

REVIEW ARTICLE OPEN



Recent advances in biodegradable polymers for sustainable applications

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The interest in producing biodegradable polymers by chemical treatment, microorganisms and enzymes has increased to make it easier to dispose after the end of its use without harming the environment. Biodegradable polymers reported a set of issues on their way to becoming effective materials. In this article, biodegradable polymers, treatment, composites, blending and modeling are studied. Environmental fate and assessment of biodegradable polymers are discussed in detail. The forensic engineering of biodegradable polymers and understanding of the relationships between their structure, properties, and behavior before, during, and after practical applications are investigated.

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INTRODUCTION

Disposal of plastic wastes is a serious environmental problem that we face today. Mass production and increased use of plastics in wide applications in our daily life^{1,2} have resulted in environmental impact. Consequently, these issues lead to the growing threat of global warming resulting from carbon dioxide emission due to burning of non-biodegradable conventional polymers such as polyethylene, polypropylene, and polyvinylchloride³. Biodegradable polymers are being developed to be used as an alternative for non-biodegradable polymer materials in a variety of applications⁴. The best option for managing non-biodegradable plastic waste is to replace the use of uneconomical non-biodegradable materials for recycling or reuse with biodegradable polymers as they are environmentally friendly⁵. Because of the environmental pollution resulting from the use of non-biodegradable materials, studies and developments have increased about biodegradable materials⁶.

Biodegradable polymers are materials that can work for a limited time before degrading into readily discarded products through a regulated procedure⁷. They might be made from a variety of wastes or/and bioresources, such as wastes of food, animal, agro-waste as well as other sources such as starch, and cellulose. Bioplastics made from renewable resources are often less expensive than those made from microbial resources prompting producers to concentrate on making bioplastics from renewable resources⁸. The use of biodegradable polymers has environmental benefits such as regeneration of raw materials, biodegradation and reduction of carbon dioxide emissions that are led to global warming⁹. Microorganisms such as bacteria and fungus may consume biodegradable polymers and convert them to H₂O, CO₂, and methane. The biodegradation process depends on the material's composition¹⁰. The polymer morphology, polymer structure, chemical and radiation treatments, and polymer molecular weight are all parameters that influence the biodegradation process¹¹. Biodegradable polymers are also called biopolymers¹². There are two reasons to use the polymers from renewable resources; (i) environmental concerns in terms of increasing plastic waste and global warming as a result of

releasing of carbon dioxide when burning waste, and (ii) petroleum resources are limited and ended¹¹.

The biodegradable plastics industry is highly promising. However, they need to be developed in tandem through examination of end-of-life processes of treatment and a world-wide integration with organic management of waste as selective biowaste collection expands. Biodegradable plastics have the benefit of being able to be handled biologically at the end of their lives (composting or anaerobic digestion). Composting of biodegradable plastics is thoroughly documented and recognized on a global scale. Product's biodegradability can be conducted under home or industrial composting conditions. In the absence of oxygen, anaerobic digestion transforms organic waste into biogas (a mixture of CO₂ and CH₄), which may then be additionally valorized through heat and power generation¹³. Developing alternative and targeted waste management solutions are also a major challenge to encourage the integration of biopolymers into the circular economy¹⁴. Biodegradable polymers may be categorized in step with their origin¹¹. To manage non-degradable waste and reduce its accumulation in the environment, biodegradable materials have been replaced nonbiodegradable materials in several uses such as plastic bags and packaging. In the field of human health care, biodegradable polymers are also used in various vital applications like soft tissue engineering, gene therapy and soft tissue engineering¹⁵.

The required properties and performance can be provided at a reasonable cost by mixing the matrix of polymers with natural fibers to form natural fiber-reinforced composites (NFRCS). Natural fibers can provide many environmental and economic advantages including being renewable and accessible in large quantities and reduce the cost of raw materials. They can be recycled, biodegradable, achieve a large stiffness to weight ratio and high thermal insulation. Natural fibers are as inexpensive as synthetic fibers such as carbon and glass fibers. Producing 1 tonne of continuous filament glass fiber products from raw materials for factory export emits between 1.4 and 2.2 tonnes CO₂-eq, with an average of 1.8 tonnes CO₂-eq. Compared to natural fibers which emit between 0.3 and 0.7 tonnes of CO₂-eq per tonne of natural fiber¹⁶. Natural fibers may be categorized in step with their origin

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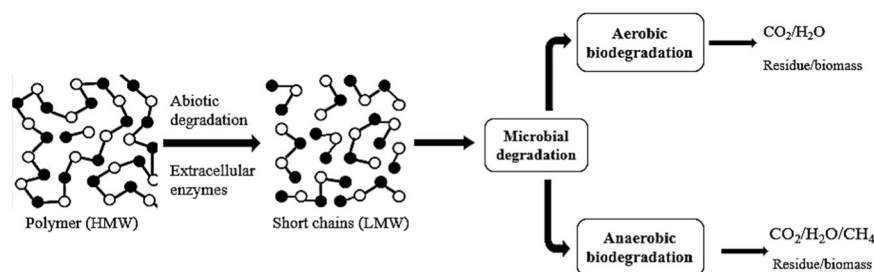


Fig. 1 The chemical biodegradation diagram. Biodegradation steps of polymers.

into plant fibers such as flax and cotton. Animal fibers can be extracted from wool and hair. Mineral fibers consist of silicates such as asbestos, inosilicates, and clay^{17,18}. Although mineral fibers are obtained from nature, they are non-renewable materials¹⁷. Because of the hydrophobic polymers character and hydrophilic character of the natural fibers, the compatibility between them is very low and a major challenge¹⁹. Therefore, many techniques are applied to treat and modify the natural fiber surfaces to decrease absorption of moisture and enhance adhesion to the matrix of polymers. These treatments can be chemical, physical, or biological treatments. There are many factors that affect the changes in the chemical composition, properties, and structural aspects of fibers such as the origin of growth, the age of the fiber plant, climatic conditions, and different extraction patterns¹⁷. For clarification, some properties and chemical composition of several fibers will be mentioned in this research.

In this article some properties of biodegradable polymers, natural fibers, and biocomposites were defined. These properties include mechanical properties, thermal properties, visco-elastic behavior, acoustic properties, surface morphology and chemical composition of materials. The thermal properties of materials were examined by using thermal gravimetric analysis. The visco-elastic behavior was reported using dynamic mechanical analysis. The chemical composition of the composite and the functional groups which are responsible for the reaction within natural fibers and provide information about covalent bonding were identified by using Fourier-transform infrared spectroscopy.

Synthetic polymers have become necessary for several applications in daily life. They are non-biodegradable materials, so they accumulate in the environment causing serious problems. Biodegradable polymers are now gaining attention as an alternative to conventional polymers. Biodegradable polymers are materials that can be degraded naturally by the action of microorganisms producing eco-friendly and useful materials such as CO₂ and CH₄. Blending two or more biopolymers and mixing with natural fibers surface modification are used to improve the mechanical and physical properties of biodegradable polymers. Biopolymers can be used in a wide range of applications such as plastic bags, parts for automotive and medical applications. Biodegradable materials have the advantage of being able to be composted with organic waste or utilized to generate biomass at the end of usage of these materials. Also, biodegradable materials can be recycled to produce useful monomers and oligomers by microorganisms and use to produce the origin products. Biopolymer failures have been explained according to forensic engineering to try to avoid or reduce these failures during the manufacture and use.

DEFINITION OF BIODEGRADATION

The biodegradation of biodegradable polymers is defined as chemical decomposition of substances, which is accomplished through the enzymatic work of microorganisms that lead to a change in chemical composition, mechanical and structural properties and forming metabolic products, which are

environmentally friendly materials such as methane, water and biomass and carbon dioxide. Figure 1 shows biodegradation steps of polymers³. Extracellular enzymes and abiotic agents such as oxidation, photo-degradation, and hydrolysis depolymerize long-chain polymers and create shorter chains (oligomers) in the first stage^{20,21}. The biomineralization process, in which oligomers are bio-assimilated by microorganisms and then mineralized, is the second stage. Either aerobic and anaerobic degradation can occur. Aerobic degradation takes place in the presence of oxygen producing CO₂, H₂O, biomass and residue. Anaerobic degradation is carried out in absence of oxygen producing CO₂, H₂O, CH₄, biomass and residue²⁰.

Factors affecting biodegradation

The biodegradation process is affected by various factors including polymer morphology, structure, chemical treatment and molecular weight. (1) Polymer structure: biodegradable polymers have hydrolyzable linkages along the chain of polymer that are exposed to degradation in the presence of microorganisms and hydrolytic enzymes. Polymers with both hydrophobic and hydrophilic structures are more degradable than polymers containing either hydrophobic or hydrophilic structures¹¹. (2) Polymer morphology: enzymes attack the amorphous regions in polymers easily than crystalline regions as amorphous regions molecules are far apart from each other which makes it susceptible to degradation. The enzymatic degradation of polymers is also affected by the melting temperature (T_m) of polymers. Biodegradation of polymers decreases by increasing the melting point of polymers²².

$$T_m = \Delta H / \Delta S$$

Where, ΔH is the enthalpy changes on melting and ΔS is the entropy changes on melting²². (3) Radiation and chemical treatments: cleavage and crosslinking of polymers caused by radicals and/or ions produced by photolysis of polymers with UV and γ -rays irradiation. Oxidation additionally takes place complicating the situation that changes the polymer's capability for biodegradation¹¹. (4) Molecular weight: the biodegradability of the polymer reduces as the polymer molecular weight increases²².

BIODEGRADABLE POLYMERS

The environment suffers from serious problems from the increasing difficulties of disposing of plastic waste that resist microbial degradation²³. Therefore, the researchers tried to produce biodegradable, non-polluting, and environmentally friendly materials²⁴. In recent times, the natural and synthetic origin of biodegradable polymers was produced with good compatibility and biodegradability¹¹. As the biodegradable polymers receive great attention because they degrade into non-toxic and environmentally friendly materials²⁴. Mechanical strength, thermal and electrical properties of common biodegradable polymers and their composites are shown in Fig. 2a–c. Polyglycolide (polyglycolic acid, PGA) has high tensile strength (70–117 MPa). Thermoplastic starch showed low tensile strength (16–22 MPa).

