently one of the most significant biopolymers when it comes to future developments and the expansion of global production capacities because it is made from renewable resources through the fermentation of polysaccharides or sugar, such as those extracted from maize or sugar beetroot, and the corresponding wastes (Harussani et al. 2020). In addition to the automobile industry, Poly Lactic Acid-based bio-composites have potential uses in building supplies, consumer items, the medical area and perhaps space travel (Faruk et al. 2014).

This review focuses on the PLA polymer's mass manufacturing state, the primary processing methods, and strategies for expanding PLA applications based on its inherent features. Additionally, this review offers a broad overview of the most common commercial applications for PLA today as well as a look at the various environments to which PLA products may be exposed over the course of their lifetime and which may cause degradation, containing hydrolysis in non-medical applications.

Bio-fiber

Bio fibers are gaining interest between researchers and academics for use in polymer composites due to their eco-friendliness and sustainability. Bio fibers are fibers that are not synthetic or man-made. They can be obtained from plants or animals (Murariu et al. 2022). Bio-fibers are prospective replacement materials for the composite industry because of their adaptability, eco-friendly model, cheap cost, renewability and local availability when compared to synthetic fibers (Dong et al. 2014a; Mohammed

et al. 2015). Despite the fact that the plants from which bio-fibers are generated, but not the fibers themselves are green and sustainable, they are neither organic nor sustainable in reality (Stevens 2010). The use of polymer composites with bio-fibers has also been elevated in the industrial sectors (Siakeng et al. 2018a). Fiber extraction method is shown Fig. 1.

The seven main categories of plant-based bio-fibers (lignocellulosic fibers) include leaf fiber (Agave, Abaca, Henequen, Banana, Sisal, PALF), stem or bast fiber (Flax, Banana, Jute, Hemp, Ramie, Kenaf), fruit fiber (Oil palm, Coir), seed fiber (Kapok, Cotton, Milkweed, Loofah), grass fiber (Corn, Bagasse, Bamboo) stalk fiber (Maize, Barley, Rice, Wheat, Oat), and wood fiber (hard and soft wood) based on the plant's component they are extracting (Ku et al. 2011). Figure 2. shows the classification of bio fiber.

To conserve the environment and biodiversity, specialists all around the world are trying to improve the sustainability and quality of eco-friendly products. Because of its bio-renewable properties and eco-friendly behaviors, people are turning to natural fibers to replace synthetic and harmful materials. To gain adequate utilization, some of the drawbacks of natural fiber, such as biocompatibility and hydrophilic properties, can be addressed through a range of surface modifications and chemical treatment processes.

Bio fibers have been effectively used in a wide range of applications, including composite materials (despite their lower density in comparison to glass fiber construction and engineering areas), textiles, biomedical, biopolymer, biosensors, and smart packaging. Natural fibers would help to reduce pollution issues such as trash, landfill, toxic, and greenhouse gas emissions (Choudhury and Debnath 2021; Jawaid and Abdul Khalil 2011; A review on natural fibers for development of eco-friendly bio-composite: characteristics et al. 2021). The main issues with the widespread use of these biofibers in various polymer matrices are their low fiber-matrix compatibility, poor dimensional stability, and intrinsically high water absorption capabilities, which cause thick swelling in biofiber-reinforced composites (Getme et al. 2020). Natural fiber problems like poor fiber/matrix adhesion, moisture absorption, low fire resistance, poor mechanical characteristics, low heat resistance, and restricted processing temperatures can be resolved with the help of fiber modification. Many strategies are used to address these problems (Siakeng et al. 2018a; S et al. 2021). Table 1. shows the types of bio fibers and their application.

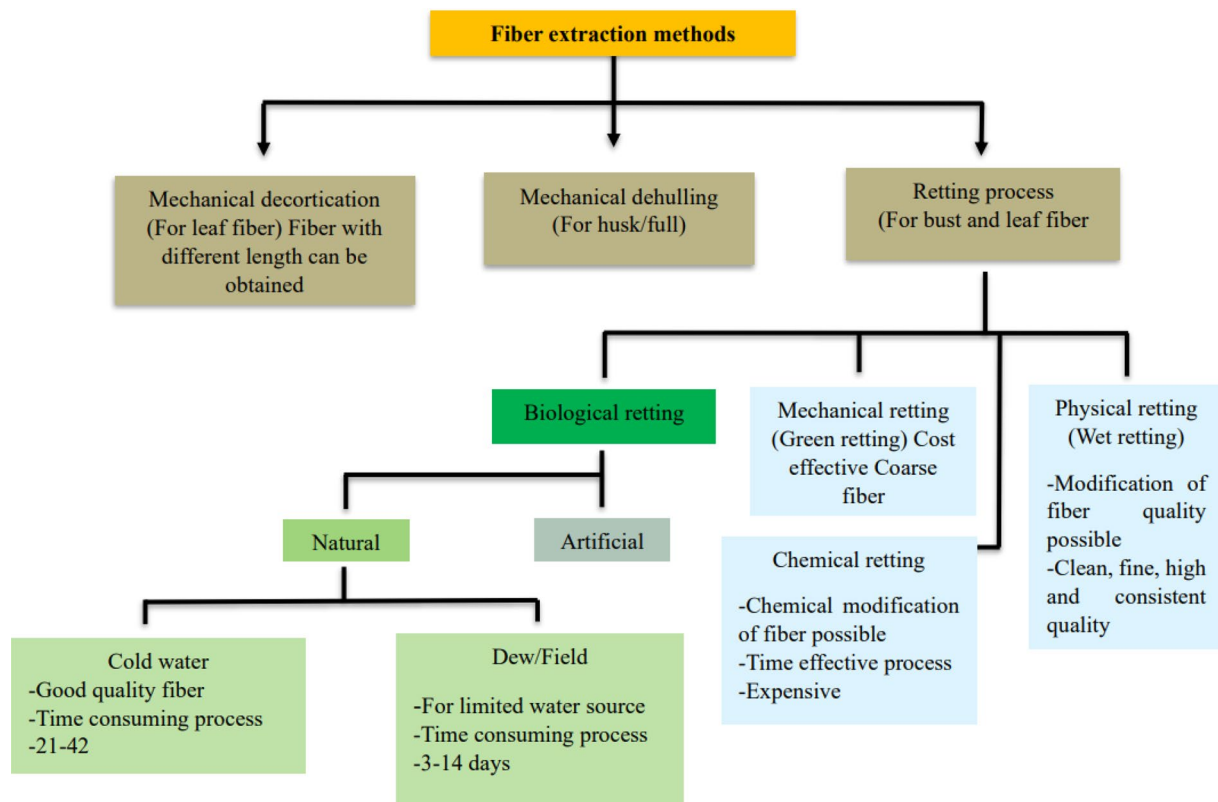


Fig. 1 Extraction Methods of bio fibers from (Faruk et al. 2012)

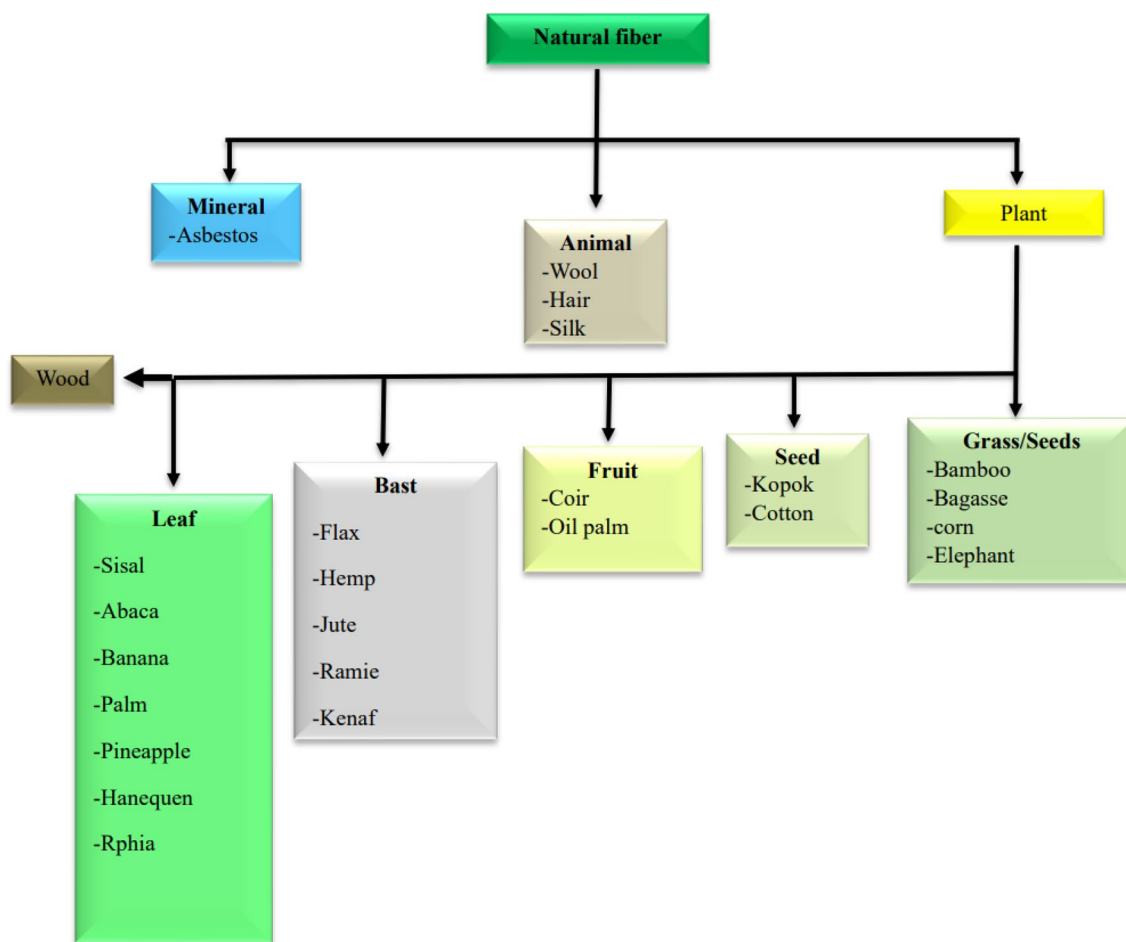


Fig. 2 Classification of bio fibers from (Faruk et al. 2012; Jawaid and Abdul Khalil 2011; Karimah 2021; Getme et al. 2020; Sathish et al. 2021)

Table 1 Types of fibers and their application from Choudhury and Debnath (2021)

Types of natural fibers	Applications
Abaca	Textiles, clothes, and useful papers such as money, journal, and check paper, as well as composites
Banana/Musa	Rope, place mats, paper cardboard, string yarn, tea bags, high-quality textile/fabric fabrics, currency note paper, mushroom, art/handicraft, cordage, cushion cover, table cloth, curtain, natural absorbent in colored wastewater, oil absorber, light weight composites, and bio-fertilizer
Coir	Filler, reinforcement in composite materials, light weight composites
Bamboo	Lactic acid, construction, vinegar, charcoal, methane, composite reinforcement, shoes, food, textiles, pulp and paper production, shocks, and bioenergy sources
Cotton	Fabric, clothes, yarn, furniture industry as coating materials
Hemp	Bags, tarpaulins, carpets, rope, furniture materials, fabric, textile, garden mulch, fleeces and needle felts, light weight composites, composites, geotextiles/geotextile insulation industry
Pineapple	Bags, table linens, mats, ropes, pulping material, handbags, composites, lightweight duck cloth, conveyor belt cord, coasters and many other interior design products, and livestock and agriculture
Sorghum bagasse	Particle board, sugar production sources, pulp, and paper
Ramie	Textile, paper, pulp, yarn, biofuel, fabric, oil, resin, wax, seed food, composites, livestock, and agriculture
Wool	Cotillion, wool yarn
Jute	Bags, sack, carpets, carpet upholstery, transportation or geotextile, electrical insulation and ropes, tarpaulins, packaging, furniture materials, fabric, light weight composites
Kenaf	Pulp and paper product

Chemical composition

Depending on the size of the fiber, the chemical makeup of bio-fibers changes. These green materials exhibit significant physical and chemical changes based on their botanical sources. Wood is a three-dimensional aggregation of bio-fibers made mostly of cellulose, hemicellulose, and lignin. Plant fibers have a complicated structure and chemical makeup (Zwawi 2021). Large fibrous cells are made of cellulose, a naturally occurring polymer with great strength and weight density. These cells may be found in a plant's seeds, leaves, or core. The three primary groups of bio-fibers are those made from plants, animals, and minerals (Sanjay et al. 2017). The majority of plant fibers are made up of waxes, lignin, cellulose, and water-soluble chemicals, with cellulose serving as the fiber's principal structural element (Bledzki et al. 1998; Eichhorn et al. 2001; John and Thomas 2008). Chemical compositions of some natural fibers shown in below Table 2.

Mechanical properties

These fibers offer excellent thermal and acoustic insulation qualities since they are hollow and lignocellulosic, much like their natural counterparts. Although the mechanical characteristics of bio-fibers are often inferior to those of synthetic fibers, these characteristics may be improved with careful surface treatment. Their low densities, cheap prices, and highly specialized modules draw a lot of interest from the industries. The common bio-fibers element (modulus/

absolute gravity) is comparable (even superior) to that of bio-fibers, despite the fact that the tensile strength of bio-fibers is greater than that of plant fibers (Azwa et al. 2013; Guzel Kaya and Devenci 2021; Madhavan Nampoothiri et al. 2010). Physical and mechanical properties of bio fibers show in Table 3.