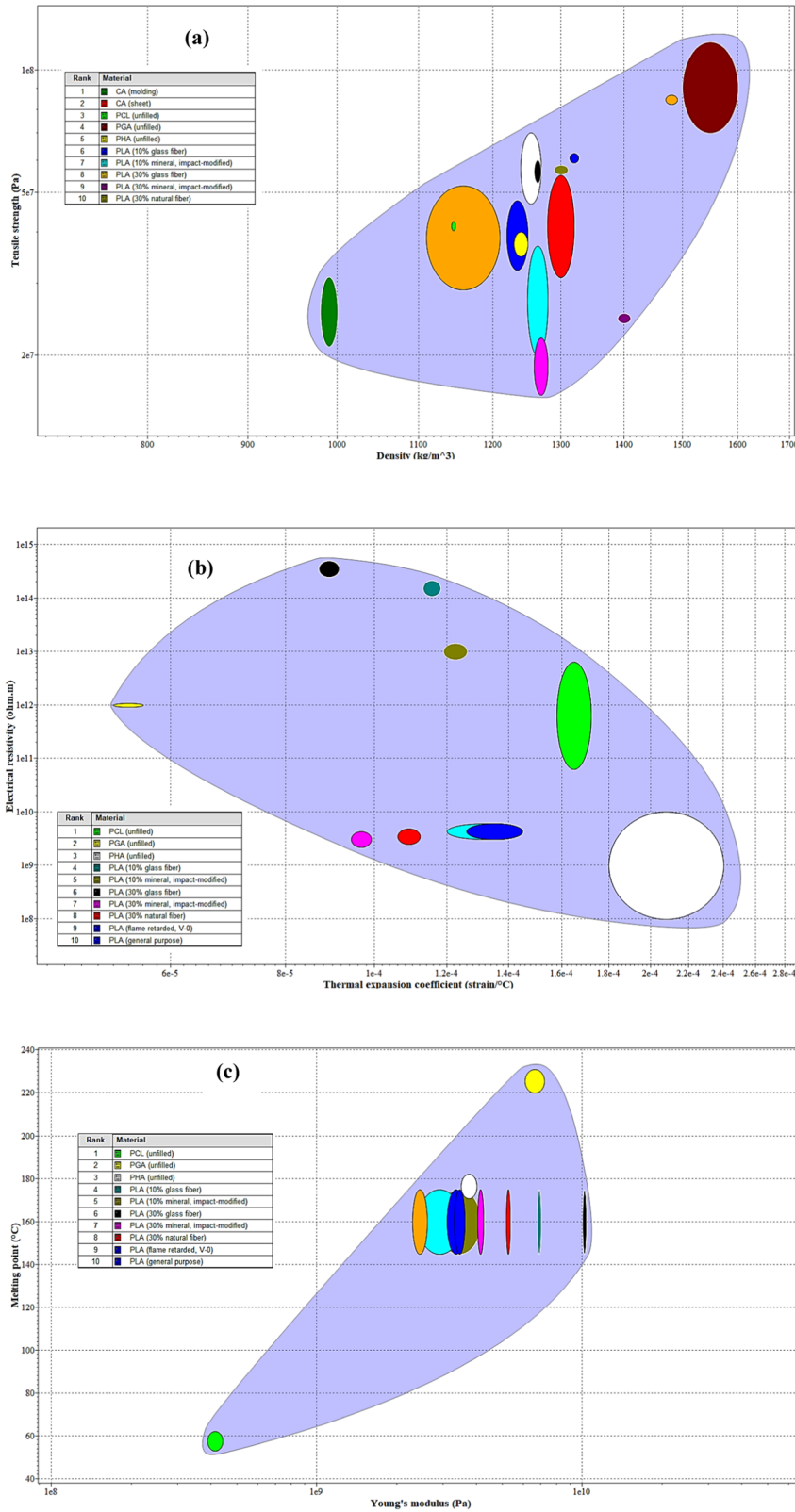


Fig. 2 Properties of biodegradable polymers and their composites. a Density vs. tensile strength. **b** Thermal expansion vs. electrical resistivity. **c** Young's modulus vs. melting temperature.

Polyactide (PLA)—glass fiber composites have high electrical resistivity (2.5×10^{22} – 4.9×10^{22}) $\mu\text{ohm.cm}$ as shown in Fig. 2b. Melting temperature of biodegradable polymers is

variant. Polyglycolide has high melting temperature (220–231 °C) and high Young's modulus (6.1–7.2 GPa) as shown in Fig. 2c.

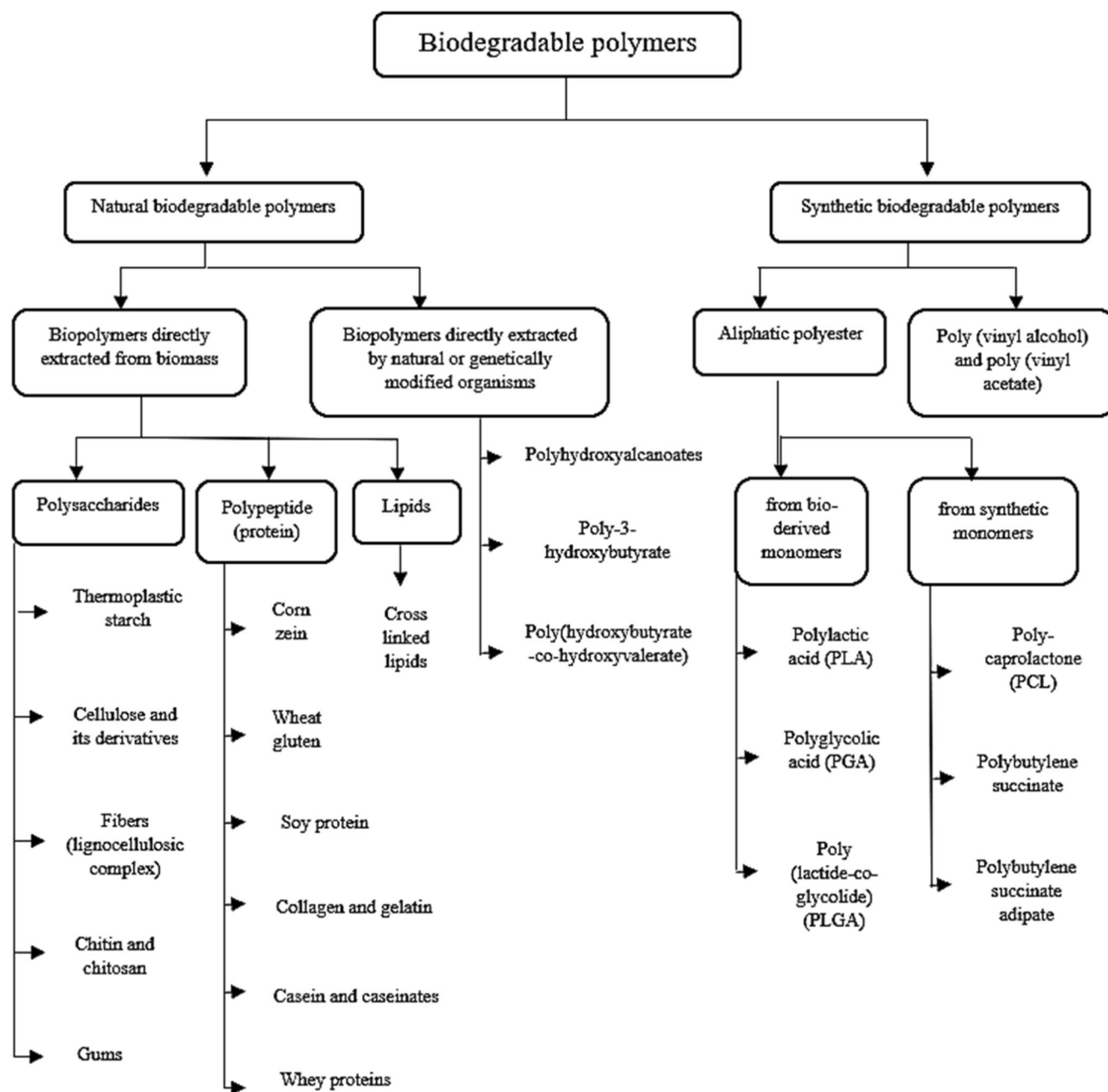


Fig. 3 Classification of biodegradable polymers. Schematic shows biodegradable polymers classification based on their origin and method of production.

Classification of biodegradable polymers

Biodegradable polymers may be categorized based on their origin and synthesis method, their chemical composition, economic importance, processing method and applications. They are categorized in this study based on their origin²⁴. Biodegradable polymers are classified into two groups based on their origin as indicated in Fig. 3. Natural biopolymers and synthetic biopolymers are made from natural resources and oil, respectively^{11,18}. Natural biopolymers are derived from renewable or biological sources such as animal, plant, marine, and microbial sources, while synthetic biodegradable polymers are manufactured chemically²⁵.

Natural biodegradable polymers. All organisms' growth cycles result in the formation of biopolymers in nature. Polymerization reactions with a chain of enzyme-catalyzed growth from active

monomers generated within cells through complicated metabolic processes are included in its production²³. They are naturally biodegradable and have good biocompatibility²⁶.

Biopolymers directly extracted from biomass. Agricultural waste is a major source for the production of bioplastics, plasticizers and antioxidant additives. The main source of polysaccharides is plant-based agricultural waste where biopolymers such as cellulose, starch, and pectin are produced^{27,28}. The use of agro-waste as a feedstock for biodegradable polymer synthesis can reduce both the cost of producing biodegradable polymers and the waste treatment cost. Biopolymers are produced through several methods, namely microbial methods, blending of biopolymers, and chemical methods²⁷. Biopolymers produced from agricultural plant waste have biodegradability, bio-functionality, biostability,

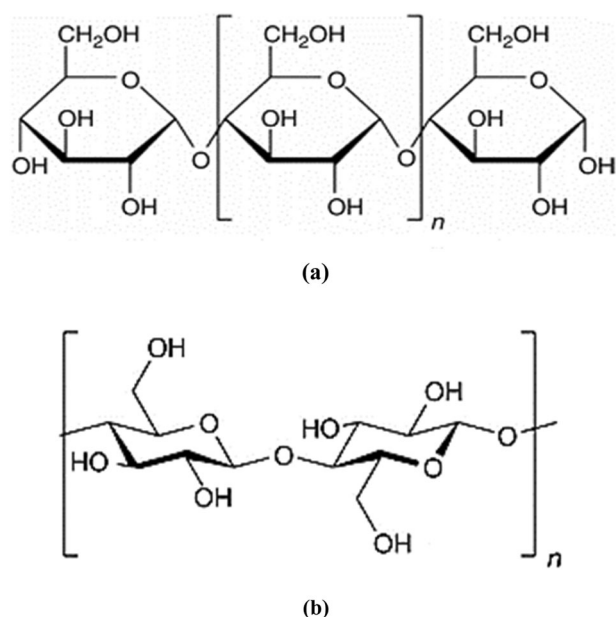


Fig. 4 Chemical structure. a starch, b cellulose.

and biocompatibility. They have wide range of chemical and mechanical properties that may be employed in several applications such as food packaging, biomedical applications, skincare, electrical electronics, vehicles and wastewater treatment²⁸.