Treatment of bio-fibers:

Many studies have been done to improve the mechanical and wear behavior by strengthening the bond between the polymer matrix as well as natural fiber reinforcement. The adhesive strength between the reinforcement and the matrix is significantly decreased as a result of the hydrophilic issue. Various tests were done on the surface of the bio-fiber to acquire the best qualities. Due to the weak link between the matrix and the fiber, the mechanical characteristics are reduced. The hydrophilic nature of the polymer matrix in bio-fibers is the cause of the weak bonding strength. It is required to treat bio-fibers in some way in order to solve these issues. Bio-fibers are subjected to chemical, physical, and biological treatments in order to reduce their hydrophilicity and remove moisture from their surfaces (Saba et al. 2014). When attempting to fill a research need in the field of natural fibers-reinforced PLA composites, Sawpan and colleagues (Koronis et al. 2013). Various physical and chemical treatments are shown Figs. 3 and 4, respectively.

conducted a review on improving the mechanical characteristics of industrial hemp-reinforced composites poly lactic bio-composites. Recently, testing of PLA for a range of random fiber material qualities has been devised that are quicker and easier. Interesting studies

Table 2 Chemical compositions of some natural fibers data obtained from Murariu et al. (2022), Mohammed et al. (2015), Siakeng et al. (2018a), Choudhury and Debnath (2021), Jawaid and Abdul Khalil (2011), Komuraiah et al. (2014)

Fiber	Cellulose (wt%)	Hemicellulose (wt%)	Lignin (wt%)	Pectin (wt%)	Wax (wt%)	Ash (wt%)
Jute	61–71	14–20	12–13	0.2	0.5	0.5–2
Wood	40–50	30–40	20–34	0–1	0.4–0.5	0.2–0.8
Flax	71–81	18–20	2–3	2.2–2.3	1.5–1.7	1.5
Hemp	70–77	18–22	3.7–5.7	0.9	0.8	0.8
Kenaf	45–57	12–22	8–13	3–5	0.8	2–5
Abaca	56–63	20–25	7–9	1	–	3
Cotton	85–90	5.7	0.7–1.6	–	0.6	–
Pina	81	7.1	12	–	–	2
Banana	63–64	12.1	5	–	–	2.2
Coir	32–43	0.15–0.25	40–45	3–4	1–2	2.7
Bamboo	26–43	30	21–31	0–0.2	1–2	1.7–5
Rice	38–57	19–33	8–20	10–15	14–17	10–20
Bagasse	33–55	17	18–25	–	–	1.7–1.8
Wheat	28–45	15–31	12–20	0–1	0.5–1	6–8
Sisal	66–78	10–14	8–14	10	2	0.6–1
Ramie	68–76	13–16	0.6–0.7	1.9	0.3	–

Table 3 Bio fibers with discussed physical and mechanical properties from Murariu et al. (2022), Mohammed et al. (2015), Siakeng et al. (2018a), Choudhury and Debnath (2021), Jawaid and Abdul Khalil (2011), Getme et al. (2020), Sathish and Loganathan (2021), Komuraiah et al. (2014)

Fibers	Density (g/cm ³)	Moisture content (%)	Tensile strength (MPa)	Tensile strength (MPa)	Young modulus (GPa)
Flax	1.5	7–12	350–1100	2–4	28–70
Kenaf	1.4	9–12	223–930	1.5–2.7	14–53
Ramie	1.5	7.5–17	400–1000	1.2–3.8	44–128
Pina	1.5	9–13	400–1627	1–3	60–82
Banana	1.4	8–12	529–759	5–6	27–32
Cotton	1.6	8–8.5	287–800	3–8	5–13
Bagasse	1.2	8.8–10	222–290	1.1–4	20–27
Wheat	0.7–1	5.1–8.3	55	2–5	22
Rice	0.6–0.8	8–9.1	10–200	2.7	1–12
Bamboo	1.3	8.8–8.9	140–230	4–7	11–17
Coir	1.2	8–10	108–252	15–30	4–6
Sisal	1.5	9–11	400–700	3–7	9–38
Abaca	1.5	7–15	418–813	3–10	31–33
Hemp	1.5	6–9	270–900	1.5–4	23–70
Jute	1.3	9–12	345–800	1.5–1.8	20–55
Wood	1.4	5–8.5	90–180	18–40	18–40

Fig. 3 Various physical treatments of bio fibers from (Karimah 2021; Getme et al. 2020; Sahayaraj et al. xxxx)

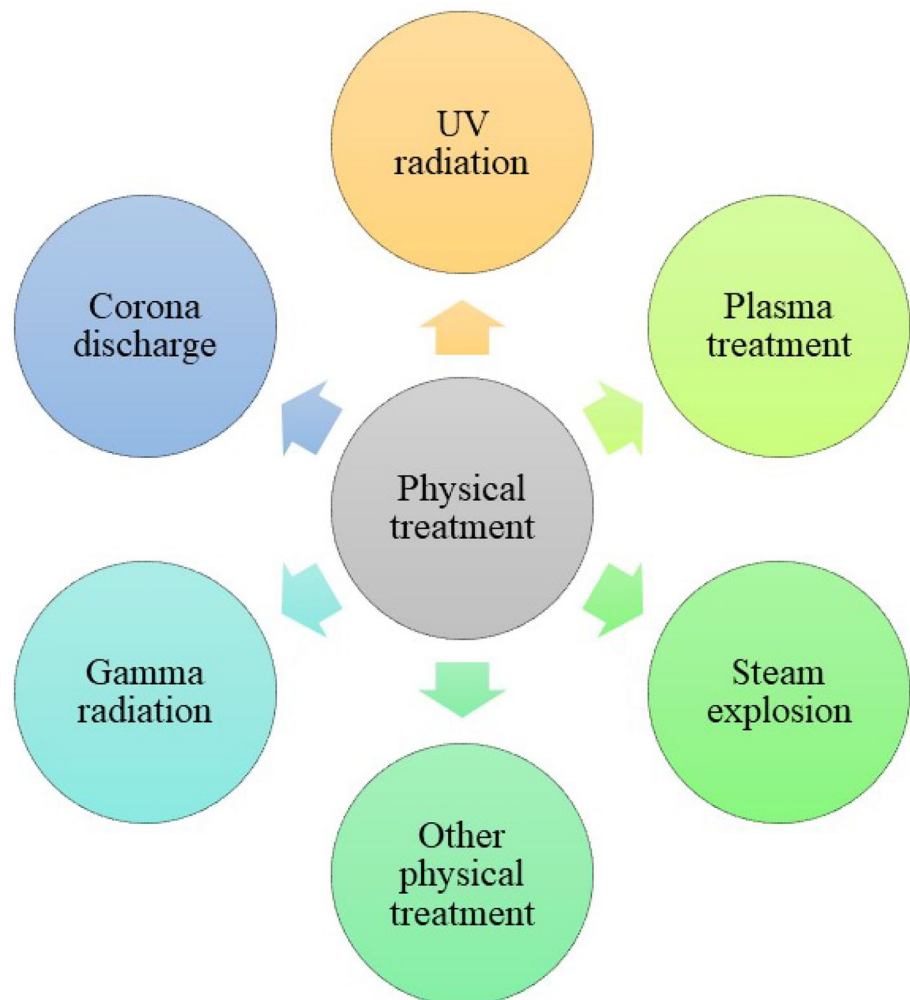
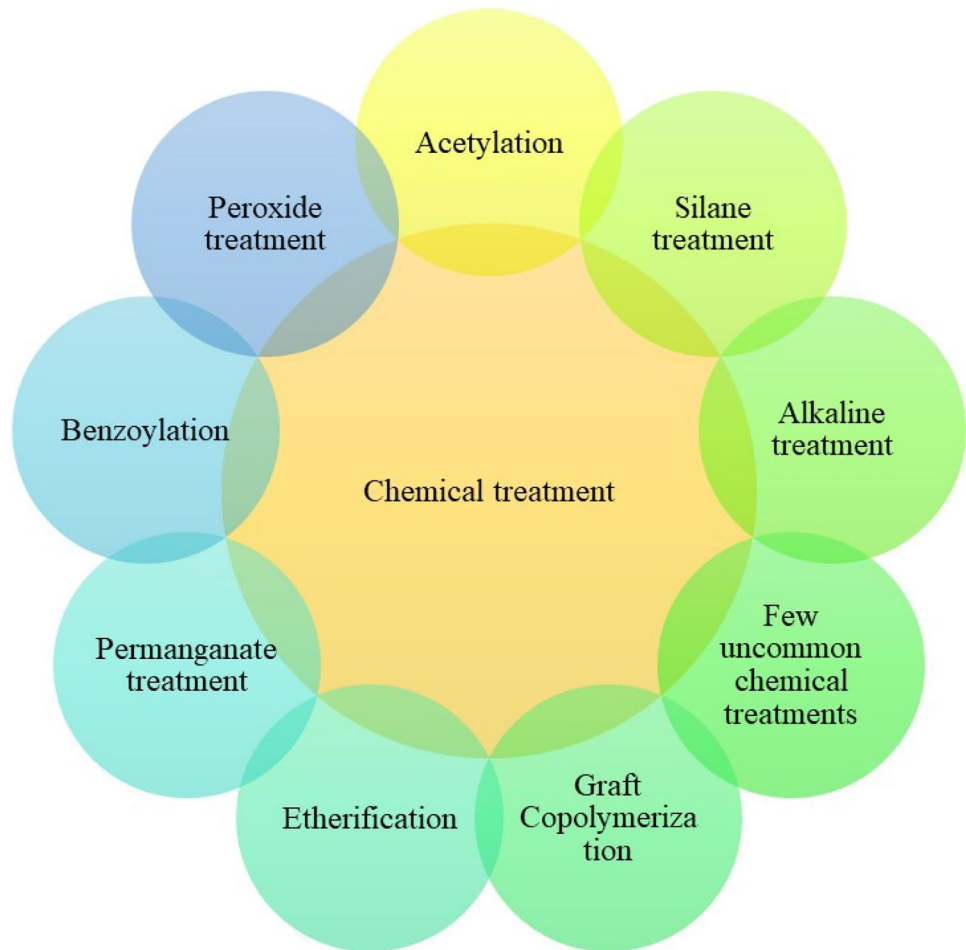


Fig. 4 Various chemical treatments for bio fibers from (Mohammed et al. 2015; Jawaid and Abdul Khalil 2011; Karimah 2021; Getme et al. 2020; Sahayaraj et al. 2021)



were conducted on aligned PLA composites reinforced with long hemp fiber (0–40 wt%). The 30% alkali-treated fiber-reinforced Polylactic acid composite (PLA/ALK) was found to have the maximum mechanical strength, measuring more than 70 MPa in tensile strength, more than 8 GPa in young's modulus, and 2.64 kJ/m² in flexural toughness. An established strategy for the configuration of long fibers and the advancement of ALK /PLA composites is the utilization of qualitative case studies. Table 4 shows the advantages and disadvantages of bio fibers.

Polylactic acid (PLA)

A biodegradable and bio based aliphatic polyester, poly(lactic acid) (PLA) is made from renewable resources such as maize sugar, potatoes, and sugar cane. For several applications, fossil-based polymers have been largely replaced by PLA (Devnani 2021). By providing more end-of-life options, PLA is seen as a possible solution to help with the municipal solid waste (MSW) disposal issue (Trivedi et al. 2023).

Table 4 Bio fibers advantages and disadvantages from Sawpan et al. (2011)

Advantages	Disadvantage
Low cost and abundantly available in nature	Poor compatibility with matrix
Biodegradable, renewable and sustainable	High moisture uptake, Tendency to form agglomerate
Low energy requirement for processing	Moderate strength
Zero impact on the environment	Poor wettability with resin
Non-toxic and recyclable	Properties vary with their origin
High specific strength	

Structure and properties of polylactic acid

PLA is one of the fastest-emerging biopolymers in the field of composite materials since it has certain features with a variety of polymers, including polyvinyl chloride, polystyrene, etc. (Lim et al. 2008 Aug 1). Comparing PLA's properties to those of common polymers indicates that PLA has a greater tensile modulus than PP, PVC, and nylon and a higher flexural strength than PP, showing that PLA has potential that can be used in a variety of technical domains. According to prior studies over the past two decades (Castro-Aguirre et al. 2016 Dec), PLA has a significant potential for enhancement in terms of its mechanical and physical attributes. PLA is a cheap and easily accessible polymer because it can be processed using a straightforward conventional method without requiring a lot of energy or time (Liang et al. 2021 Jan; Bajpai et al. 2014 Jan; Kühnert et al. 2017). The possibility of industrial applications makes the physical observation of PLA significant. Shiny and translucent, with low oxygen and water penetrability and good resistance of grease and oil, it maintains its stability at low temperatures. Due to these characteristics, it may be used to manufacture bottles, film, trays and cups (Ranakoti et al. 2019 May 29). Variable stereochemistry, a special property that can be applied to PLA, causes major variations in the characteristics and opens up new opportunities for customizing the properties, which is something that sets PLA apart from other petrochemical polymers. According to the ratio of the D- and L-content, stereochemistry results in semicrystalline and amorphous chemical structures in the PLA (Ranakoti and Rakesh 2020). The chemical composition of PLA affects its physical, mechanical, and biodegradation rates. The D-content is a crucial characteristic that enables the alteration of the PLA's properties and an increase in crystallization rate, which lowers the melting point (Pantani et al. 2014). Table 5 shows the advantages and disadvantages of poly lactic acid.

Production of PLA:

It is possible to create PLA, a thermoplastic polymer with high strength and modulus, using renewable resources like maize and sugar beets. It is a member of the aliphatic polyester family and is thought to be compostable and biodegradable since it is often manufactured from hydroxy acids like polyglycolic or mandelic acid (Kuru and Mehmet. 2023) (Table 6).

There are many processes used to make PLA, Viz. enzymatic polymerization, poly-condensation, azeotropic dehydration and ring opening polymerization are some of these techniques (Ranakoti et al. 2022). The most popular techniques among them are ring opening polymerization and direct condensation of lactic acid. The world's biggest producer of PLA, Cargill Dow LLC, created a proprietary, low-cost continuous method for making lactic acid-based polymers (Garlotta 2001). This method produces PLA in melt rather than solution, which has significant positive effects on the environment and the economy. Dextrose is first produced by converting natural or renewable resources like sugar beet, potatoes, maize, etc. Lactic acid is produced by the fermentation of the dextrose. Lactic acid is converted to lactide in the presence of a catalyst. The molten lactide is further refined using vacuum distillation. Subsequently, it goes through a polymerization step to create PLA. The remaining lactide monomer is reused in subsequent steps. In addition to these advantages, the manufacture and use of PLA also significantly decrease greenhouse gas emissions and energy consumption. To guarantee a sustainable ecosystem, these variables are crucial (Ajioka et al. 1995). The total energy required to produce one kilogram of PLA is 75.4 MJ/kg. This covers the whole procedure, starting with the growing of the corn and ending with the transportation of the PLA pellets. The usage of fossil fuels and renewable energy sources may be included to the total energy consumption. The portion made up of renewable energy makes up 24.6 MJ/kg. The fossil energy usage, on the other hand, is 50.8 MJ/kg, and the main sources of this

Table 5 Advantages and disadvantages of poly lactic acid from Farah et al. (2016)

Polymer	Advantage	Disadvantage
Polylactic acid	100% Biodegradable Anti-bacterial and anti-fungal properties, bio-compostable Good mechanical and physical properties Gloss and transference are fine Processing ability Blended with petroleum-based polymer Easy to 3D printing application	High cost Permeability to moisture and oxygen is high, low heat resistance Low melting point. A slow rate of deterioration Impact resistance and hardness are weak Not suitable for high-temperature environment Not suitable for long-term food storage application

Table 6 Mechanical properties of PLA composites

Composition	Tensile strength	Preparation method	References
PLA-30percent rice straw	22.27 MPa	Solvent casting	Ogin et al. (2016)
PLA-5percent Lignin	48.39 MPa	Twin-screw micro compounder	Diyana et al. (2021)
PLA-30percent kraft lignin	25.3 MPa	Extrusion	Ghorbani Chaboki et al. (2019)
70percent PLA-20percent PBAT-10percent office wastepaper	49 MPa	Injection molding	Gao et al. (2019)
PLA-8percent oil seed fillers	62.6 MPa	Co-rotating twin-screw extruder	Bassani et al. (2019)
PLA-60percent kenaf	5.2 MPa	Brabender mixer and hot press machine	Xu et al. (2019)
PLA-30percent okra fiber	58.4 MPa	Co-rotating twin-screw micro extruder	Finkenstadt et al. (2007a)
PLA-63percent cellulose-2.9percent starch-24percent carnauba wax	3.27 MPa	Mixing, blending, followed by compression molding	Islam et al. (2017)
PLA-50percent pine wood flour	66.2 MPa	Counter rotating twin-screw micro extruder	Fortunati et al. (2013)
PLA-10percent coir fiber	57.9	Twin-screw extruder	Ranjeth Kumar Reddy and Kim (2019)
PLA-30percent ramie fiber	53 MPa	Compression molding	Altun et al. (2013)
PLA-50percent jute fiber	32.3 MPa	Compression molding	Zhang et al. (2017a)
PLA-hemp fiber	72.1 MPa	Twin-screw extruder	Yu et al. (2014)

Table 7 Published research on the dynamic mechanical study of hybrid biopolymer composites reinforced with natural fibers data obtained from Hazrol et al. (2020)

Reinforcement	Bio-polymer matrix
Flax/basalt fiber	Poly (lactic acid)
Hemp/sisal fiber	Biodegradable epoxy
Sisal/corn fiber	Poly (lactic acid)
Flax/jute fiber	Poly (lactic acid)
Oil palm/kenaf fiber	Poly (lactic acid)
Cotton/starch	Poly (lactic acid)
Hemp/sisal fiber	Poly (lactic acid)
Basalt/cissus quadrangularis fiber	Poly (lactic acid)
Cassava bagasse/sugar palm fiber	Cassava starch

energy are oil, coal, gas, and nuclear power (Drumright et al. 2000) (Table 7).

PLA biodegradation

Algae, fungus, and bacteria are necessary for the PLA to biodegrade spontaneously. Figure 4 illustrates the breakdown of the PLA's chemical structure into the water, carbon dioxide, and inorganic chemicals. Additionally, some biomass is created throughout the process and may be used as manure. When the residue created is invisible and non-toxic, the terms "biodegradability" and "composability" are interchangeable (Scaffaro et al. 2019). Only under certain circumstances controlled by humidity, pressure, and temperature is biodegradability guaranteed. Environmentally unfavorable circumstances will slow down biodegradation,

which even in the presence of enzymes may produce residue that is different from that of natural biodegradation. The two stages of PLA biodegradation are homogeneous (intramolecular degradation) and heterogeneous (surface degradation). Any polymer may degrade in one of three ways: first via an intersectional chain, second through the main chain, and third through a side chain. The lengthy polymer chain of PLA is broken down into shorter monomers, dimers, and oligomers, which are exactly known as carboxylic acid and alcohol, by the emission of ester linkages during biodegradation. In contrast to PLA with lower molecular weight, PLA with greater molecular weight degrades more gradually. The rate of degradability of PLA will decrease when its melting point rises (Vink et al. 2007). There are two steps to PLA's biodegradation in aerobic circumstances. Water diffuses through the hydrolysis chain in the first step; in the second stage, microorganisms attack the frail molecular chain and break it down into H₂O, CO₂, and microbial biomass. It should be noted that only when the molecular weight drops to 10,000 Dalton or below can an enzymatic assault on the PLA take place. Cutinase, lipase, subtilisin, alkaline proteases, elastase and trypsin are a few of the enzymes that are important in the quick destruction of PLA (Wang et al. 2021). These enzymes are easily accessible in the bodies of bacteria, fungi, and algae (Fig. 5).

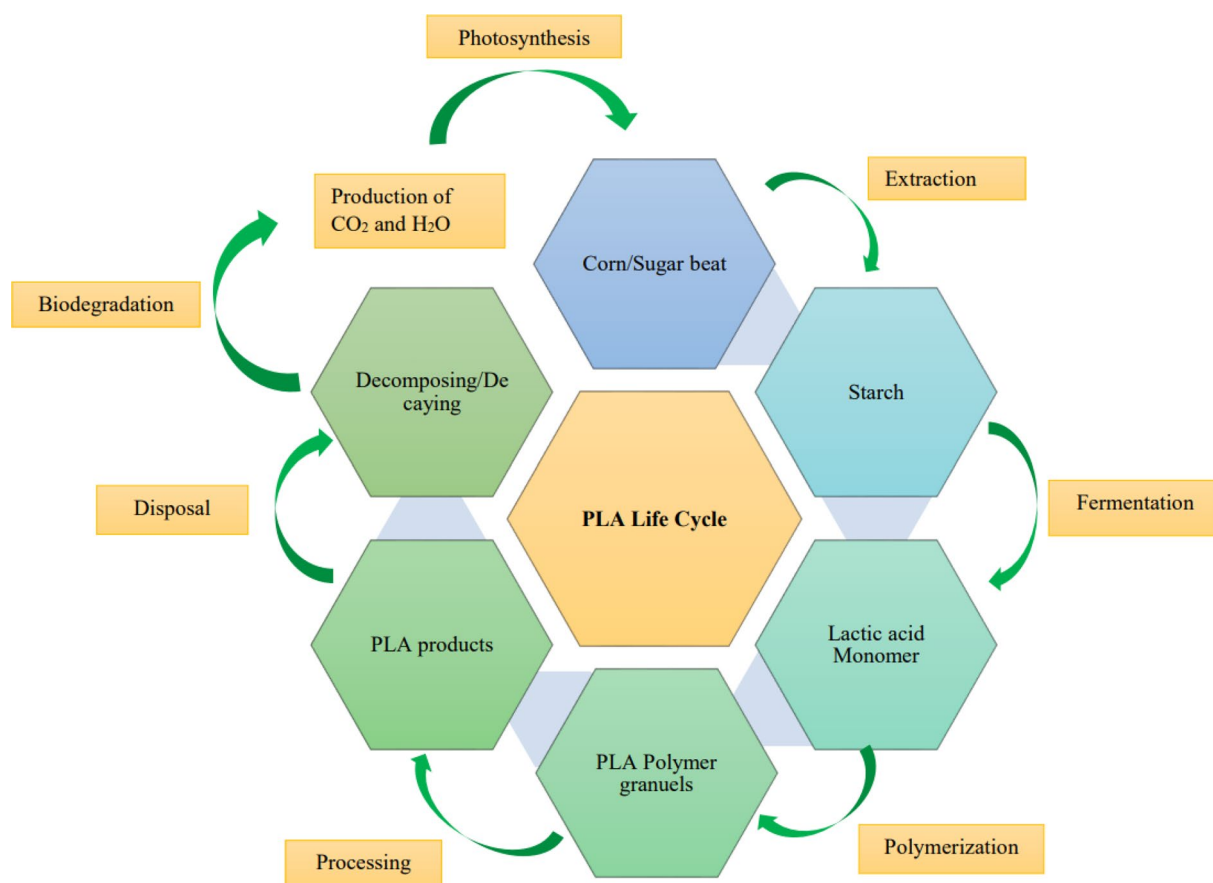


Fig.5 Life cycle of PLA from (Sawpan et al. 2011)

Composite

Materials made by two or more stages and isolated by various interfaces that have diverse chemical and physical properties are referred to as composite materials (Shahar et al. 2022; Ahmed et al. 2018a). To achieve a system with more value functional or structural features that cannot be attained by each of the components alone, the different systems are carefully integrated (Nurazzi et al. 2019; Abrial et al. 2020). In general, composite materials have unique properties with a high strength-to-weight ratio. Since composites may be formed into intricate designs, another advantage of composite material is that it provides

structural adaptability. The bulk of the beneficial qualities of composite substances are obtained by a strong link between a stiff reinforcement—typically fibers (filaments) or reinforcements with different geometric shapes, like platelets, particles and a flexible matrix (Mohd Nurazzi et al. 2021). Numerous research have started to look at PLA composites with natural fiber reinforcement. The investigation was focused on the variables affecting tensile strength performance. According to Ku et al. paper's (Gurunathan et al. 2015), the tensile strength of composites including fibers was discovered to be 20–40% lower in the perpendicular way than that of composites containing fibers in the parallel way. The average score for natural fiber was that it could replace conventional

Fig.6 Chemical structure of polylactic acid; **b** isomers of lactic acid (González-López et al. 2020)

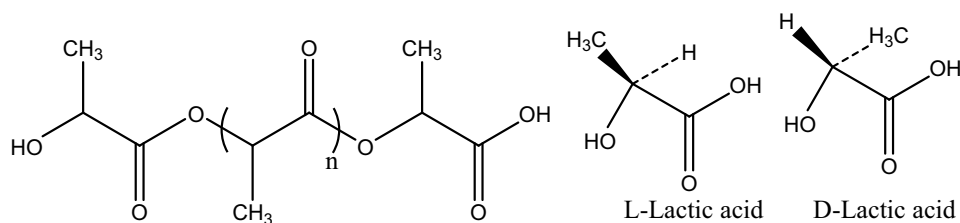


Table 8 Application of DMA obtained from Hazrol et al. (2020)

Polymer properties & characterization	Polymer composite characterization	Industrial applications
Polymer rheological and thermal properties	Investigation of an ideal curing schedule of Fiber-reinforced polymer composites	Paints and lacquers industry
Melting point of semi-crystalline polymers	Mechanical, viscoelastic properties, melting point, vulcanization in elastomeric polymer composite	Structural pipeline repair
Polymer blends and phase morphology	Storage and loss moduli of polymer composite	Chemical industry
Effect of orientation on the mechanical properties of solid polymers	Sol gel transformation in polymer composite	The curing reactions and Tg of the materials
Polymer and polymer compatibility	Evaluation of the interfacial bonding in polymer composites	Melting point, dynamic modulus, Tg of chemicals
Rate and extent of curing properties of thermoset resins	Characterization of the thermo-rheological properties of gel systems	Oil and gas industry
Polymer storage and loss moduli	–	Food industry, Glass transition and gelation point, Automotive industry Curing reactions, damping behaviour, dynamic modulus of auto and aerospace components
Polymer damping properties	–	Optimization of the formulation of pharmaceutical drug delivery systems

fibers in the strengthening of plastic content. The tensile characteristics of the composites are enhanced for natural fiber reinforcements via matrix interfacial bonding, novel manufacturing processes, physico-chemical modification approaches (Fig. 6).

Bio-fiber-reinforced PLA composites

The search for a truly recyclable composite with acceptable mechanical qualities seems to be over by the development of PLA bio-composites (Table 8). Numerous bio-composites were created using a variety of production techniques to examine the ability of PLA as a polymer matrix. In an injection molding machine, sugar beet pulp of 7–40 weight percent was combined with Polylactic acid in an attempt to create bio-composites (Singh et al. 2018). The inclusion of the sugar beet pulp boosted the PLA's Young's modulus by 45% while lowering its tensile strength by 29%,

according to the results. The short fiber's length caused an ineffective stress transfer process between the matrix and fibers, which was blamed for the bad outcome (Table 9). When reinforced with oil seed fibers, the PLA's mechanical characteristics were reduced by 50% (Finkenstadt et al. 2007a). The mechanical qualities of PLA were diminished as a consequence of the improper manufacturing process' distribution of reinforcement (Fig. 7).

Previous studies on the natural fiber have placed a lot of emphasis on reinforcing it using polymer composites. An acceptable degree of bond between the polymeric resin and the surface of hydrophilic natural fiber cellulose is normally guaranteed, as shown in research on natural fiber surface changes and the performance of the resulting bio-composites by Mohanty and colleagues (Mohanty et al. 2001). Treatments such as peroxide, alkali, isocyanate and coupling agents, along with bleaching, dewaxing, acetylation and vinyl grafting may improve the properties of natural fiber composites. The crystalline structure of cellulose may

Table 9 properties of popular bio-fiber-reinforced PLA composites

PLA bio-composite	Processing	Fiber (%)	Tensile strength (MPa)	References
Flax	Compression molding	30	53	Oksman and Selin (2004)
Cotton	Compression molding	–	4.12 2	Pradhan et al. (2010)
Ramie	Compression molding	30	52.5	Oksman and Selin (2004)
Kenaf	Compression molding	30	32	Oksman and Selin (2004)
Corn stover wheat straw	Extrusion injection	30	58	Pradhan et al. (2010)
Coconut	Extrusion compression molding	0.5	67.99 3.75	Pradhan et al. (2010)
Jute	Compression molding	30	48	Oksman and Selin (2004)
Hemp	Injection molding	30	75	Oksman and Selin (2004)