Polysaccharides: Polysaccharides, proteins, and lipids are found in numerous applications in biodegradable products. Potatoes, corn, and rice are basic sources of starch production, where the chemical composition and granules size of starch varies according to the source of production¹¹. As shown in Fig. 4a, starch is a combination of linear (amylose) and branching (amylopectin) poly-(1,4) - α -glucose²⁹. The ratio of amylose to amylopectin has a substantial impact on the physicochemical characteristics of starch³⁰. Starch has poor mechanical properties, low impact resistance, water sensitivity, and brittleness properties. The properties can be improved by reinforcing the starch matrix with fibers or modifying the starch chemically or physically with other biodegradable polymers to enhance its properties^{24,31}. Starch is not completely soluble in water. It is partially soluble in water at room temperature, depending on the proportions of amylose and amylopectin^{32–34}. There are many advantages to starch biopolymers, such as high biodegradation, renewability, and good oxygen barrier properties that make them suitable in some commercial applications such as packaging applications, bags, cosmetics, adhesives, medical applications, and agricultural mulch films^{3,30,35}. One of the most common biopolymers is cellulose²⁹. It contains reactive OH groups in the backbone³¹. Cellulose is a polysaccharide with a molecular structure that is similar to starch. However, d-glucose units are attached to β -glycosidic bonds in cellulose. In starch, d-glucose units are linked to α -glycosidic bonds. As illustrated in Fig. 4b, cellulose consists of polymer chains made up of unbranched β (1 \rightarrow 4) connected D-glucopyranosyl units²⁹. Cellulose is the primary component of lignocellulosic plant cell walls. Hemicellulose, lignin, and other carbohydrate polymers are components of a gel matrix embedded with cellulose which exists in the lignocellulosic materials¹¹. Cellulose and cellulose derivatives are ecologically friendly materials that are widely utilized due to their ability to decay, compatibility with other materials, and regenerate. Because of cellulose's hydrophilic nature, moisture absorption causes the material's mechanical characteristics to deteriorate. As a result, cellulose derivatives are created through chemical modification, in which bonds are formed between the reagents and the OH

groups in the cellulose improving solubility and mechanical characteristics by reducing cellulose's hydrophilicity³¹. Cellulose and its derivatives are used in different applications such as fiber industries, wood products, textile, and pharmaceutical industries³.

Chitin is a polysaccharide can be produced by fungal fermentation processes or found in marine crustaceans such as shrimps, insects, the shells of crabs and lobsters. Chitosan is a deacetylated chitin derivative that makes up the exoskeleton of arthropods¹¹. Chitosan is a derivative of glucan that is produced by repeated monomers of chitin. Biodegradability, high strength, and nontoxicity are among the chemical and physical properties of chitosan and chitin³⁶. They have several applications in the field of medical and pharmaceutical sciences, tissue engineering applications, absorption of heavy metal ions, membrane barriers, the cosmetic industry and food packaging material^{3,36}.

Polypeptides (Proteins): Proteins are natural polymers produced from various vegetables and animals. Collagen is a protein found in both vertebrate and invertebrate connective tissue. It accounts for around half of all human protein. Collagen has a hydrophilic nature as a result of the enormous substance of essential, acidic, and hydroxylated amino corrosive buildups that lipophilic deposits¹¹. Collagen properties are exceptionally compelling in many fields, for example, leather chemistry, surgery, pharmaceutical industries like capsule production, etc.^{3,37}.

Biopolymers derived directly from naturally occurring or genetically engineered organisms. Microorganisms are a source for research on bioplastic materials and biopolymers that use agricultural wastes as a medium for growth. Example of microbiological compounding plastic is polyhydroxyalkanoate (PHA), which is produced from various groups of bacteria and cheap renewable resources. It is completely aerobic decomposed by microorganisms³⁰. polyhydroxyalkanoates are produced from a biodegradable aliphatic polyester family. They are formed in nature and produced directly by bacterial metabolism^{38,39}. They are genuinely biodegradable and profoundly biocompatible thermoplastic materials and can be developed from a variety of renewable resources. Polyhydroxyalkanoate is the potential alternative to the non-degradable polyethylene and polypropylene. Poly(3-hydroxybutyrate) (PHB) is the most representative member of this family^{11,38,39}. It is a natural polymer that formed by various chains of bacteria. It is produced from low-cost, renewable feedstock and without causing much impact on the environment. It is degraded in anaerobic and aerobic conditions and it doesn't produce any poisonous materials from its degradation²². Copolymer of hydroxybutyrate (PHB) and hydroxyvalerate (HV) is known as poly(3-hydroxybutyrate)-hydroxyvalerate (PHBV). It is a highly crystalline polymer. By increasing the HV unit content, the melting point, the glass transition temperature, the crystallinity and the tensile strength decrease but the impact strength increases. The copolymer PHBV is less brittle than PHB. The degradation rate of PHBV is higher than PHB^{40,41}.

Synthetic biodegradable polymers. Synthetic biodegradable polymers are produced by conventional polymerization procedures such as aliphatic polyesters, polylactide, aliphatic copolymer⁴². Due to the appropriate degradation time and their production in industrial scale, the degradable polyesters are considered as the most potential materials utilized comparing to conventional plastics. They are produced in several forms such as polylactic acid, polycaprolactone and polybutylene succinate. They have low ecological contamination⁴³. More than 90% of the biopolymers are polyester as they contain easily hydrolyzable bonds of esters. Synthetic biodegradable polymers may be categorized into bio-based polymers like PLA and oil-based monomers such as PCL^{12,42}.

Aliphatic polyester synthesized from bio-derived monomers (water-based monomers). The most significant biodegradable polymer is

polylactic acid (PLA)³. It is well-known for being biodegradable and biocompatible polymer²⁹. It is synthesized from renewable materials such as potato starch, wheat, rice bran corn and biomass^{22,31}. Polylactic acid is linear aliphatic polyester thermoplastic polymers²². Polycondensation and ring-opening polymerization of lactic acid monomers are two processes that may be used to synthesize PLA³. PLA's high molecular weight is achieved by polymerization of ring-opening. The final properties of the polymer can be controlled using this method⁴⁴. Lactic acid monomer exists in three diastereoisomeric structures which are D-lactide, L-lactide, and meso-lactide (DL-lactide)¹¹. Mechanical, thermal and biodegradable properties of PLA depend on the selection of stereoisomers distribution within the polymer chain. PLA is used in many applications such as interference screws, sutures, dental, ligating clips, bone pins and rods³.

Polyglycolic acid (PGA) is a linear aliphatic polyester that is synthesized by polymerization of ring-opening of cyclic glycolide monomers¹². It is a thermoplastic and biodegradable polymer. The degradation process of this polymer takes place in two stages. In the first one, water is dispersed in the amorphous regions of the polymer matrix where the ester bonds are separated. In the second stage, crystalline segment of the polymer gets vulnerable to hydrolytic attack¹¹. This polymer is used in several applications such as tissue engineering, medical devices, and drug delivery³. Glycolic acid monomers (GA) are copolymerized with L-lactide and LD-lactide (LA) generating poly(lactide-co-glycolide) (PLGA). The rate of degradation decreases when the L/G proportion increased⁴⁵. Mechanical and chemical properties are controlled by adjusting the ratio of monomers in the combined polymerization of the PLA and PGA without affecting the compatibility and biodegradability. The degradation of the PLGA takes place by hydrolysis of the ester bond and to form LA and GA which are eco-friendly materials³¹.

Aliphatic polyester synthesized from synthetic monomers (petroleum-based monomers). Polycaprolactone (PCL) is a biodegradable synthetic linear aliphatic polyester. It can be synthesized by polymerization of ring-opening of caprolactone monomers within the sight of metal alkoxides catalysts. Aluminum isopropoxide and tin octoate are common catalysts used in polymerization process^{22,46}. Polycaprolactone is dissolved in numerous solvents such as methylene chloride⁴⁶. The melting point of PCL is between 58–60 °C. It is degraded by the activity of anaerobic and aerobic microorganisms. The degradation rate of PCL depends on the level of crystallinity and its molecular weight²². This polymer is used in medical fields such as tissue engineering, long-term drug and vaccine delivery vehicles³. The biodegradability of PCL can be improved by copolymerization with aliphatic polyesters. PLA and PGA copolymer has lower crystallinity and melting point compared with the homopolymer^{22,45,46}.

Polybutylene succinate (PBS) can be prepared by polycondensation of 1,4 butanediol with aliphatic dicarboxylic acid succinic acid⁴⁷. It is a thermoplastic aliphatic polyester that has a melting point between 90–120 °C^{11,47}. It is decomposed by a hydrolysis mechanism²². Because PBS is crystalline materials, it degrades slowly¹¹. It is normally blended with different materials such as adipate copolymer which acquiring polybutylene succinate adipate (PBSA)²². This copolymer has a higher degradation rate than PBS¹¹.

Biodegradable polymer blends and composites

The main characteristics of biodegradable polymers are their primarily hydrophilic nature, high rate of decomposition, and potentially undesirable mechanical qualities. These properties could be improved by mixing natural and synthetic polymers^{11,48}. Blending polymers is a category of substances where two polymers at least are mixed collectively to produce a new material

with various physical properties. The purpose of mixing two or more polymers is to develop a blend that combines each polymer's preferred properties⁴⁹. The preparation of biodegradable polymer blends is normally associated with the blending of a thermoplastic resin with biodegradable one. These types of blends are expected to be more biodegradable than conventional plastics⁵⁰. After the degradation process of the biodegradable materials, the residuals components are more ecologically friendly and do not cause environmental pollution³.

NATURAL FIBERS

Natural fibers are perfect reinforcing materials for polymer composites (thermoplastics, thermosets, and elastomers). Natural fiber-reinforced polymer composites are gaining popularity due to their excellent mechanical properties and considerable manufacturing benefits, as well as the fact that they provide a solution to environmental contamination. Composites based on natural fibers have better impacts in the industry due to these fibers have high specific properties and low density. They don't have health hazards because of they are non-toxic⁵¹. Natural fibers are renewable materials, relatively high in tensile strength, low-cost, and light⁵². Natural fibers have become a substitute for non-renewable and expensive synthetic fibers (glass, carbon and kevlar fibers)⁵³ in different applications because of environmental aspects and their properties as shown in Fig. 5⁵⁴. Natural fibers contain some desirable properties, including high specific strength and modulus, flexibility during treatment, and excellent corrosion resistance⁵³. However they show some limitations such as high anisotropy, moisture absorption, limited compatibility with conventional resins, and inferior homogeneity when compared to glass and carbon fibers^{55,56}. For centuries, natural fibers used in several applications such as clothing, making baskets, ropes and various parts of automobiles⁵⁴.