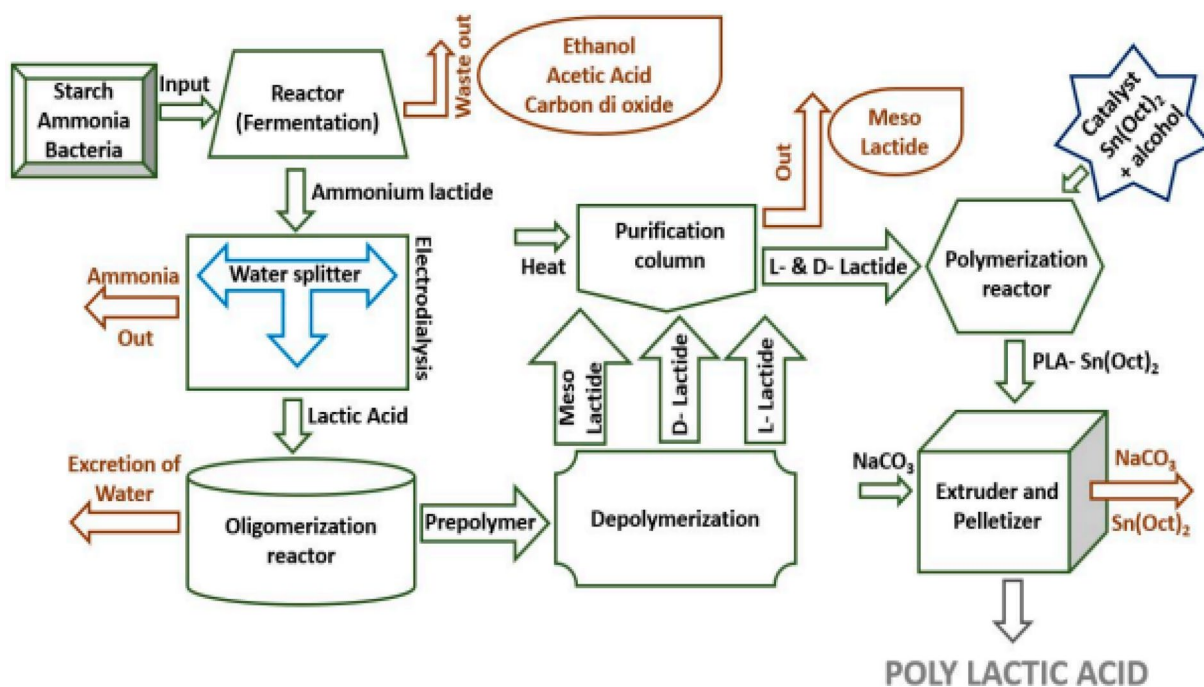


Fig.7 Production of polylactic acid procedure (González-López et al. 2020)

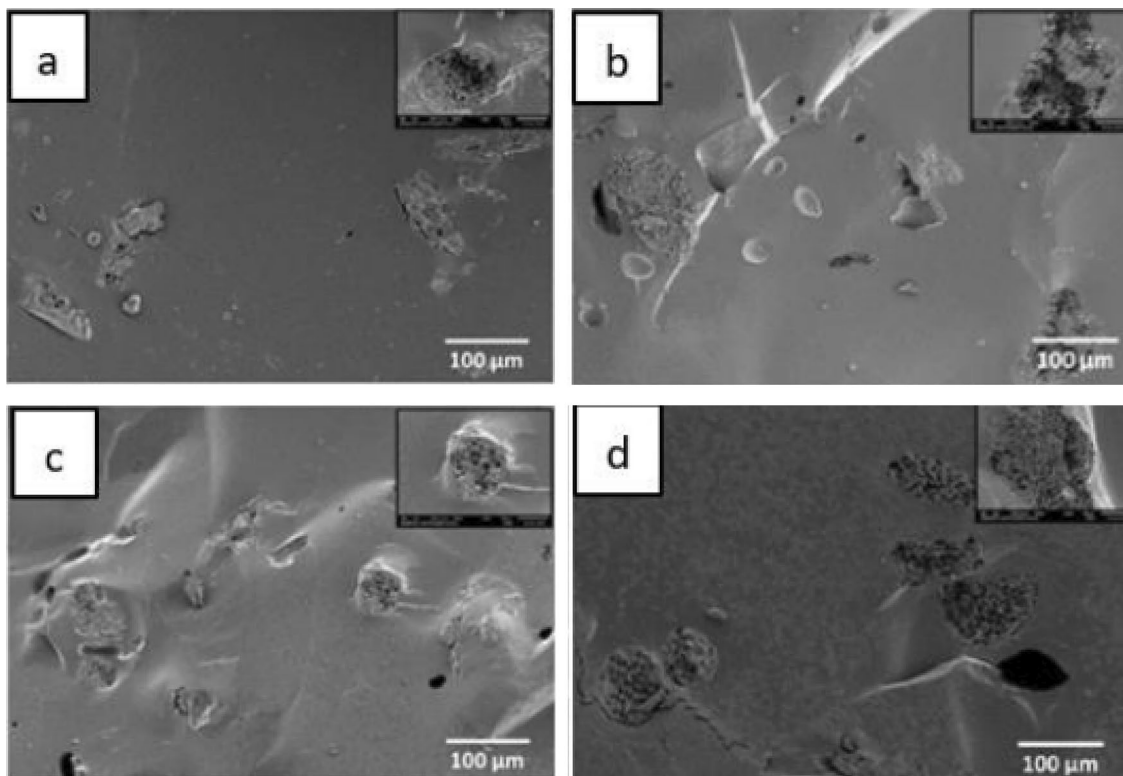


Fig.8 Micrography of **a** thermoplastic starch (TPS) 5% raw fiber, **b** TPS 5% alkali treated fiber, **c** TPS 10% raw fiber, **d** TPS 10% alkali treated fiber, (Amir et al. 2019)

be disrupted by substituting a chemical function for the hydroxyl group. Because the substituted groups serve as the plasticizer in this de-crystallization process, the cellulose's thermoplastic properties are improved (Martinez Villadiego et al. 2022 Jan) (Fig. 8).

The use of DMA to investigate fiber/matrix interactions

Composites made of natural fibers have emerged as a new standard for replacing traditional materials. Dynamic mechanical analysis (DMA), a flexible methodology that supports the more conventional procedures, is one of the newest techniques developed to suit this new tradition of material testing. DMA is a method for assessing the viscoelastic properties of materials, particularly polymers and composites, by measuring the stress or strain induced by a dynamically varying tension or strain given to the sample. While analyzing the temperature response, dynamic mechanical thermal analysis (DMA) is another name for DMA. The time, temperature, or frequency dependent composite material DMA parameters of relevance, as well as storage modulus, loss modulus, and damping factor. The elastic behavior and stiffness of a material are explained by its storage modulus (E_0), often known as its dynamic modulus (Fig. 9). Young's modulus and this parameter have a theoretical relationship, but they are not the same (Mohanty et al. 2001; Hazrol et al. 2020).

Brief on PLA composites in brief

Global production of PLA, a thermoplastic polymer made from naturally occurring resources, is expected to reach 211,000 tons in 2020 (Ashok et al. 2019). Additionally, the production of chitin and chitosan compounds was about 107,000 tons (Haris et al. 2022) whereas the capacity of cellulose and PHA production in the world was over 30,000 tons, and 580,000 tons respectively, in 2020 (Ganesh Saratale et al. 2021). According to Jem's law (Jem and Tan 2020), the worldwide market for PLA is predicted to quadruple every four years. This is corroborated by the fact that widespread research into the use of biodegradable items has been sparked by environmental contamination brought on by excessive manufacturing of plastics made from petroleum and by pressure from global warming. Natural fiber-reinforced polymer composites were used in the automobile sector in a landmark work by Holbery and Houston (2006). Natural fibers, including hemp, kenaf, jute, sisal and flax, were shown to have advantages in terms of weight, cost, CO₂ reduction, and reduced reliance on external oil supplies. In order to create the composite, natural fiber preforms or mats were combined with a thermoplastic binder method. Research on bio-composites reinforced with natural fibers was created by Faruk et al. (2016). The kind of fiber, climatic conditions, manufacturing techniques, and surface modification of the fiber are only a few of the variables that may have an impact on the bio-composites' physical, thermal, and mechanical characteristics. These processes are influenced by a wide variety of bio-composite

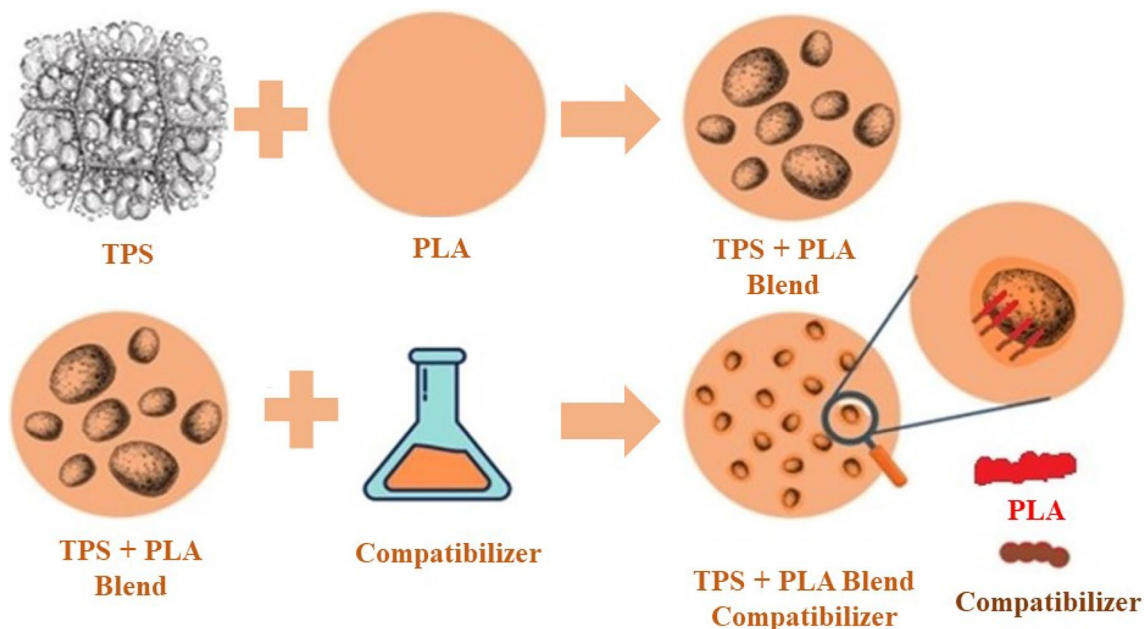
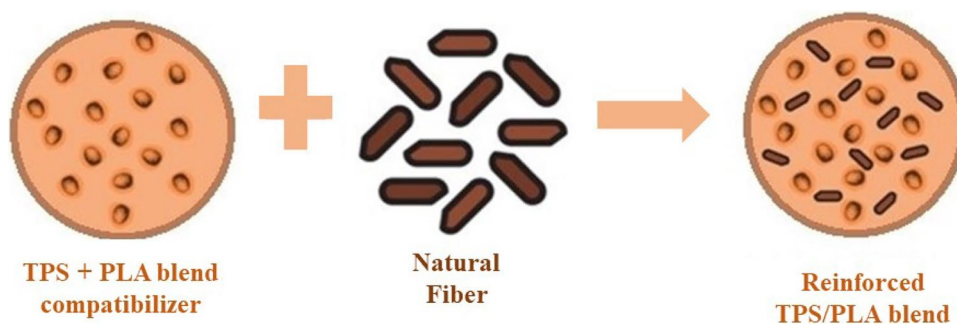


Fig. 9 Compatibilization process of TPS/PLA blends (Song et al. 2013)

Fig. 10 Reinforcement with natural fibers of TPS/PLA blends (Song et al. 2013)



processing methods and factors, including moisture quality, fiber shape and content, binding agents, and their impact on the characteristics of composites (Fig. 10).

Development of PLA bio composite

Numerous research have systematically examined natural fiber-reinforced polylactide (PLA) composites. An increasing corpus of research has highlighted the value of creating composite materials by mixing biodegradable polymers with plant fibers. Several research have examined the effectiveness of natural fiber reinforcements in PLA composites. Studies have been done on reinforcing PLA-based composites utilizing natural fibers derived from leaves, such as sisal and leaf fiber. The effect of benzoyl peroxide surface treatment on the mechanical characteristics of BSF-reinforced composites of Poly lactic acid was examined based on earlier study by Asaithambi and colleagues (2014a) on reinforced PLA hybrid composites with BSF. In their study of the tribology of PLA composites augmented with natural fibers, Bajpai et al. (2013) discovered that adding natural fiber sheets to the PLA matrix considerably improved the parameters of neat polymer wear. Natural fibers, including nettle, *grewia optiva*, and sisal were combined with the PLA polymer in order to make a laminated composite utilizing the hot compression method. The working conditions were established such that the applied weights ranged from 10 to 30 N, the sliding speeds ranged from 1 to 3 m/s, and the sliding lengths ranged from 1000 to 3000 m. According to the experiment's findings, the friction coefficient was reduced by 10–44% compared to plain poly lactic acid, and the true wear rate of the composites was increased by more than 70%. The mechanical properties of synthetic and natural cellulose fibers reinforced with PLA composite were studied by Graupner et al. (2009). Compression molding was used to create several kinds of natural fiber composites, such as kenaf, cotton, hemp, and human-made fibers of cellulose, with a fiber mass of 40% and the addition of PLA.

Non-wood jute-derived natural fiber polymer composite reinforcement with PLA is being investigated in a few

research (Graupner et al. 2009). Jiang et al. (2019) research on hydrothermal aging as well as structural degradation discovered by X-ray tomography in jute/PLA composites. According to the research, biodegradable PLA composites reinforced with natural fiber might take the position of traditional composites reinforced with synthetic fiber. However, these composites may age more quickly due to the combined impacts of heat and moisture. Oksman and Selin (Oksman and Selin 2004) found that PLA plastics and composites demonstrated that PLA may be utilized as the matrix in a composite system where natural fibers are employed as reinforcements. It has been discovered that PLA composites reinforced with flax are 50% stronger than several other thermoplastic composites bonded with flax that are presently utilized in automobile panels. These PLA composites may be readily extruded and compacted. The rigidity of the PLA was raised from 3.4 to 8.4 GPa by using 30% flax fibers. The composability and biodegradation rate of soy straw/PLA and wheat straw biocomposites were examined by Pradhan et al. (2010). They observed that PLA composites for untreated soy and wheat straw were shown to be obviously biodegradable materials. The presence of the natural biomass slows down the degradation of the PLA component, showing that composites may employ modified/treated materials. Additionally, Omar et al.'s work (2020) on the use of kenaf fiber-reinforced composite in the automobile sector was quite instructive. Recent developments in kenaf fiber-reinforced composite were examined in the research.