Classification of natural fibers

Natural fibers may be categorized based on their sources⁵⁷. Natural fibers come from three main sources namely; animal, plant, and mineral materials⁵⁸. Plant fibers include bast (stem) fibers, seed fibers, leaf fibers, fruit fibers, core fibers, cereal straw fibers, wood fibers and other grass fibers. Animal fibers include hair and wool. Mineral fibers such as asbestos and glass fiber.

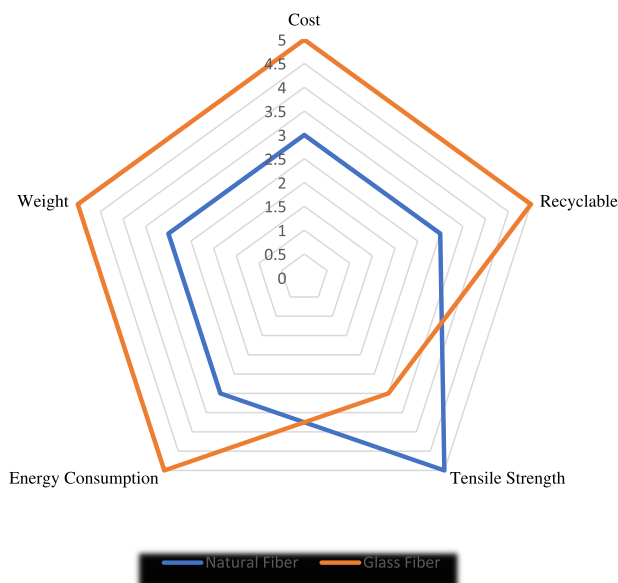
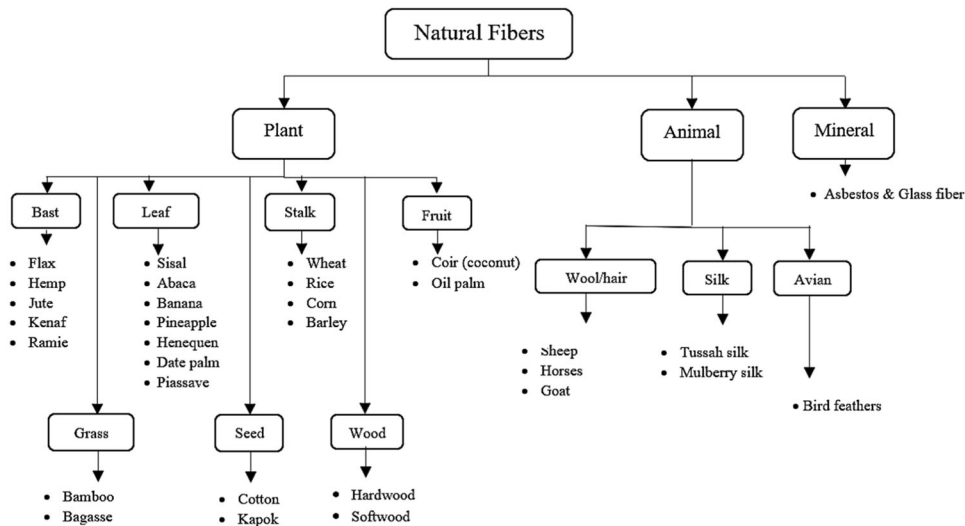
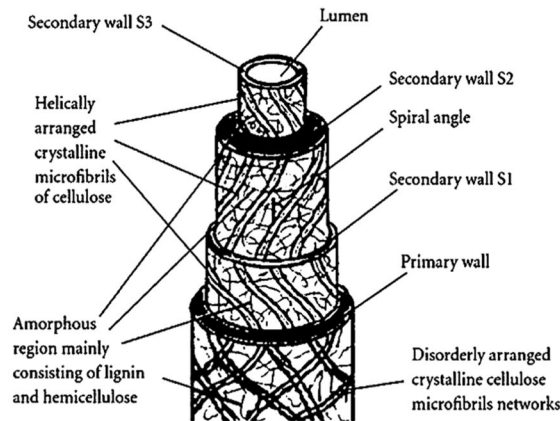


Fig. 5 Properties and limitations. Natural fiber vs. glass fiber⁵⁴.



(a)



(b)

Fig. 6 Natural fibers. **a** Classification of natural fibers. **b** The schematic picture shows the cell wall of natural plants⁵⁵.

Figure 6a, b presents the classifications of the different natural fibers and cell wall of natural plants respectively^{55,57–59}.

The plant fibers can be categorized into bast fibers, fruit fibers, leaf fibers, grass fibers, seed fibers, wood fibers, and stalk fibers. Bast fibers like ramie, hemp and flax. They come from the skin and bast around the plant stem. Fruit fibers that are collected from fruits such as coconut and palm. Leaf fibers that are extracted from leaves such as sisal, pineapple, abaca and banana. Seed fibers that are developed from seeds and seed shell such as kapok and cotton. Stalk fibers which are generated from the stalks of the plants such as wheat, rice, and corn. Grass fibers include bamboo and bagasse. Finally, wood fiber (hardwood and softwood)^{58–61}.

Cotton fiber. The purest form of cellulose is cotton. Cotton is hydrophilic. It has high elastic, flexibility and elongation at break. It is degraded by acids but it has resistance to alkalis. Cotton has low microbial resistance however the fibers are highly resistant to beetles and moths. Cotton is burning very fast⁵⁷. Although cotton fibers are widely used, their mechanical performance suffers behind other natural fibers, notably different types of bast fibers. This can be attributed to the low degree of fiber orientation⁵⁹.

Coir fiber. Coir is developed from the coconut fruit⁶². It is found between the outer shell of the coconut and its husk⁶⁰. Among several natural fibers, coconut fiber has many desirable properties such as durability, high hardness, acoustic resistance, resistance to fungi and moths, and hardly burns. Also, this fiber is more moisture resistant than other natural fibers. It was reported that the coir fiber has saltwater and heat resistance⁶³.

Flax fiber. Flax fiber is the strongest fiber comparing with the other natural fibers⁶⁴. It is a renewable, and biodegradable material. Great attention increased for flax fiber-reinforced plastic composites because of its great toughness, high strength, stiffness, low density, and bio-degradability. When compared to E-glass fiber, flax fiber has a higher specific tensile strength⁶³. Flax fiber is widely used in several applications such as towels, carpets, ropes and fabrics. Also, flax fiber has been used in biocomposites fabrication⁶⁵.

Kenaf fiber. The outer (bast) and inner (core) fibers of the kenaf plant are used to produce kenaf fiber⁵⁷. Kenaf fibers are used in polymer matrix composite as reinforcement materials. It is non-abrasive during manufacturing, has a low density, and good

Table 1. Some natural fibers' chemical composition.

Fiber	Cellulose (wt %)	Hemicellulose (wt %)	Waxes (wt %)	Lignin (wt %)	Pectin (wt %)	Reference
Cotton	89	4	0.6	0.75	6	79
Flax	71	18.6–20.6	1.7	2.2	2.3	77
Kenaf	53.5	21	—	17	2	79
Jute	61–73.2	13.6–20.4	—	12–16	—	59
Hemp	81	20	0.8	4	0.9	79
Ramie	68.6–76.2	13.1–16.7	0.3	0.6–0.7	1.9	77
Abaca	62.5	21	3	12	0.8	79
Sisal	67–78	10–14.2	2	8–11	10	77
Coir	46	0.3	—	45	4	79
PALF	70–82	—	—	5–12	—	77

specific properties. It is also biodegradable⁶³. On the other hand kenaf fibers are rough, brittle, and hard to process. It is widely used in textiles, paper materials, ropes, absorbents, and building materials⁶⁶.

Hemp fiber. Hemp fiber belongs to the bast fiber family⁶³. It is used in polymer composites reinforcement⁶⁷. It consists of 55–72% cellulose, 8–19% hemicellulose, 2–5% lignin, 4% minerals and less than 1% wax⁶³. It has excellent mechanical strength, elastic modulus and moisture resistance^{46,52}. Hemp fiber has chemical composition comparable to flax fibers, however flax is less resistant to moisture than hemp. Hemp fiber has a toughness that is roughly 20% greater than flax, although they have a poor elongation⁵⁹. Hemp fiber is used to produce paper, fabrics, construction materials, plastic and composites⁶⁸.

Jute fiber. Jute fiber is extracted from the ribbon of the stem. Jute fiber has wood-like properties. It has good mechanical and thermal properties, high aspect ratio, high strength to weight ratio and good properties of insulation⁶³. Jute has a microorganism's resistance, but it is sensitive to photochemical and chemical attacks. Jute fiber is brittle because of high content of lignin and a small extension to break. It has tensile strength lower than flax or hemp. Exposure to sunlight causes loss of the tensile strength of the fiber and has little resistant to acid and moisture. Jute is the plant fiber with the highest hygroscopicity. Jute is silky texture, biodegradable material and resistant to fire and heat which makes it adequate for use in industries. It is used in travel luggage, fashion, furnishings, carpets and other floor coverings. Jute fibers may be used to strengthen polymer composites (thermoset and thermoplastic)⁶⁹.

Ramie fiber. Ramie fiber is white in color, very fine and silk-like. Ramie fiber is naturally resistant to germs, insects, and mildew. Ramie fiber is not harmed by mild acids and stable in alkaline media⁵⁷. The physical properties of ramie fiber showed high tenacity, high luster and brightness⁷⁰. The fiber has an exceptional strength which slightly increases when wet. Ramie fiber is gaining attention as a polymer matrix reinforcement in composites⁵⁷. Natural fiber has much less density than that of synthetic fibers⁶³.

Sisal fiber. The sisal fiber comes from the sisal plant leaves which is a hard fiber⁴⁶. As compared to glass fiber, sisal fiber has a higher strength and stiffness⁷¹. Sisal fibers include a number of environmentally friendly benefits such as low density, excellent specific modulus, and strength. These advantages make them appropriate for use as reinforcement in composites⁷². When the temperature of the sisal fiber increased, the tensile strength, modulus, and toughness for fiber decrease. It is smooth, yellow, straight and degraded easily in saltwater⁵⁶. The fiber increases

the toughness of polymer than increasing the modulus and strength. The sisal fiber composites had maximum toughness than other fibers⁷³.

Pineapple leaf fiber (PALF). PALF is made from the leaves of the pineapple plant⁵¹. It's a cheap waste product from pineapple farming⁵⁷. PALF consists of high content of cellulose about 70–82%, polysaccharide, and lignin^{51,57}. PALF has superior mechanical properties because of the high content of cellulose⁵³. As compared to cotton, dyes faded more quickly due to the high content of lignin and wax materials in pineapple leaf fiber. Also, dyes difficult to penetrate due to the high coarseness of PALF⁵¹.

Abaca. The abaca is a banana species. that comes from the leaf sheaths of the banana plant^{57,59,63}. It is yellowish-white in color and lustrous fiber. It is the strongest fiber among all other plant fibers. Abaca tensile strength is twice times higher than sisal and three times higher than cotton⁵⁷. It is resistant to degradation in saltwater than other fibers, so it is preferred to use in marine applications⁶³. Abaca fibers are used in many applications such as manufacturing ropes, bags, slippers, placemats and doormats. Also, abaca fiber is used as a reinforcement of composites⁷⁴.

Bagasse and bamboo. After sugar is extracted from the sugarcane stalk, bagasse is a fibrous residue that remains⁶³. It is used in many applications like biofuel in the production of renewable energy and used as a natural fiber-reinforced polymeric composite^{63,75}. Bamboo fiber is a kind of cellulosic fiber⁷⁵. It has eco-friendly characteristics such as high growth rate, lightweight, high strength and it is a biodegradable material. It is used in many applications such as flooring to ceiling and transportation, furniture manufacturing, etc. Also, it is used as a reinforcement of composites⁷².

Palm fiber. It is biodegradable, compostable, and has the potential to break down into its essential parts and return to the environment naturally. Palm fiber has five times the tensile strength of structural steel and is comparable to flax, hemp, and sisal. It has better vibration dampening and acoustic insulation than glass and carbon fiber as well as better heat insulation than carbon⁶⁰. Palm fiber has a cellulose purity of up to 70% and a thermal stability of up to 226 °C.

Animal fibers. Animal fibers are the most significant natural fibers after plant fibers. They are employed in composites as reinforcement⁵⁷. Animal fibers include mainly three types: hair, avian and silk fibers. Animal hair fibers are obtained from animals and hairy mammals such as horses, sheep, and goats. Silk fibers are obtained from insects while preparing cocoons or from dried saliva from insects. Avian fibers are produced from bird feathers⁵⁸.

Structure and composition of natural fibers

Natural fiber cell walls are made up of three layers: a main cell wall, a secondary cell wall, and an intermediate cell wall known as lumen⁵⁹. Fig. 6b depicts the structural organization of a natural fiber cell wall^{52,59,76}. This structure is known as microfibril⁶³. During cell formation, the wall of the primary cell is the first layer formed. Secondary walls S1, S2, and S3 are the three layers that make up the second cell wall⁷⁷. The lumen layer is responsible for the transportation of water⁷⁸. The cell walls are formed of a semi-crystalline cellulose microfibril, hemicellulose, lignin, wax, pectin and water-soluble compounds^{63,77}.

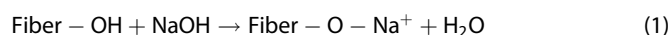
The physical properties of the fiber are connected to the inner structure and components of the plant material that is being used. The fiber of plants is lignocellulosic structures which consist of hemicelluloses, cellulose, and lignin, as well as pectin, protein, wax, ash, tannins, and inorganic salts⁵⁹. These components are vary according to the fibers sources, growth conditions, age of plant and processes of digestion^{59,63}. The chemical composition of some of the common natural fibers are presented in Table 1^{59,77,79}. Cellulose content of fibers is the most important factor in determining their characteristics and mechanical performance when utilized as reinforcement in composites. In contrast to cellulose, an increase in non-cellulose components causes a decrease in fiber strength and modulus, which has severe consequences for composites reinforced with natural fibers⁵⁹.

Surface modification of natural fibers

Natural fibers have a variety of drawbacks in reinforcement composites, including poor compatibility with the polymer matrix due to the hydrophobic character of the polymer matrix and the hydrophilic character of the fibers. Their moisture absorption, and dimensional stability are considered as main limitations^{63,80}. The high wettability can be attributed to the existence of OH groups, and polar groups. The mechanical properties of natural fibers decreased when the moisture content in natural fiber increased. This led to a loss in dimensional stability and degradation. This causes weak adhesion between the polymer matrix and the natural fiber when the natural fibers used as reinforcement in composites¹⁷. Surface adhesion is a key factor in describing component mechanical and physical characteristics^{80,81}. These problems can be solved by surface modification treatment such as chemical, physical and biological treatments¹⁷.

Chemical treatment. Since natural fibers are hydrophilic and polymer matrix is hydrophobic, there is an inherent incompatibility between them, this results at the interface in weak bonding. Chemical treatment methods will reduce the fiber's hydrophilic nature by removing hydrophilic OH groups from reinforcing fiber⁸¹. This treatment strengthens the adhesion of the polymer matrix to the fiber via chemical reactions^{53,60}.

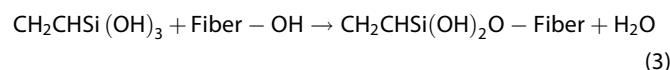
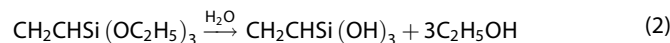
Alkaline treatment or mercerization: This method of treatment is the simplest, most cost-effective, and efficient for improving the adhesive capabilities between polymer matrix and natural fibers⁵³. In this chemical treatment, aqueous sodium hydroxide (NaOH) is used. Natural fibers are soaked in a predetermined concentration of NaOH for a certain time and temperature. Non-cellulosic components like lignin, hemicellulose, oils, and wax are removed during this process⁶⁰. The following reaction shows alkali treatment in below equation⁸⁰.



The removal of non-cellulosic components modifies the polymerization degree and structural orientation of cellulose crystallites, altering the chemical composition of the fibers. Also, this treatment has a permanent effect on mechanical fibers behavior, particularly on their stiffness and strength. Various alkaline treatment studies indicated that mercerization would increase

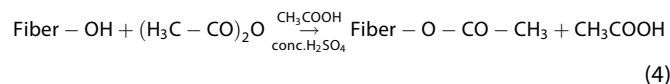
the amorphous cellulose amount at the crystalline cellulose expense and remove hydrogen bonds in the network structure⁸⁰. Neutralizing the fibers can be achieved by using acetic acid to end the reaction by removing the rest of the hydroxyl groups after washing the fibers with distilled water⁸².

Silane treatment: Silane is a molecule of multifunctional. It is used as a binding agent to adjust the surface of fibers. Silane coupling agent develops a siloxane bridge chemical bond between the polymer matrix and the fiber. Silanols form when moisture and a hydrolysable alkoxy group are present. One silanol end reacts with the matrix functional group during condensation, whereas the other end reacts with the cellulose hydroxyl group⁸¹. The following reactions show silane treatment in below equations⁸³.



The number of hydroxyl groups in natural fibers is decreased during this treatment, which leads to a reduction in absorption of moisture and improves the fiber's mechanical properties⁸². The saline treatment increases the composite's tensile strength while also reducing the influence of moisture on its characteristics. It also enhances fiber-polymer matrix adhesion⁸⁰.

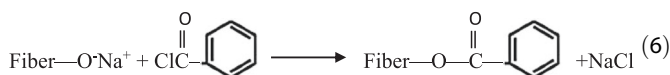
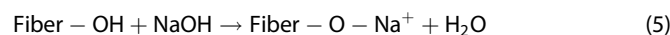
Acetylation treatment: In acetylation treatment, acetyl groups are grafted to the cellular structure of the fibers by using acid catalyst⁸⁴. The fibers are firstly soaked in acetic anhydride. They are processed for 1-3 h in acetic anhydride at an elevated temperature (70–120 °C) in order to accelerate the reaction. This process is known as esterification method for plasticizing natural fibers with the formation of acetic acid as byproduct. The hydrophilic hydroxyl groups of the natural fiber react with the acetyl group (CH₃CO). To swell the cell wall and activate the reaction, a small amount of acetic acid is required in the reaction mixture. Equation (4) shows the acetylation treatment of natural fiber⁸⁰.



When the natural fiber's moisture content decreases, the fiber's hydrophilic nature decreases⁶⁰. The removal of non-cellulosic components from the fiber improves moisture resistance⁸¹. The dimensional stability and degradation of composites are also enhanced by this procedure⁸⁰.

Benzoylation treatment: Benzoyl chloride is utilized in this procedure. This treatment reduces the fiber's hydrophilic nature, improves fiber-polymer adhesion, and increases composite strength. Alkaline treatment is used while treating benzoylation. Non-cellulose compounds are removed during the alkaline treatment process. The amount of reactive OH groups on the fiber's surface likewise increases. After that, the fibers are exposed to benzoyl chloride treatment. The hydroxyl groups are replaced by benzoyl groups and found in the cellulose during benzoylation treatment⁸¹. This treatment minimizes the absorption of moisture and enhances the thermal stability of the fiber.

Equations (5) and (6) describe the steps of benzoylation treatment^{85,86}.



Peroxide treatment: The connection between the natural fiber and the matrix can be enhanced by peroxide treatment⁸¹.

Peroxide is a specific functional group with function group RO—OR' which contains divalent ion O—O. Peroxide tends to decompose to free radical in the form of 2 RO[•]. The peroxide decomposition is completely achieved by heating the solution at high temperature^{81,86}. The hydrogen group of the cellulose fiber and the matrix react with the free radical RO[•]. Benzoyl peroxide and dicumyl peroxide are used in the modification of natural fiber surface. In this treatment, dicumyl peroxide or benzoyl peroxide is used to coat the fibers in acetone solution for about 30 min at a temperature of 70° C^{81,87} after pre-treatment of alkali. The concentration of peroxide solution is about 6% and saturated peroxide solution is used in acetone⁸⁶. The following reactions show the peroxide treatment in below equations⁸¹.



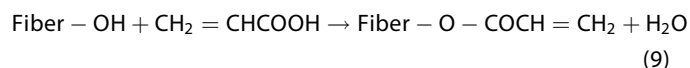
where:

R = the functional group of peroxide

Pretreatment on the fiber with benzoyl peroxide or dicumyl peroxide produces high mechanical properties of the composition⁸¹. After treatment, the treated fiber should be washed with distilled water⁸⁷. This treatment increases moisture resistance and improves thermal stability⁸⁶.

Maleated coupling agents: Efficient interaction between the polymer and the fiber can be achieved by maleated coupling agent⁸¹. Maleic anhydride is commonly used to treat the surface of the fibers as well as the polymeric matrix to enhance the compatibility between polymer and natural fiber. This treatment differs from other chemical treatment methods as it is used to treat both the fibers' surface and the polymeric matrix. The mechanical properties of polymer matrix-natural fiber composites are improved by this treatment⁶⁰. hydroxyl groups in the amorphous region of the cellulose structure interact with maleic anhydride during grafting, eliminating hydroxyl groups from the fiber. This treatment decreases the hydrophilic tendency⁵³.