Processing method developments of PLA bio-composite

Numerous prior natural fiber investigations have focused on the use of PLA composites with natural fiber reinforcement. Natural fiber-reinforced composite laminates were researched by Jauhari et al. (2015). By building the short or long bundles of natural fibers, scientists have been capable to explore natural fiber reinforcing. In terms of polymeric composites, it produced a flat sheet made up of one to 10 layers of fibers. Different approaches were put

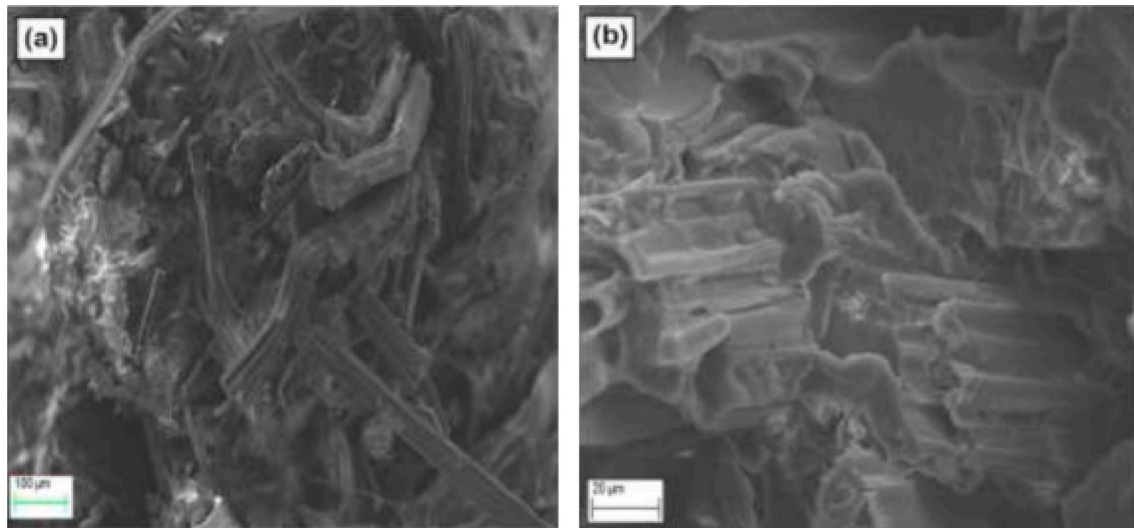


Fig. 11 SEM images of tensile fractured surfaces of **a** untreated PLA/jute composites and **b** silane 2 treated PLA/jute composites (reproduced with permission from Elsevier, license number: 5046471264297) (Bajpai et al. 2013)

out to categorize the mechanical qualities of the mechanical interlock between the fibers and a binder. An ANSYS Software model and analysis of a Polylactic acid drop-off laminate was also conducted in order to comprehend the behavior of FRPs under axial load.

Khan and coworkers (2016) examined the mechanical characteristics of reinforced Poly lactic acid composites for woven jute textiles, proposing them as a sustainable substitute to non-recyclable synthetic fiber. Hot pressed molding have been utilized to make the Polylactic acid reinforced plain WJF (woven jute fabric). The mean values for tensile modulus, tensile strength, flexural modulus and flexural strength, of the raw warp-directed woven jute composite expanded by roughly 211, 103, 42.4, 95.2, and 85.9 percent after reinforcement, whereas the strain at highest tensile stress raised by 11.7%. The methods of manufacturing PLA polymer composites shown in Fig. 11.

Several studies have shown the usefulness of polymer composites reinforced with natural fibers. Ogin et al. (2016) conducted fascinating research on the elements, construction, as well as generic mortification of composite materials, documenting the fundamental constituents of composites as well as the generic flaws originating from the production procedure and exterior stress of the material. Resin shrinkage is a common cause of process-related defects such as shrinkage cracking, porosity, and fiber matrix debonding.

Description and usage of natural fiber qualities for composites were explored in one research by Nechwatal et al. (2003). The tensile strength and Young's modulus of composites have increased as a result of the reinforcing with natural fiber. Additionally, a brand-new method for

manufacturing thermoplastic granules with long fiber reinforcement utilizing standard plastic machinery was also put forward. The single fiber and fiber bundle tests were designed to assess a thread-like structure.

The significance of bio fibers in the PLA matrix

A new direction in the field of polymer composites has been explored by the development of a biodegradable polymer-based bio composite reinforced with bio fibers. The kind of fiber, percentage of fiber content, interfacial adhesion between matrix and fiber, surface fiber modification, and inclusion of additives like compatibilizer, binding agent, and nanofiller, among others, all influence the properties of bio composite materials. PLA composites reinforced with biofibers have garnered a lot of attention lately. Developing a bio composite, or entirely biodegradable material, is practically achievable by combining a biodegradable matrix with biofiber reinforcement. There are a number of advantages associated with PLA composites, including high specific resilience, composability, robust processability, high durability, renewability, and recyclability. The mechanical characteristics of completely natural materials have been enhanced by the bio fibers used in PLA composites (Jawaid and Abdul Khalil 2011; A review on natural fibers for development of eco-friendly bio-composite: characteristics et al. 2021).

PLA-based green composites

Bio composites that use renewable resources for both the matrix and reinforcement are known as green composites.

The necessary composite stiffness and strength characteristics serve as a guidance when choosing an appropriate fiber from the natural fiber pool for a particular polymer. The mechanical properties of the composite are anticipated to be dependent on adhesion at the fiber-matrix interface, fiber aspect ratio, volume percentage, and orientation in addition to the intrinsic qualities of each component (fibers and matrix) (Khan et al. 2016). Nature has offered a vast choice of safe, healthy, sustainable, and, most importantly, environmentally friendly supplies. Right now, it is necessary to identify these resources and develop methods for making the best use of them, such in the example of green composites. The majority of composites are "green" and have industrial use. Green composites are eco-friendly and could be a viable alternative to petroleum-derived polymers and polymer composites. It is feasible to reduce the use of fossil fuel resources by developing biopolymer matrices that use renewable resources such as vegetable oils, carbohydrates, and proteins, such as those found in cellulose-reinforced green composites. Vegetable oils, which can be utilized to manufacture sustainable polymers, are cheap and readily available. These have increased the potential for sustainable and "biodegradable" composites, which can be referred to as "green" composites since they satisfy the requirements for "green materials," and they have also increased the usage of plant fibers as reinforcements.

Therefore, in order to produce "green" composites, "green" polymers that serve as a matrix must be obtained (.K et al. 2022). Various types of natural fibers are utilized in green composites, shown in Fig. 12.

"Green composites," made from natural fiber composites reinforced with PLA, have promising mechanical qualities when compared to non-renewable petroleum-based materials. Fiber addition can also improve PLA's barrier qualities, impact strength, and heat deflection temperature.

The majority of green composites are made using methods that are essentially the same as those used to create conventional synthetic FRP matrix composites, which are categorized as either open mold or closed mold. Open mold processes include filament winding, hand layup, spray up, tape layup, and autoclave technique. Closed mold procedures include transfer molding, injection molding, and compression molding. The three most often utilized processing methods for composites are extrusion, compression, and injection molding. Low melting point materials are used in green composites, which means that processing-related deterioration is a possibility. In a similar vein, processing becomes challenging when hydrophilic components are present (Nechwatal et al. 2003). The many processing techniques utilized for developing green composites are depicted in Fig. 13 and properties of green composite show in Fig. 14.

Rajat Rathore and his research team used injection molding to examine the tensile and flexural characteristics of sisal fiber/poly(lactic acid) and jute fiber/poly(lactic acid)-based green composites (Jadhav et al. 2019). Using the hot melt mixing process, Tuan Anh Nguyen and Thi Huong

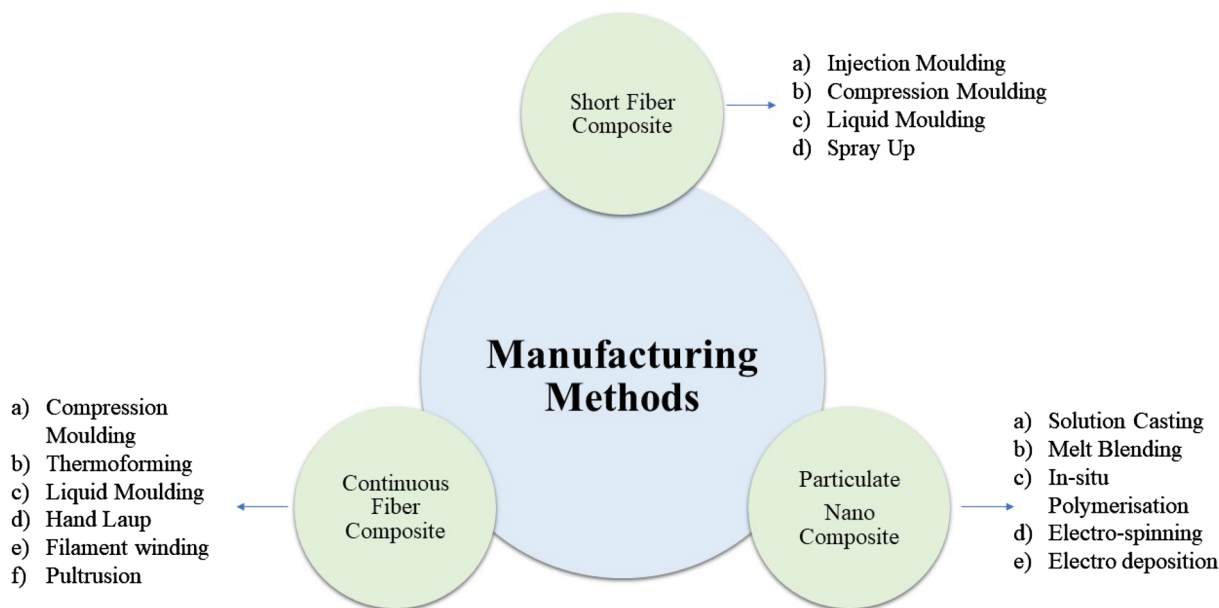


Fig. 12 The method of manufacturing PLA polymer composites from (Qin et al. 2011)

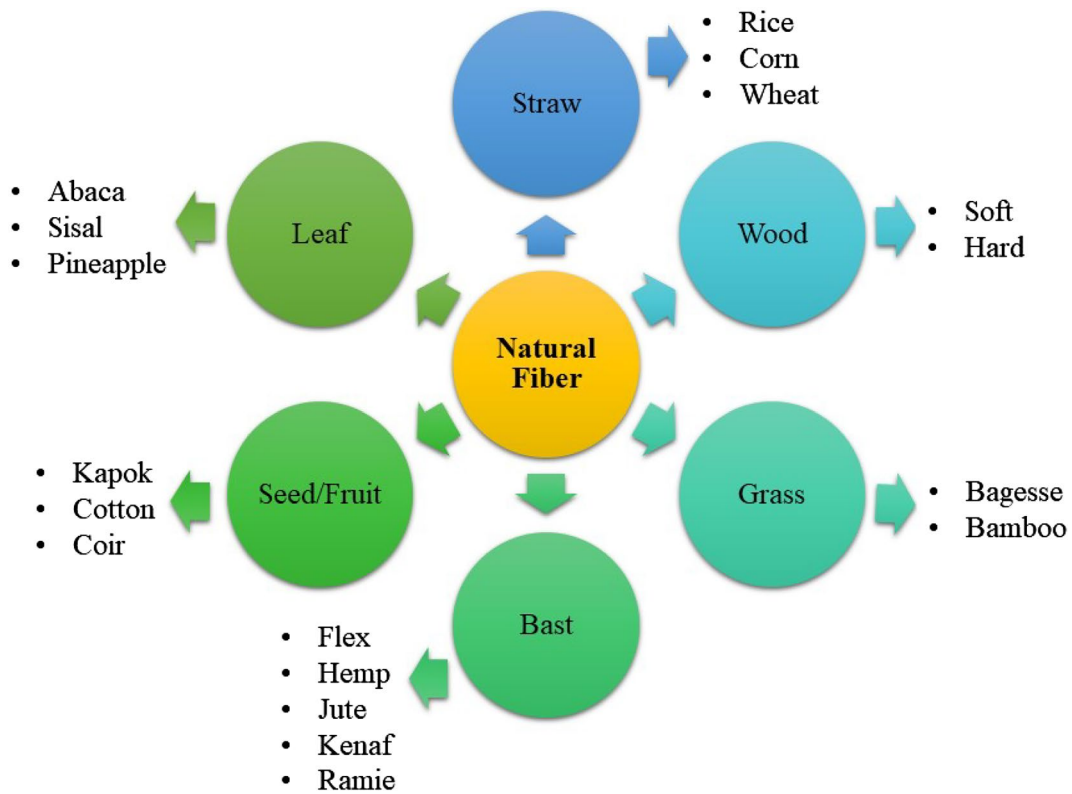


Fig.13 Various types of natural fibers are utilized in green composites from (Nirmal Kumar et al. 2022)



Fig.14 Various processing methods for making green composites from (Nechwatal et al. 2003)

Nguyen created a green composite based on banana fiber and polylactic acid, then examined its mechanical properties. The results obtained indicated that using 20% of banana fiber by weight produced satisfactory outcomes and maintained the mechanical strength values (tensile strength: 52.57 MPa, flexural strength: 70.35 MPa, impact strength: 155.45 J/m, and hardness: 23.8 Hv) at the specified level. SEM observations provided visible proof that the NaOH treatment eliminated surface contaminants from the fiber (Mhatre et al. 2019). Yu Dong and colleagues synthesized a green composite material including polylactic acid and coir fiber, and analyzed its mechanical performance and multifunctional characteristics. With and without alkali treatment, the mechanical, thermal, and biodegradability characteristics of PLA/coir fiber green composite have been effectively assessed (Naik et al. 2022). The impact of four distinct enzymatic treatments on the mechanical characteristics of the coir fiber-reinforced PLA composites was assessed by Kubra Coskun and her research team while synthesizing coir fiber/poly(lactic acid) bio composites. Tensile, flexural, impact, DMA, and SEM studies were performed to characterize the composites (EFFECT OF INJECTION PARAMETERS ON TENSILE AND FLEXURAL PROPERTIES OF GREEN COMPOSITES. 2022). The properties of the

Fig.15 Properties of green composites from Nechwatal et al. (2003)

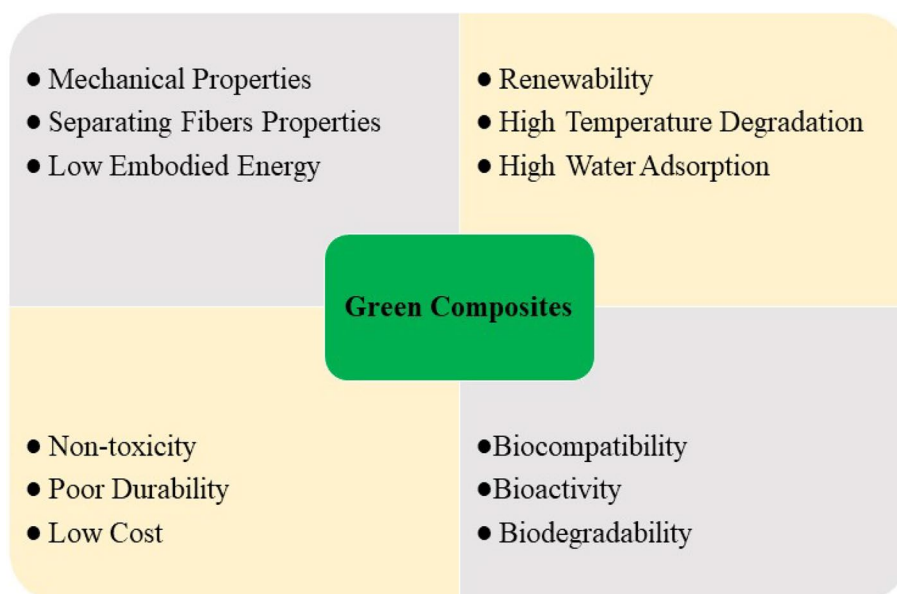


Table 10 Natural fiber properties that could be employed in the fabrication of green composites data obtained from Zini and Mariastella (2011)

Fibers	Diameter (mm)	Density (g/cm ³)	Tensile strength (Mpa)	Youngs modulus (Gpa)	Elongation (%)	Price (\$/Kilo)
Hemp	25–250	1.47	550–900	38–70	1.6–4	1.55
Sisal	50–200	1.45	468–700	9.4–22	3–7	0.65
Abaca	10–30	1.5	430–813	31.1–33.6	2.9	0.345
Bamboo	25–40	0.6–1.1	140–800	11–32	2.5–3.7	0.5
Flax	40–600	1.5	345–1500	27–39	2.7–3.2	3.11
Curaua	7–10	1.4	500–1100	11.8–30	3.7–4.3	0.45
Kenaf	2.6–4	1.5–1.6	350–930	40–53	1.6	0.378
Jute	25–250	1.3–1.49	398–800	13–26.5	1.16–1.5	0.925
Ramie	0.049	1.5–1.6	400–938	61.4–128	1.2–3.8	2

Kenaf Fiber/PLA and Kenaf Fiber/PP Composites, which were developed by Seong Ok Han and their research team utilizing melt compounding and injection molding, were compared to those of kenaf-reinforced polypropylene (PP) composites. The PLA and PP composites have a modulus of 2.96 GPa and 6.64 GPa, respectively, at 40 wt%. The addition of 40 wt% kenaf fibers to PP improved its flexural modulus, flexural strength, and storage modulus at room temperature by approximately 260%, 50%, and 134%, respectively, whereas PLA's characteristics were enhanced correspondingly by 118%, 4%, and 64% (Nguyen and Thi. 2022). Continuous pineapple leaf fiber-reinforced PLA composite was developed by Jaya Suteja and her research team for the purpose of examining the properties of 3D printed (Dong et al. 2014b). PLA/Wood fiber/MAH-g-PLA composites were developed by Lei Zhang, Shanshan Lv, and their research team using melt blending and injection molding with various compatibilizer

addition ratios. PLA was used for the matrix phase and wood fiber for the reinforcing phase. Using techniques such as thermogravimetric analysis (TGA), scanning electron microscopy (SEM), X-ray diffraction (XRD), and dynamic mechanical thermal analysis (DMA), the crystallinity, microstructure, thermal stability, and dynamic thermomechanical property of the composites were examined. The mechanical parameters depicted in Fig. 15 include tensile strength, elongation at break, bending strength, and modulus for PLA/Wood fiber/MAH-g-PLA composites (Coskun et al. 2019) (Table 10).

The effects of kenaf fiber-reinforced composites made at 20%, 30%, 40%, and 50% fiber concentrations were examined by El-Shekeil et al. Thirty percent fiber content adds hardness but reduces heat stability and abrasion resistance (Seong et al. 2012). Jianghu Zhan and his research group developed composites based on poly(lactic acid) and ramie fiber to examine the effects of chemical treatments

Table 11 Some examples of PLA-based green composites utilizing diverse natural fibers from distinct processing methods

Natural fibers	Methods	References
Sisal	Injection molding	El-Shekeil et al. (2012)
Silk fiber	Internal melt mixer and compression molding	Jianghu Zhan (2021)
Coffee ground and bamboo flour	Injection molding	Gorrepotu et al. (2023)
Aloe Vera Fiber	Injection molding	Saurabh Chaitanya and Inderdeep Singh (2017)
Chicken feather fiber	Twin screw extruder and Injection molding	Buasri et al. (2013)
Wheat straw	Twin screw extruder and injection molding	Baek et al. (2013)
Banana fiber	Melt blending	Chaitanya and Singh (2018)
Flax fiber	Compression molding	Cheng et al. (2009)
Ramie	Melt extrusion and solvent casting	Nyambo et al. (2011)
Kenaf	Compression molding	Shih and Huang (2011)
Jute	Compression molding	Suneel Motru et al. (2020)
Pineapple leaf fibers	Melt blending	Sharma et al. (2021a)
Bamboo fiber	Hot pressing	Manral and Pramendra (2019)
Kenaf fiber	Melt blending	Singh et al. (2019)
Kenaf fiber	Melt blending	Farahiyan et al. (2015)
Kenaf fiber	Single screw extrusion	Takagi et al. (2012)
Kenaf fiber	Single screw extrusion	Chen et al. (2017)
Kenaf Fiber	Extrusion and compression molding	Pan et al. (2007)
Banana Fiber	Injection molding and compression molding	Fundamental study and modification of Kenaf fiber-reinforced polylactic acid bio-composite for 3D printing filaments (2023)
Bamboo fiber	Melt blending	Lau et al. (2023)
Bamboo fiber	Compression molding	Hassan et al. (2019)
Bamboo fiber	Compression molding	Komal et al. (2020)
Sugarcane bagasse fiber	Compression molding	Shih and Lai (2020)
Untreated sisal and coir fibers	Compression molding	Roy Choudhury and Debnath (2020)
pineapple leaf and coir fibers	Hot pressing	Choudhury et al. (2022)
Hemp	Injection Molding	Khoo and Chow (2017)
Pineapple leaf	Twin screw extruder	Duan et al. (2017)
Pineapple leaf	pre-pregging	Siakeng et al. (2019)
Pineapple leaf and chicken feather fiber	Internal plasticizer and hot-pressing	Elen et al. (2023)
Chicken feather fiber	twin-screw extruder and injection-molded	Todhanakasem et al. (2022)
Hemp and jute fiber	Compression molding	Mustafa et al. (2022)
Pineapple leaf fiber	Twin screw extruder	Enthil et al. (2023)
Coir and pineapple leaf fiber	Hot pressing	Akderya et al. (2020)
Pineapple leaf fiber	Compression molding	Arockiasamy et al. (2022)
Ramie	Autoclave foaming and molding	Kaewpirom and Worrarat (2014)
Ramie	Injection Molding	Siakeng et al. (2018b)
Ramie	hot-pressing	Agung et al. (2018)
Banana/sisal fiber	Twin screw extrusion	Li et al. (2023)
Sisal	Hot press molding	Li et al. (2022)
Sisal	Compression molding	Soemardi et al. (2023)
Wood	Hot press molding	Asaithambi et al. (2014b)
Wood	Hot-pressing foaming	Jayamani et al. (2015)
Wheat	Injection molding	Wang et al. (2019)
Wheat and bamboo	Melting–blending and injection molding	Guo et al. (2013)
Banana	Compression molding	Wang et al. (2022)
Banana	Compression Molding	Chai et al. (2023)
Wheat	Hot pressing	Zhang et al. (2020)
Bamboo	Hot pressing and cold pressing	Ramasamy et al. (2022)

Table 11 (continued)

Natural fibers	Methods	References
Bamboo	Twin-screw extruder	Weerasinghe et al. (2023)
Bamboo	Co-rotating twin-screw extruder and hot and cold press	Chougan et al. (2022)

and Compatibilizers. The PLA/RF/TGIC composites' tensile and flexural strengths increased by 49.8% and 46.5%, respectively, in comparison to pure PLA (Suteja et al. 2022). Using an injection molding technique, Surya Rao Gorreputu at El. developed composites made of polylactic acid green and pineapple leaf fiber. The research examined the impact of several parameters such as pressure, speed, temperature, fiber length, and fiber loading on the characteristics of PLF/PLA green composite (Zhang et al. 2017b). Some examples of PLA-based green composites utilizing diverse natural fibers from distinct processing methods show in Table 11.

Green nanocomposites based on PLA

Nanocomposites are gaining traction in the global plastics processing industry, and their enormous potential for a wide range of product development is well known. The upcoming trend in high-efficiency, low-cost, lightweight, sustainable, and environmentally friendly nanocomposites is anticipated to be green or eco-friendly materials. To create sustainable products, a variety of manufactured and natural green polymers and green nanofillers have been used. The unique characteristics of sustainable polymers and eco-reinforcement are embodied in green polymeric nanocomposites. Applications for the eco-friendly nanofiller reinforced green nanocomposites can be found in the packaging, energy, automotive, and biological

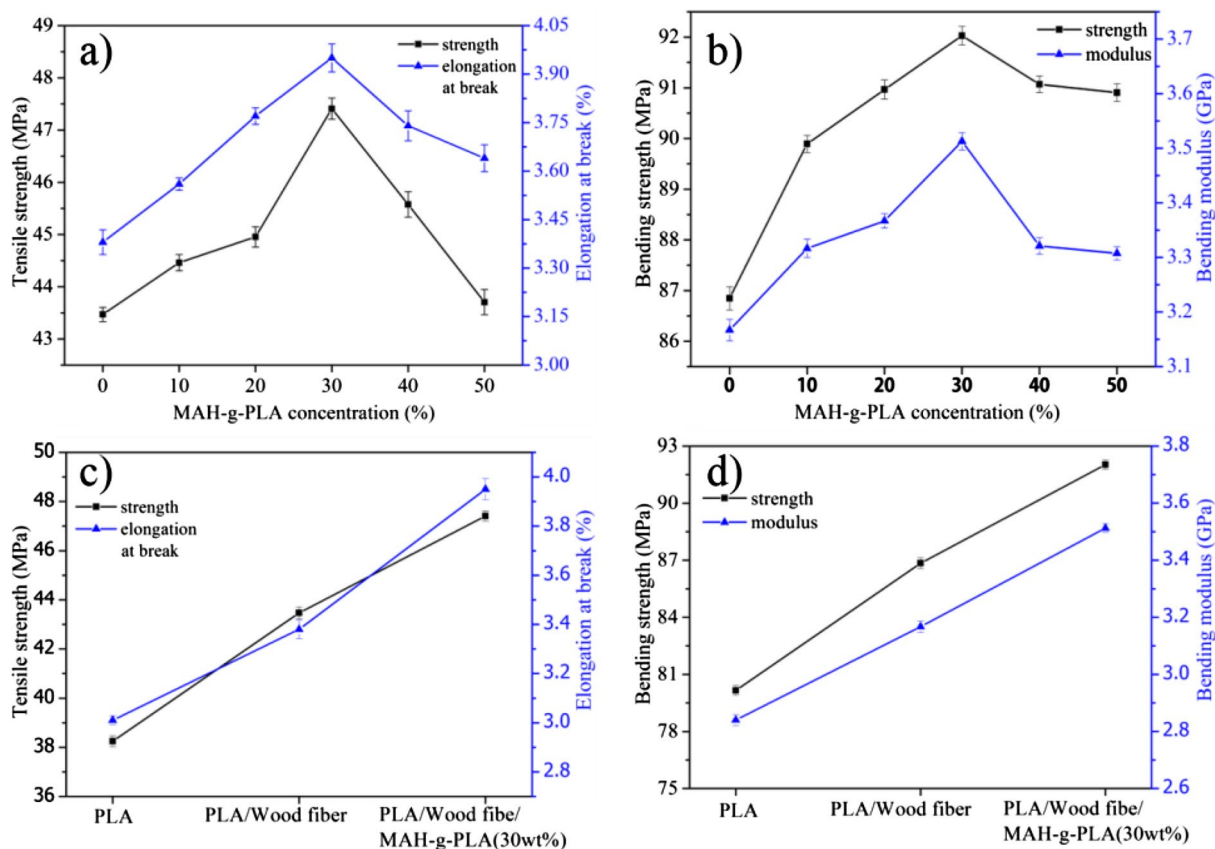
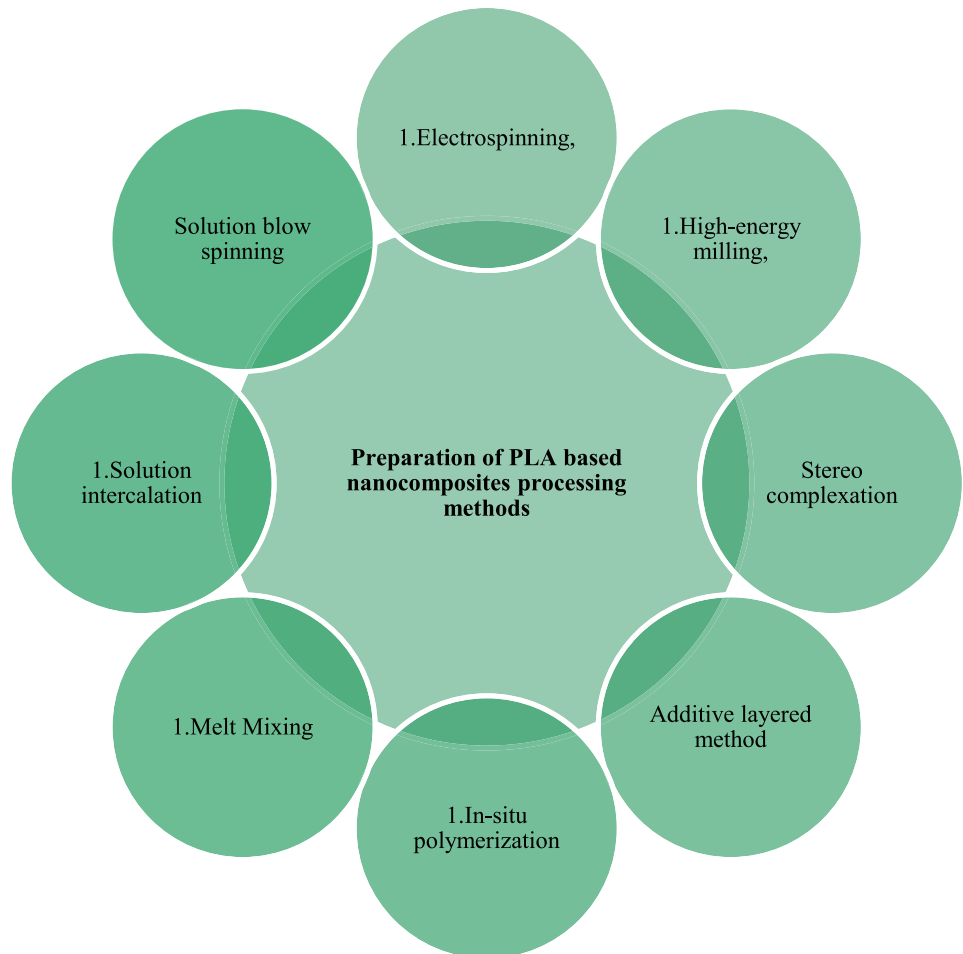