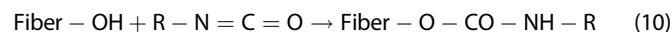
Acrylation and acrylonitrile grafting: This treatment is used to improve the bonding between the polymer matrix and the fiber by using acrylic acid (CH₂=CHCOOH). It reacts with hydroxyl groups of the fiber⁵³. The chemical reaction between the carboxylic acids in the coupling agent and the cellulose OH groups form linkage of ester. Equation (9) shows acrylation treatment⁸⁸.



This treatment reduces the fiber's hydrophilic character and increased its moisture resistance. As a result, peroxide radicals are employed to start the grafting of acrylic acid on the matrix. Peroxide's O—O bonds remove hydrogen atoms from the tertiary carbon of the polymer chain. The stress transfer capacity at the interface between the fiber and the polymer matrix is enhanced by acrylic acid and therefore improves composite properties⁸¹. After 1 h of fiber immersion in various concentrations of acrylic acid at elevated temperatures, alkali-pretreated fibers are washed with an aqueous alcohol solution and then oven-dried^{84,88}.

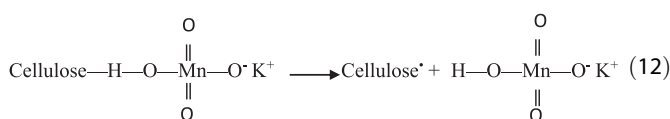
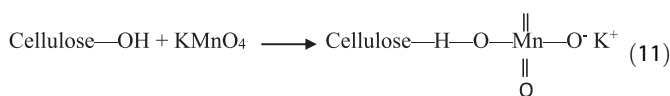
Fiber grafting with acrylonitrile (AN) (CH₂=CH-C≡N) is a method for modifying the surface of the fibers. Free radicals are initiated by acrylonitrile and reacted with the molecules of cellulose in fibers and the matrix monomer. The interlocking efficiency at the interface is improved as a result of the copolymerization process between the polymer matrix and the fiber⁸¹. It was reported that sisal fibers showed 25% lower tendency to absorb water, high tensile strength, and modulus characteristics of the polymer composite as a result of acrylation treatment⁸⁴.

Isocyanate treatment: Isocyanate is a coupling agent used to enhance and modify the surface of natural fibers⁸¹. In this treatment, the reaction between the functional group of isocyanate (-N=C=O) and the OH groups of cellulose and lignin components of the fibers forms a urethane group^{81,89,90}. Equation (10) shows the isocyanate treatment of natural fiber²⁴.



The bonding properties between the polymer matrix and the fibers are improved by forming strong covalent bonds⁵³, resulting in high compatibility with the binder resin in the composites^{89,90}. Also, urea is formed because of the reaction between the moisture content that exists on the surface of the fiber and isocyanate. This urea can react with the celluloses' OH groups⁸¹. The second reaction results in a fiber with a stronger moisture resistance property as well as a good bonding with the polymer matrix. As a result, mechanical properties and compatibility of polymer composites are improved^{53,81}.

Permanganate treatment: The chemical crosslinking at polymer matrix and the natural fibers interfaces can be improved by treating the natural fibers using permanganate⁵³. Permanganate treatment is conducted using potassium permanganate (KMnO₄) in a solution of acetone. Reactions between highly reactive permanganate ions (Mn⁺³) and the OH groups of cellulose are carried out in this treatment. This treatment forms cellulose-manganate which improves thermal stability. The alkaline treatment is required included with this treatment⁸¹. Chemical reactions using the permanganate treatment are given in below equations⁸⁵.



Also, the hydrophilic character and water absorption of the fiber-reinforced composites decrease^{85,86}. This procedure improves the adherence of the polymer matrix to the fiber⁵³. A high concentration of KMnO₄ (more than 1%) leads to a decrease in the hydrophilic tendency of the fiber⁹¹ and causes excess delignification inside the cellulosic structure and reduces the properties of the fibers. According to Sheng et al., reported that treatment of natural fiber using potassium permanganate increased the surface area and made it coarser during the oxidation reaction, which improved mechanical entanglement with the matrix⁸¹.

Physical methods. Natural fibers can be physically treated to improve the mechanical interaction between the polymer matrix and the fibers while maintaining their chemical properties⁶⁰. Physical treatment includes the use of plasma, corona, ultrasound and UV light¹⁷. These procedures are only utilized and applied to change the surface qualities of natural fibers⁵³. Plasma treatment is the most common physical treatment because it stimulates the substrate surface when employed in the treatment of natural fibers^{92,93}. Surface contaminants are the cause of changes in surface qualities including dyeability, wettability, and flammability, hence plasma treatment can be employed to reduce surface contamination¹⁷. Generally, the hydrophobicity of the natural fiber is increased as a result of plasma treatment¹⁶. Plasma treatment would increase surface roughness resulting in good binding to the polymer and the adhesion between the polymer matrix and natural fiber¹⁷. Corona treatments can be employed as a

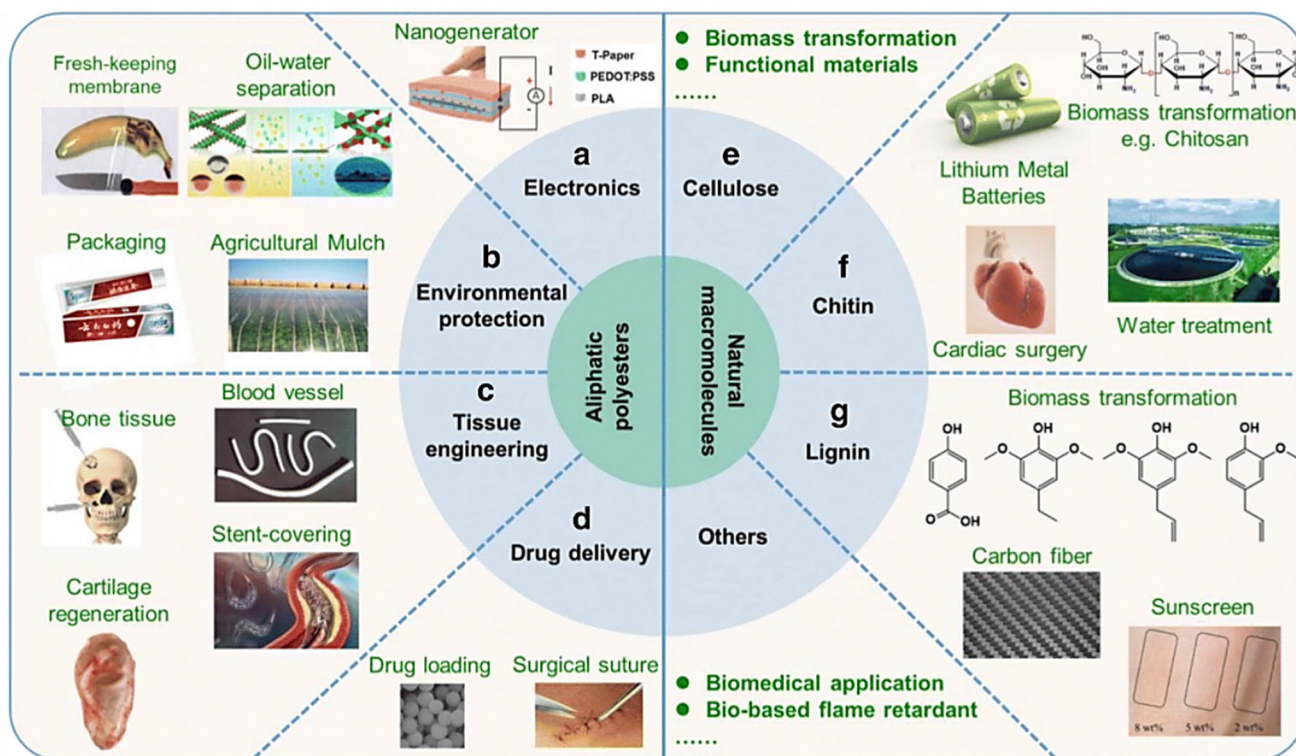


Fig. 7 Applications of biodegradable materials. Snapshot of the main fields of application¹⁰¹.

stand-alone surface modification or as a pre-treatment to activate cellulose in preparation for future chemical treatments like grafting^{59,94}. Corona treatment improves the compatibility of hydrophobic polymer matrix with hydrophilic fibers.^{60,95} Ultraviolet treatment is also used in surface fiber modification which leads to an increase in the fiber's thermal stability⁹⁶.

Biological treatment. Natural fibers are commonly treated using chemical and physical processes. However, they have some drawbacks such as usage of large volumes of solvents and hazardous chemicals, generation of wastes and pollutants, energy waste, and required costly equipment and chemicals. Microorganisms like fungi, enzymes and bacteria could be used in natural fibers treatment¹⁷. Biological treatment is an eco-friendly method⁹⁷, more economical than other treatments, less energy waste and improves the thermal stability of the fibers. It was also found to be selective in removing pectin and hemicellulose⁹⁸. Fungal treatment is a new biological treatment that removes of non-cellulose components of the fiber surface by using specific enzymes, increased fineness of the fiber, and fiber individualization can be achieved^{98,99}. Extracellular oxidizing enzymes produced by white-rot fungus react with lignin and aid in its elimination, as well as enhancing hemicellulose solubility and lowering the fiber's hydrophobicity. Furthermore, fungi generate hyphae, which causes fine pores on the surface of fiber as well as a rough surface to improve fiber/matrix interaction. Without the use of fungus, enzymes might be applied directly to natural fibers. By increasing cellulose content and eliminating lignin and hemicellulose, enzymes like xylanase and laccase increase hardness, tensile, and flexural strength¹⁰⁰.

APPLICATIONS OF BIODEGRADABLE POLYMER

The use of biodegradable polymers is rapidly growing with a global industry worth many billions of dollars annually. Biodegradable polymers are used in a variety of applications, including

food packaging, computer keyboards, automotive interior parts, and medical applications like implantable large devices, medical delivery and tissue engineering^{101–104}. Figure 7 shows various applications of biopolymer materials¹⁰¹.

ENVIRONMENTAL FATE AND ASSESSMENT

Biodegradable polymers have been designed and developed over the past 20 years. They are used in applications that advantage biodegradation. Biodegradation is a biological process in which bacteria digest dead organic matter and convert it to microbial energy and biological mass while releasing inorganic compounds as by-products. Biodegradation is used in waste management to convert biowaste into compost for soil fertilization through organic recycling. Anaerobic digesters are another type of organic recycling system that produces biogas and digestate, which is subsequently transformed into compost. Biopolymers are developed to be reused in composting facilities and anaerobic digesters with bio-wastes. Biopolymers are also utilized in agricultural plastics that are made to be left in the field and biodegrade after usage. The environmental effect of compounds produced during polymer biodegradation and composting, which might then be dispersed into the environment by compost fertilization or directly diffused during their biodegradation in soil, is a recurring subject. End-of-life possibilities for biodegradable polymers are represented in Fig. 8.