Fig.16 Mechanical properties of PLA/Wood fiber/MAH-g-PLA composites. **a, c** Tensile strength and elongation at break, **b, d** Bending strength and modulus from (Coskun et al. 2019)

Fig. 17 Various processing methods for preparation of PLA-based nanocomposites from Lin et al. (2018), Kausar (2021)



sectors. Green composites or nanocomposites developed entirely of renewable resources offer lower prices and great ecological balance as compared to standard non-green materials. The success of green materials is determined by a variety of factors, including processing methods, ease of fabrication, and desirable physical properties (Yang et al. 2015; Chen et al. 2020; Lin et al. 2018). When designing a high-performance PLA nanocomposite, the optimal combinations of nanofillers, preparation methods, and processing parameters must be carefully taken into consideration. In addition to multi-walled carbon nanotubes (CNT) and montmorillonite (MMT), which are well-known as nucleating agents to achieve high thermal and mechanical properties, other nanofillers, like silver nanoparticles (AgNP), are important in enhancing the antibacterial and high-performance properties of PLA. The properties of the polymer nanocomposite are determined by the processing technique employed, which in turn determines the dispersion state of the nano-reinforcements throughout the matrix. (Kausar 2021). PLA nanocomposites are made up of nano-fillers such as aluminum hydroxide, hydroxyapatite, layered titanate, carbon nanotubes (CNT),

and layered silicates. In PLA nanocomposites, the nanofiller is at least one dimension's nanoscale, or less than 100 nm (Hassan et al. 2012; Ates et al. 2020; Sanusi et al. 2020). Various processing methods for preparation of PLA-based nanocomposites and PLA-based nanocomposites shown in Figs. 16 and 17, respectively.

Green nanocomposites based on poly(lactic acid)/banana fiber/nanoclay were developed by VP Sajna and his research team using a melt blending approach and injection molding. Within the poly(lactic acid) matrix, untreated and chemically changed banana fibers as well as organically modified nanoclay were utilized as reinforcing agents. The mechanical properties, dynamic mechanical analysis, differential scanning calorimetry, thermogravimetric analysis, heat deflection temperature (HDT), morphological properties using scanning electron microscopy and transmission electron microscopy, and water-absorption studies were used to assess the effects of chemically altering banana fiber and incorporating nanoclay in the biocomposites and bionanocomposites. The inclusion of 3% nanoclay and fiber treated with slane produced enhanced strength and modulus because the nanoclay established hydrogen bonds

at the interfaces between the fiber and matrix, according to tensile and flexural evaluation of biocomposite and bionanocomposites (Sharif and Sudipta Hoque 2019).

Procedures to increase the ductility of PLA

Because PLA is an intrinsically brittle polymer, it needs to have better ductility for a larger range of applications. There are two categories of techniques for doing this. The first category consists of physico-chemical techniques such as plasticizers, impact modifiers, mixing, or copolymerization to increase the ductility of PLA. However, PLA may be mechanically modified using methods like stretching (uni- or bi-axial), for example, to make it more ductile. This section discusses each of these options.

Plasticizing

All plasticizers typically have a linear or cyclic carbon chain and are carbon-based chemicals. A plasticizer's lower molecular size than the base polymer's enables it to fill the intermolecular gaps between the base polymer chains, decreasing the secondary forces between them and increasing the mobility of the molecular chains as a result.

Plasticizer usage has two outcomes. First off, PLA is a polymer with great stiffness and strength but poor impact resistance. When subjected to dynamic influences, it acts as a hard polymer. In this situation, plasticizers are employed to toughen the polymer and change how it behaves, boosting its impact strength and elongation at break. This can only be done, however, by sacrificing some rigidity and strength. Additionally, at greater plasticizer contents (> 10 wt%), elongation at break typically only rises. Second, by diffusing into the polymer chains and lowering the glass transition temperature, plasticizers may be employed to increase the molecular chain mobility of the supplied polymer (T_g). In this instance, a plasticizer is used to enhance

the base polymer's overall crystallization. The wider crystallization window between T_g, which is lowered by the plasticizer, and the melting temperature also helps crystallization (T_m). Although the plasticizer lowers T_m as well, the temperature differential between T_g and T_m often widens (Raquez et al. 2013; Basu et al. 2017). Both justifications for employing a plasticizer are necessary for PLA. 10–20 wt% of plasticizer is often required for both a significant drop in the T_g of PLA and acceptable mechanical characteristics, however adding more plasticizer to the polymer than 20–30 wt% typically causes phase separation (depending on the Mw of the plasticizer). Thus, the quantity of plasticizer to be blended with PLA will determine how much plasticization is possible. Additionally, the quantity of plasticizer employed, Mw, and miscibility with the host polymer all affect how well plasticizers are utilized (Table 12).

In addition to being renewable resource-based, non-volatile, biodegradable, biocompatible (for medical applications), and non-toxic (for food packaging), an appropriate plasticizer for PLA should lower the T_g and boost elongation at break with little leaching or migration during storage. Injection molded components made from plasticized PLA are inappropriate for use in food contact applications because a poorly chosen plasticizer may migrate out of the base polymer and cause difficulties with product safety. Finding out what kinds of plasticizers have been used for PLA, how successful they have been, and most crucially, which ones may be used in applications where migration is prohibited, was one of the objectives of the literature study (Sajna et al. 2014; Saeidlou et al. 2012 Dec 1; Auras et al. 2011).

Impact modification

Natural rubber

Addition of another ductile renewable resource-based material is one of the best strategies to improve the impact

Table 12 Various application of green composites from (Faruk et al. 2012)

Sector	Applications
Short life product	Packaging applications, Rope, bag, broom
Electronics industries	Poly(lactic acid)/ kenaf dummy cards in personal computer, printed circuit board, Mobile phone, Radio
Sport industries	Toys, Flax reinforced snowboard, Tennis rackets, Bicycle frames
Biomedical industries	Articular cartilage, Trachea, Sutures, Drug delivery system and grafts, Scaffolds for tissue engineering and bone fixators
Structural application	Concrete elements, fencing, decking, siding, bridge, fiber cement, Composites soil
Household components	Ceiling, floor, window, Wall partition, table chair, kitchen cabinet, decks, suitcase, Roofing
Automobile industries	Door trim panels, Spare tire cover, Seat backs, dashboard, headliner, luggage compartment, Engine and transmission enclosure for sound insulation

strength or ductility of PLA (Liu and Zhang 2011 Aug 1). Isoprene, the primary ingredient in both isoprene rubber (IR), and natural rubber (NR) is one of these biomaterials (Grigale et al. 2010 Jun). While synthetic rubber (SR) is utilized to replace natural rubber (NR), NR is still the most significant material in the rubber industry and is used to make tires among other things. Latex, which is produced by tapping the "rubber-tree" (*Hevea brasiliensis*), is the sap. Latex is a cis-1,4-Poly(isoprene) aqueous suspension with a poly isoprene ratio of around 30–38 weight percent. We discovered that there are "island-like" NR phases in the PLA matrix and that the adhesion between the two phases in PLA/NR mixes is poor (Arrieta et al. 2014 Dec).

According to Lopez-Manchada and colleagues (Courgneau et al. 2013 Aug), NR produces a dispersed phase that resembles a droplet in PLA, and the processing conditions may affect the droplet diameter. For example, a rise in melt temperature during internal mixing increased the size of the NR droplets, which was a result of the PLA droplets' ability to fuse due to a decrease in viscosity. This internal mixing method resulted in an average NR droplet of 1–2 μm . The adhesion between the phases of the 80/20 wt% PLA/NR mix vulcanized with dicumyl peroxide enhanced when a cross-linking (or curing) agent was also added to the compound. The mechanical characteristics of the mix were also significantly enhanced by the web-like structure the NR phase created in the PLA (Li and Shimizu 2007 Jul 9). Santawitee et al. (Alias and Ismail 2019 Sep 2) looked at the cross-section of PLA/NR and PLA/ epoxidized NR (ENR) blends. Because PLA and ENR are only partially compatible, the scientists discovered that the epoxidation of NR results in improved size distribution (smaller droplets). Another research looked at the morphology of PLA/NR mixes that were 65/35 and 80/20 by weight. The combination with 35% NR, according to the authors, has better mechanical qualities than pure PLA, such as a seven-fold increase in impact strength. This effect is thought to have been brought on by the web-like structure created by the NR. By etching the NR content for 1 and 4 min, this structure was shown (Bitinis et al. 2011 Oct 3).

Mechanical characteristics of PLA bio composites

Poly (lactic acid)-based composites have attracted significant commercial attention in the composites sector among the biopolymers due to their excellent mechanical characteristics and biodegradability (Yuan et al. 2014 Sep). For materials that are entirely natural, the fibers utilized as fillers in PLA composites have increased mechanical characteristics (Pongtanayut et al. 2013 Jan). Fiber volume/weight fraction, fiber layer stacking order, processing techniques, fiber treatment, and environmental effects are all taken into account during mechanical characterization

(Saba et al. 2014). PLA was reinforced with bio-fibers such ramie, hemp, rice straw, kenaf, wood, abaca, sisal, coir, rice husk, bamboo, oil palm, and flax (Xu et al. 2014 Aug; Yussuf et al. 2010 Sep). Some researchers have looked at adding plasticizer, impact additives, and other additives to soft polymers in an effort to increase PLA's impact power (Azwa et al. 2013; Nunna et al. 2012 Jun; Yu et al. 2010 Apr 1). According to the findings, the aforementioned procedures greatly increased PLA's durability at the expense of stiffness. Due to this, there has been a lot of attention in developing compostibilizing graft copolymers in recent years to provide superior PLA and fiber compatibility as well as high impact strength (Fortunati et al. 2013). Bio-composite PLA/ bamboo flour was made using a poly (lactic acid)-graft-glycidyl methacrylate (PLA-g-GMA) graft copolymer that was synthesized (Asim et al. 2017 Jan; Tawakkal et al. 2014 Nov). Since GMA is a bifunctional monomer made up of an epoxy group at one end and an acrylic group at the other, its graft copolymer was employed as an appropriate material in composites. While the acrylic group experiences polymer chain coupling, the epoxy group of the GMA may react with the fiber or -COOH group of the -OH group in PLA, forming a durable chemical link between the PLA matrix and fiber (Wu 2014 Oct).

Thermal characteristics of PLA bio composites

Composites that are heated are likely to see significant changes to their chemical and physical characteristics. The substance will go through processes that depend on temperature and time, including sublimation, evaporation, water absorption, etc. As a result, changes in mechanical, thermal, electrical, and magnetic characteristics will occur (Xu et al. 2012). Because natural fibers degrade with time, the decline in thermal stability in PLA-based green composites may be more pronounced (Haafiz et al. 2013 Oct 15). Natural fibers typically degrade between 200 and 210 °C, which is the temperature at which PLA is processed, but PLA degrades starting at around 300 °C. Therefore, it is essential to research the heat degradation of PLA composites made of cellulose.

3D and 4D Printings of PLA Bio composite

The primary natural raw material for 3D printing is polylactic acid, a thermoplastic aliphatic polyester. It is a thermoplastic polymer that is entirely biodegradable and was created using renewable basic resources (Wang et al. 2014 Jun). One of the most widely used feedstocks for additive manufacturing is PLA, which is one of the materials available for 3D printing. One of the easiest

materials to print, PLA has the advantage of being easy to process, even though it tends to slightly contract after 3D printing (Ray and Cooney 2018). One major advantage of PLA over ABS is that it can be 3D printed at much lower temperatures (between 190 and 230 degrees Celsius) since it does not need a heated printing substrate (Ngaowthong et al. 2019 Sep). PLA also does not need much post-processing since the supports are often extremely simple to remove and the material may be treated with acetone or sanded as needed. Many producers, including Amolen, Prusament, PolyMax, Polymaker MatterHackers, Proto Pasta, Colorfabb, Fillamentum, Paramount 3D and Sunlu, created PLA filament for use with 3D and 4D printing.

This material is utilized as a substitute for petroleum-based plastics for packaging applications, notably in the food sector since it interacts well with foods (Azlin et al. 2020 Jan). FDM (fused deposition modeling) technique, which produces components by the extrusion of thermoplastic filaments, may be used to 3D print using PLA. PLA is one of the frequently used materials for this technology. Composites are incredibly useful for producing lightweight parts with robust mechanical properties. The fundamental criterion for classifying composites as fiber-reinforced materials is the ability of the fibers to provide mechanical strength to pieces without increasing weight (Chia and Wu 2014 Dec 17; Mazzanti et al. 2019 Jun 28). In order to create 3D printed structures that can alter their form or qualities over time (Jamshidian et al. 2010 Sep; Ilyas and Sapuan 2019 Dec 1), four-dimensional printing has become a popular trend. The distinction is that whereas 3D printed products keep a fixed shape like any plastic or metal component, 4D printed objects may change over time. The evolution through time is

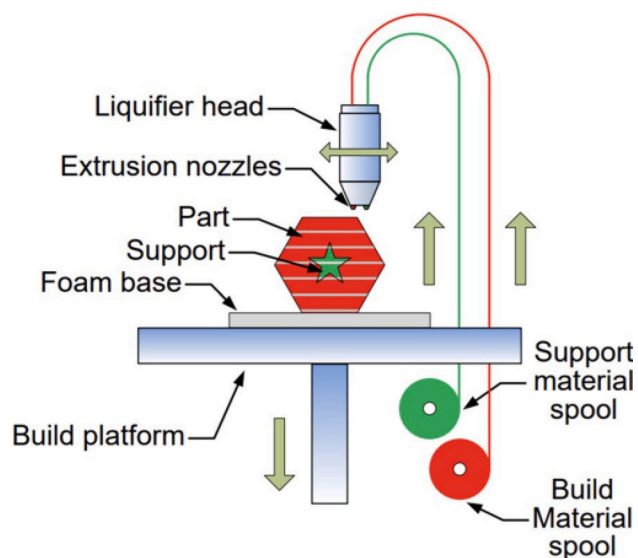
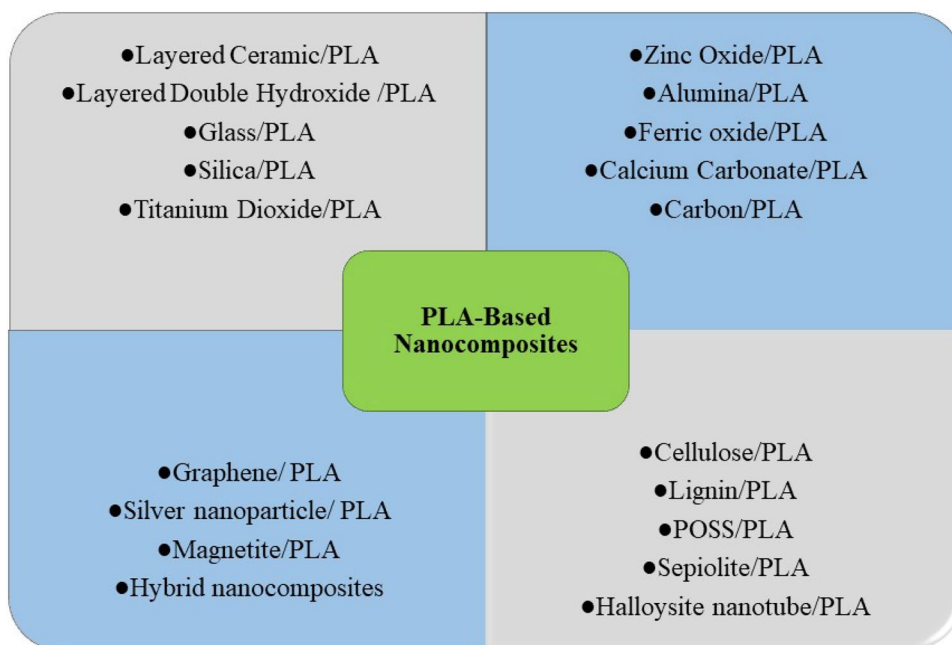


Fig.19 Fused deposition modeling is shown schematically. Elsevier granted permission for this reproduction, license number 5170280890406 (Ouhsti et al. 2018)

the fourth dimension of 4D, where 4D printing technology produces intelligent structures by using cutting-edge production methods like 3D printing, superior materials, and specialized design. A stimulus is required for 4D printed items to enter the deformation phase; examples of possible triggers include exposure to water, heat, light, or magnetic fields (Ilyas and Sapuan 2020 Aug 1; Champeau et al. 2020 Aug; Spiegel et al. 2020 Jun; Zolfagharian et al. 2020 Mar).

Fig.18 Various nanocomposites based on PLA from Hassan et al. (2012), Ates et al. (2020), Sanusi et al. (2020)



Fused deposition modeling (FDM)

FDM, also known as FFF (fused filament fabrication) or material extrusion, is a 3D printing process with a number of benefits, including as affordability, tolerable working conditions, and a high build volume capacity (Quanjin et al. 2020 Jan). Nevertheless, it has drawbacks such poor precision, sluggish printing rates, and shrinking brought on by temperature fluctuations (Ma et al. 2020 Apr). The 3D CAD model is digitally divided into layers as part of the FDM process (Fig. 7), and the data is then transformed into G-code (Geometric code for computer numerical control), which is sent to the 3D printer, which builds the component layer by layer. The thermoplastic filaments, which typically have diameters of 1.75 or 3 mm, are then fed into the heated chamber and melted. The molten material is subsequently extruded in a layer-by-layer arrangement using nozzles. The final item is then taken off the build platform, and the support material may also need to be taken off (Rahim et al. 2019 Oct 2) (Fig. 18).

Several research (Jadhav and Wankhade 2017 Sep; Hart 2019) have examined the impact of FDM printing settings on the functionality of 3D printed objects. The build orientation, raster angle, layer thickness, infill density, and extrusion temperature are among the factors that influence the quality of an FDM produced item (Fig. 19).

One of the structural factors that has the greatest impact on the mechanical characteristics of FDM components is build orientation. FDM items produced in a horizontal or vertical orientation have much worse mechanical characteristics than parts printed perpendicular to the build platform (Liu et al. 2019 Jun 19; Popescu et al. 2018 Aug). The mechanical qualities of FDM printed samples are also influenced by raster angle. The greatest tensile characteristics are found in samples printed with a raster angle of 0°, followed by 0°/90°, 45°/45°, and 90°, according

to studies (Domingo-Espin et al. 2015 Oct; Smith and Dean 2013 Dec 1). The quality and mechanical qualities of FDM printed products are also greatly influenced by layer thickness and infill % (Fig. 20).

Depending on the material used, FDM can print at its fastest possible rate. According to research on the effects of printing speed on FDM components, interlayer bonding between the filaments decreases as printing speed rises, which is explained by the insufficient time for polymer plasticization (Wu et al. 2017 Aug 19; Dizon et al. 2018; Ouhsti et al. 2018).

Application of PLA bio composites

The creation of biopolymers on a broad scale has been made possible by the increase in the price of oil, the depletion of oil reserves, the global awareness of sustainable development, and the implementation of various laws like the End of Vehicle Directive. In light of this, PLA has distinguished itself as a biopolymer with high potential to replace synthetic polymers. A range of uses are possible for PLA thanks to its controllable characteristics (Ning et al. 2015; Duigou et al. 2019 Oct).

As a plastic that is thought to be environmentally benign, PLA is a good choice for agricultural items such bags for storing agricultural products, mulch films, paper coating for packaging, and bags for fertilizers, composting, and pesticide release systems (Kariz et al. 2018 Mar; Subash and Kandasubramanian 2020 Jul). The potential for PLA in textile production is enormous. It may be melt-spun, pulled to remove crystallization stress, extruded, or molded to create fiber. In addition to being used as a fiber fill in quilts and pillows, PLA-based fibers are also used in carpet as continuous filament, as filament and spun yarns in clothing, and as a variety of biocompatible and nonwoven

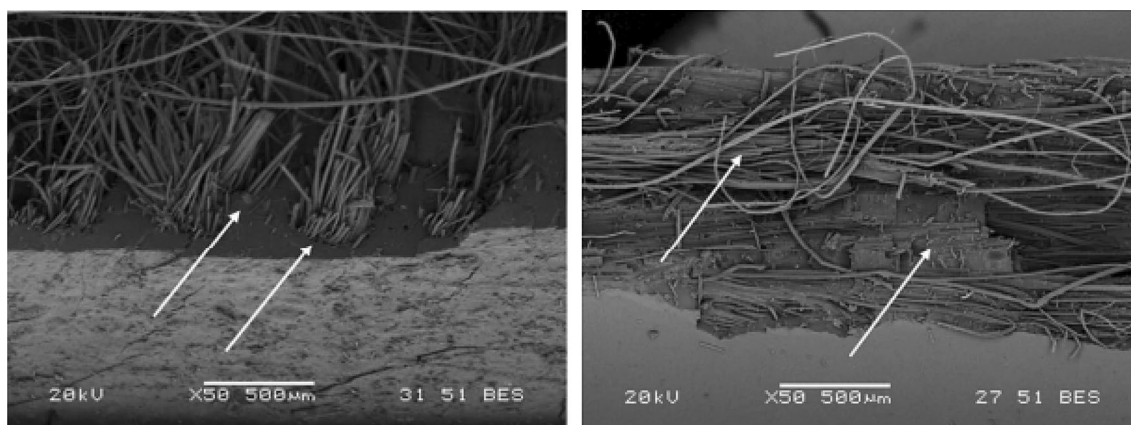
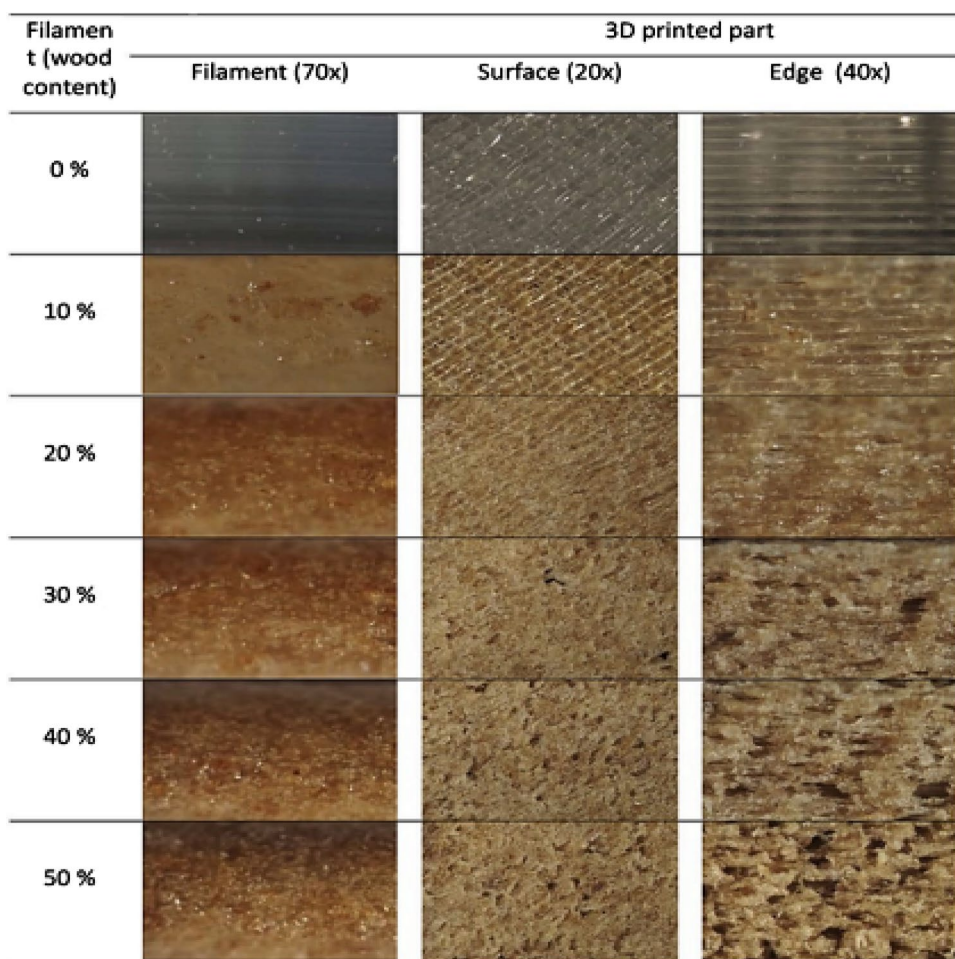


Fig. 20 SEM pictures of the fracture surfaces of PLA/flax yarn composites manufactured using FDM **a** longitudinal fracture and **b** transverse fracture (Yang and Yeh 2020)

Fig.21 Optical micrographs of a wood/PLA filament, surface, and edge (Miazio 2019) are shown. Elsevier granted permission for this reproduction, license number 5167000723403



fibers like binders and self-crimp (Ahmed et al. 2018b). PLA is used in cushioning, reusable clothing, canopies, and diapers, especially in nonwoven form. Typically, fibers must have great heat resistance and stiffness because to the thermal and mechanical stresses they must withstand. Given that high crystalline fiber can function under stressful circumstances, meso lactic compound (8 to 20percent) is added to Poly (lactic acid) to create cross-linking chains that will strengthen the fiber's bond with other fibers (Nagarjun et al. 2022 Dec 2; Asyraf et al. 2021 Dec 31; Ali et al. 2021 Apr 15). Today, PLA is widely used to construct items like single-use cups of tea and coffee, as well as packaging for mustard oil as well as cosmetics. Additionally, PLA's low smoke production after burning, excellent UV resistance, and effective wicking properties make it a useful substance for the industries of textile and dye (Asyraf et al. 2022 Feb; Sharma et al. 2021b) (Fig. 21).

Conclusion and future scope

Natural fibers, an abundant and sustainable supply of raw materials for eco-friendly manufacturing, have been crucial to the development of human civilization. PLA-based bio composites/green composites have grown in popularity as people become more aware of the benefits of using environmentally friendly materials in various applications. PLA/biofiber based composites are a good substitute for current non-biodegradable petroleum-based products since they have sufficient mechanical and thermal properties. Applications for PLA-based bio composites include short-lived products, the electronics, sports, biomedical, and structural industries as well as household and automotive components. Bio-composites are important for many applications, and the underlying matrix's characteristics may be improved by adding other elements. Diverse renewable and non-renewable elements are used to create bio-composites. PLA is a special natural thermo-plastic polymer that is one of the most environmentally friendly and has superior qualities among the renewable polymers. Creating composites based on PLA is a key strategy for

resolving issues with PLA scaffolds. The use of composite methods considerably increases the variety of biomaterials' components and possible applications. Combining PLA with other materials may result in products with balanced physical and biological properties. The capacity of mixes of Poly (lactic acid) and copolymers to function as parallel polymers in a variety of packaging and fiber sectors is considerable. With the advent of research and development in biobased goods, PLA processing has become simple. Extensive research studies have been conducted to better understand the behavior and properties of PLA and bio-fiber based PLA composites under various processing environments. Current and future research should focus on the development and enhancement of PLA-based composites with various types, ratios, and forms of natural fibers for multifunctional applications. PLA/Bio fiber-based composites are utilized in a wide range of applications, but further research and development is required to lower processing costs, improve performance, and increase utilization in industrial applications. PLA and its bio composites are the most commonly utilized materials in 3D printing worldwide. PLA/biofibers-based bio composites with form memory and other advantageous features could be extremely valuable for 4D printing.

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Author contribution Who voluntarily contributed to this study.

Declarations

Conflict of interest The author declares no conflict of interest.

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