Biodegradation end products

Chemical elements can be found in nature as components of organic molecules (such as polysaccharides, and so on) as well as in inorganic substances (such as NH_3 , CO_2 , and so on). Microorganisms transform lifeless organic materials into inorganic chemicals during biodegradation. Glucose molecules, for example, are converted back into the inorganic compounds that plants used to create glucose via aerobic biodegradation. This process is termed as mineralization as it leads in the transformation of

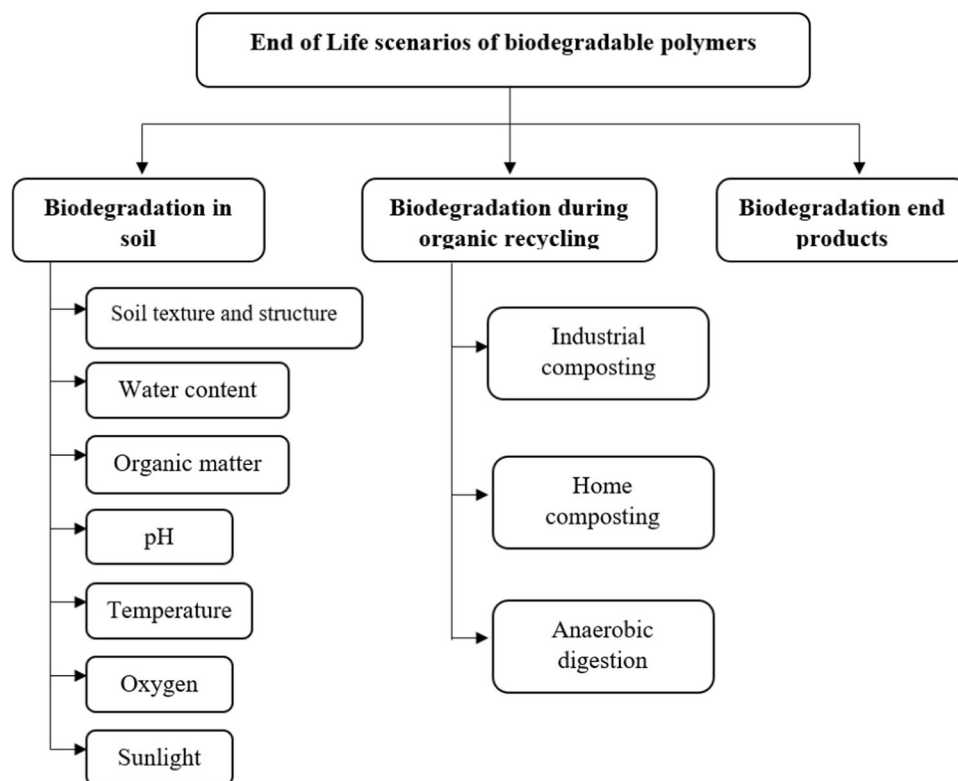


Fig. 8 Environmental assessment. End-of-life scenarios of biodegradable polymers.

organic material molecules into inorganic compounds and minerals. Organic molecules, like natural polymers and certain man-made polymers, are affected by the biodegradation process. Biopolymers are used in the manufacture of plastic materials that are designed to decompose in the soil or compost plants. Biodegradation of a polymeric compounds in which part of the original carbon (C_{polymer}) is mineralized (CO_2), part is consumed by microorganisms for their own development and reproduction (C_{biomass}), and the rest remains as polymeric residue (C_{residue}). Other kinds of microorganisms are engaged in the biodegradation process under anaerobic circumstances. As a result, products such as CO_2 and CH_4 are produced.

Biodegradation during organic recycling

Organic recycling is a treatment process of bio-waste that results in the generation of compost (stabilized organic matter) that is utilized as a soil fertilizer in agriculture. Organic recycling is standardized and industrial biotechnology that involves a multistep biodegradation process in anaerobic digesters or aerobic composting plants. Organic recycling involves three strategies namely; industrial composting, home composting, and anaerobic digestion.

Industrial composting. This technique can be used to handle bio-waste obtained by home, industrial, and agricultural processes. In addition, bio-waste from sewage treatment, park and garden upkeep are used. Composting refers to organic matter recycling method that turns waste into compost. Biodegradable plastic materials that have been used before are an ideal feedstock for industrial composting. Bio-waste is collected in industrial composting plants, where a variety of elements come together to provide a perfect environment for microorganisms to improve the composting process: temperature, moisture, and pH change over time. During the process, O_2 should be available. Microorganisms get energy and chemical

ingredients for their own survival, development, and reproduction through this mechanism. Microbial metabolism generates heat, which leads to increase pile temperature. As the temperature of the mass increases, quicker reactions take place, speeding up the biodegradation process.

Home composting. When compared to industrial composting, home composting is applied to a lower amount of bio-waste created through domestic activities or garden upkeep and is done in a much more varied manner. As a result, home composting can provide different results due to the fact that numerous elements influence the process such as moisture, temperature and types of microorganisms. Because the compact dimensions of the composting masses may not be allowed for high temperatures to be attained, home composting is frequently slower than industrial composting.

Anaerobic digestion. Bio-waste is decomposed by a bacterial population in the absence of oxygen, resulting in the formation of biogas (methane and carbon dioxide) and digestate, with little or no exothermic heat released. A two-step method is used by most commercial anaerobic digestion systems. Anaerobic fermentation is the initial process, followed by aerobic composting in the second step.

Biodegradation in soil

Several biopolymer-based applications that end up in the soil after use are fast growing in the market. Furthermore, it is used to improve soil quality, mature industrial compost, which is made from a feedstock containing biopolymers, ends up in the soil. Soil is a heterogeneous material governed by a mix of environmental and seasonal elements that tightly control the microbial population's creation and activity. For example, bacteria colonize an alkaline-neutral humid soil, however fungi require acid dry soil to thrive and operate.

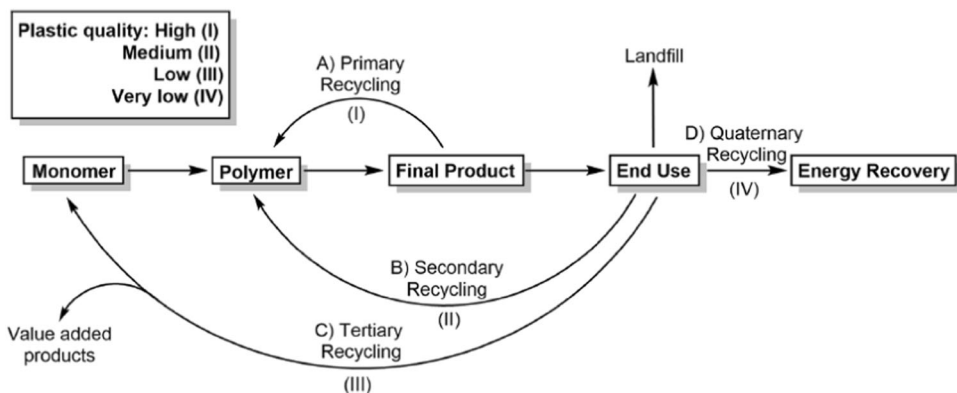


Fig. 9 Recycling routes. Different types of plastic waste recycling and their impact on plastic quality¹⁰⁶.

Soil texture and structure. Soil texture is formed based on the percentages of clay, sand, and silt particles that make up the soil. The size of clay particles is less than sand and silt particles. Microbial colony development is influenced by soil texture and structure: sandy, dry, and well-aerated soil. It promotes fungal colonization. Clay compact soil with inadequate aeration supports facultative aerobic or microaerophilic bacterial colonies.

Water content. The texture, structure, and organic material concentration of the soil contribute to the soil's water retention, which has a big influence on microbial selection. The formation of bacterial colonies is aided by humidity, whereas the growth of fungal colonies is aided by dryness.

Organic matter. Organic matter is the preferred substrate for the formation and expansion of microbial colonies since it is the major fuel source for biodegradation. It also acts as a soil buffer, contributes to soil aeration and humidity, and has a good impact on the preservation of microbial habitat.

pH. Microorganisms display varying degree of sensitivity at different pH values. Therefore, pH changes have a significant impact on microorganisms growth. As a result, soil pH is an important factor in restricting microbial colonization. Bacteria prefer a slightly alkaline neutral pH, whereas fungi prefer an acidic pH.

Temperature. The temperature has an impact on microbial activity and presence because it permits microbe colonies to form. Warm temperature speeds up the chemical processes of microbial metabolism and the enzymatic degradation of polymers.

Oxygen. The amount of oxygen in the soil distinguishes between aerobic and anaerobic biodegradation by stimulating the growth of microorganisms that can live with or without oxygen. The amount of oxygen in the air is usually inversely related to the amount of water and carbon dioxide in the air.

Sunlight. Microbial development on the soil surface is inhibited by ultraviolet radiation. As a result, biodegradation takes place within a few millimeters under the soil surface with the highest concentration in the first 10 cm. Temperature, organic matter content, aeration, moisture, and oxygen need to be adjusted to meet optimal values for microbial activity. Mulch films are agricultural biodegradable plastic films that are not removed after cropping. They remain on or in the soil where they are diced up and mixed with the most biologically active soil layer by milling and ploughing activities promoting material biodegradation⁹⁷.

RECYCLING OF PLASTICS AND BIOPOLYMER

All possible recycling techniques should be investigated to optimize the environmental advantages of these materials¹⁰⁵. There are four different recycling paths as shown in Fig. 9. They are involved after collection, separation, and purification of plastic garbage¹⁰⁶. They are primary recycling, secondary recycling (mechanical recycling), tertiary recycling (feedstock or chemical recycling), and quaternary recycling (energy recovery)¹⁰⁷.

Because of its simplicity and low cost, primary recycling is the most used method¹⁰⁷. Primary recycling is mechanical recycling which is a closed-loop recycling technology that can only be used on high-quality plastic trash with a known history¹⁰⁵. This method entails reusing things in their original form. The disadvantage of this method is that there is a certain limit on the number of cycles for each material¹⁰⁷. Primary recycling allows the recycled material to be utilized in applications that have the same properties and performance as virgin plastics. It is usually not linked to post-consumer plastics rather to the conversion of uncontaminated plastic waste (e.g., production leftovers) into its original pellet or resin form within the same manufacturing facility. Hence, it does not need sorting and cleaning.

Secondary recycling is the mechanical reprocessing of waste and plastics after consumption. Materials recycled through secondary recycling have lower mechanical properties compared to the mechanical properties of the original product. The lower mechanical properties of secondary materials recycled are attributed to lower material purity and deterioration processes that result during the life of the product. Secondary recycling may be economically inexpensive if the amount of waste is small or/and contaminated. Otherwise, the cost of recycling increases due to the separation and purification steps. Although mechanical recycling is a well-established recycling approach for conventional plastics, it should be used with caution when it comes to biodegradable plastics¹⁰⁵. This is due to the sensitivity of biodegradable materials to heat¹⁰⁸. One of the advantages of this method over chemical recycling is that mechanical recycling is lower in cost processing, less potential for global warming, and less use of non-renewable energy¹⁰⁶.

Chemical recycling is the process of converting polymers into monomers and partially depolymerizing them into oligomers by chemical processes that modify the polymer's chemical structure. The resulting monomers can be utilized to recreate the original or a related polymeric product by new polymerizations. Starting with monomers, oligomers, or combinations of various hydrocarbons, this process is capable of reducing the plastic substance into smaller particles appropriate for use as raw material¹⁰⁷. Chemical recycling provides the following benefits over mechanical recycling: the ability to create valuation products and the potential for a circular polymer manufacturing economy, since recovered

Table 2. Advantages and challenges of the recycling techniques.

Technique	Challenges	Advantages	References
Mechanical recycling	Deterioration of product's properties, pre-treatment	Cost-effective, efficiency, well-known	108,109
Chemical recycling	Mainly limited to condensation polymers	Operational for PET, simple technology	109,110
Energy recovery	Not ecologically acceptable	Generates considerable energy from polymers	111,112

virgin monomers may be repolymerized for an endless number of recycles. However, one of the downsides of this technology is that it is both economically and environmentally expensive¹⁰⁶.

Quaternary recycling is the process of recovering energy from low-grade plastic garbage by incineration^{106,107}. This method is considered to be the best method for reducing the volume of organic matter and producing large energy from polymers¹⁰⁷. However, this method of waste recycling should be used as a last resort¹⁰⁶. This method is not environmentally acceptable because of the resulting pollutants and health risks from toxic materials that are transmitted through the air, such as dioxins¹⁰⁷. Chemical recycling is the only approach permitted under the principles of sustainable development since it results in the production of the monomers from which the polymer is created¹⁰⁷. Table 2 lists the benefits and drawbacks of the various approaches^{108–112}.

NATURAL FIBER REINFORCED COMPOSITES

Natural fiber polymer composite (NFPC) is a composite substance comprising of a matrix of polymer reinforced with natural fibers with good strength properties¹¹³. The natural polymer reinforced composite has taken great interest in many applications. This is because natural fibers offer more and good benefits and qualities than synthetic fibers. Natural fibers have low weight, low cost, renewable and available materials. They are considered less harmful to processing equipment. It also has relatively good mechanical properties like flexural modulus and tensile modulus, as well as an enhanced surface finish of the molded parts composite¹¹⁴, stability during manufacturing, biological degradation, and limited health risks. Natural fiber polymer composites with excellent mechanical strength and stiffness may be made by integrating a light-weight and durable natural fiber into a polymer matrix (thermoset and thermoplastic)¹¹⁵. They are used in many applications which are rapidly increased in various engineering fields. The different types of NFPCs have got great attention in various automotive applications. Also, the other natural fiber composites are found in various applications like building and construction industry, window frame, panels, sports, aerospace, and bicycle frame¹¹⁶.

CHARACTERIZATION OF NATURAL FIBERS, BIODEGRADABLE POLYMERS AND BIOCOSITES

Mechanical properties

Natural fibers have good mechanical strength, modulus of elasticity and they are tough. The composites from natural fibers are served for commercial purpose and become a good substitute of synthetic fibers in various applications¹¹⁷. When comparing natural fibers with glass fibers, the most important features of natural fibers are inexpensive, good mechanical properties due to lower density of them, easy processing and handling, renewable resources, recyclable, and has good acoustic and thermal insulation ability¹¹⁸. Kim et al.¹¹⁹ reported that natural fiber-reinforced composites with high-strain rates absorb more energy than glass-fiber-reinforced composites. Natural fibers have drawbacks such as low strength, variation in quality, high absorption of moisture, treatment temperatures are limited, low durability and less resistance to combustion^{117,118}. In general,

bast fibers are distinguished for having the better properties for structural applications. Among these, flax fibers give the best possible properties between low cost, lightweight, hardness and high strength. Jute is more common. However, it is not as stiff or strong as flax. Overall, natural fibers are characterized by a low density and renewable materials and can be obtained cheaper than glass fiber. Conversely, the strength of natural fibers is much less than synthetic fibers. In situations where stiffness and weight are crucial, natural fibers outperform synthetic fibers. Natural fibers have various unique features, such as biodegradability, renewability, and eco-friendliness, that make them good candidates for several applications¹¹⁸. Mechanical and physical properties of some different natural and synthetic fibers are listed in Table 3^{118,120–124}.

When comparing the mechanical properties of biodegradable polymers and non-biodegradable polymers such as polystyrene (PS), polyethylene (PE), polypropylene (PP), there is a wide values range for the strength of both materials, but conventional polymers are generally stronger than biodegradable polymers. These biodegradable polymers have relatively lower strength than conventional polymers¹²⁵. Among these biodegradable polymers, PLA is considered as one of the most important polymers due to its good mechanical properties that make them suitable for several applications^{125,126}. The mechanical and physical properties of several biodegradable and non-biodegradable polymers are listed in Table 4^{120,127–131}.

Natural fibers are used to enhance mechanical properties of biodegradable polymers. Dong et al.¹³² examined the coir fiber/PLA composite mechanical properties. Figure 10a–d shows that flexural and tensile moduli of PLA biocomposites at the fiber loading from 5 to 20 wt% are moderately improved when compared with neat PLA. It was found that the tensile modulus of untreated coir fiber biocomposites improved to 25.6%, and the flexural modulus was enhanced by 13.4% in the presence of 5 wt% and 20 wt% coir fiber respectively. Increased coir fiber loading to 10 wt% resulted in lower tensile and flexural modulus, which became obvious at 30%. Treated fibers may not improve the modulus of biocomposite particularly at higher fibers loading (20 and 30 wt%). This is because the condition of adhesion between the polymer matrix and the fiber, which has less influence on the tensile modulus of biocomposites than tensile strength.

As shown in Fig. 10a, b, biocomposites have lower flexural and tensile strengths than neat PLA. Biocomposites with treated natural fibers have better strength levels than biocomposites with untreated fibers. Biocomposites have lower elongation at break than PLA at fiber loading 5–20 wt%, as illustrated in Fig. 10c. It was found that the brittleness of PLA composites is higher than that of neat PLA. This is because of the rigid nature of the reinforcement of wet coir fibers with the PLA matrix. When fiber loading reaches 30% in the composite, these composites have a high elongation at break compared to polylactic acid. The increase in elongation at break for PLA biocomposites reinforced with coir fiber up to 30 wt% coir fiber can be attributed to the presence of bundles of excessive non-wet fibers, as shown in Fig. 10d, e. This helps biocomposites deform longitudinally under tensile loading. The alkali treatment of fibers works to remove pectin and lignin from the coir fiber. Thus, increase the cellulose content improves the elasticity and flexibility of coir fibers.

Table 3. Some common natural and synthetic fibers' physical and mechanical.

Fiber	Density (g/cm ³)	Tensile strength (MPa)	Tensile modulus (GPa)	Specific strength (MPa/g.cm ⁻³)	Specific Young's modulus (GPa/g.cm ⁻³)	Elongation (%)	Reference
Natural Fibers							
Flax	1.4–1.5	343–2000	27.6–103	—	45	1.2–3.3	118
Hemp	1.48	550–900	70	372–608	—	0.8–3	120
Ramie	1.5	200–1000	41–130	147–625	27–81	1.5–4	120
Kenaf	1.2	223–1191	11–60	641	10–42.9	1.6–4.3	120
Jute	1.3	393–773	26.5	—	—	1.5–1.8	121
Sisal	1.3–1.5	507–855	9.4–28	362–610	6.7–20	2–2.5	122
Abaca	1.5	400–980	6.2–20	—	9	1–10	118
Coir	1.25	220	6	—	—	15–25	123
Banana	1.35	711–789	4–32.7	444	3.6–27.3	2.4–3.5	120
Bagasse	0.89	350	22	—	—	5.8	123
Bamboo	1.1	500–575	27–40	454	50–67.9	1.9–3.2	120
Cotton	1.5–1.6	287–800	5.5–13	190–530	3.7–8.4	3–10	122
PALF	0.8–1.6	180–1627	1.44–82.5	—	35	1.6–14.5	118
Synthetic fibers							
C-glass	2.52	3310	68.9	—	—	4.8	124
E-glass	2.5–2.59	2000–3500	70–76	—	29	1.8–4.8	118

Table 4. Physical and mechanical properties of some biodegradable polymers and non-biodegradable polymers.

Polymer	Tensile strength (MPa)	Tensile modulus (MPa)	Strain (%)	Density (g/cm ³)	Melting point (°C)	T _g (°C)	Ref
Biodegradable polymers							
PHB	40	1700–3500	3–6	1.25	168–172	5	120,127,128
PLA	28–50	1200–2700	7–9	—	177–180	60–65	120,127
PCL	41	300	900	1.145	60	–60	120,129
PBS	57	—	700	—	115	–32	120
Non-biodegradable polymers							
PC	55–70	2100–3500	200	1.2	155	147	120
LDPE	10–15	200	300–500	0.910–0.925	80–110	—	127,130,131
PP	35–40	1700	150	0.9–0.92	160–176	–10 to –23	120,127
PS	25–69	4000–5000	1–2.5	1.04–1.06	110–135	—	120

Thermal properties

Thermal analysis is used to examine chemical, physical, and structural response as a result of a temperature change. Temperature is a basic state variable that impacts the majority of chemical processes, physical qualities, and structural changes. Thermal analysis may be defined as any scientific or technical assessment of a material in which temperature is manipulated¹³³. This term, however, has long been restricted to approaches using thermogravimetric and calorimetric effects¹³⁴. The basic techniques used in thermal analysis are TGA thermogravimetry analysis (TGA), differential thermal analysis (DTA), and differential scanning calorimetry (DSC). The difference in temperature between a sample and a reference is carried out by differential thermal analysis (DTA). The loss of weight measured by thermogravimetry is measured by thermogravimetry analysis (TGA). The determination of heat flow done by differential scanning calorimetry (DSC)^{53,134}. Thermogravimetry analysis is widely used to describe the thermal stability of natural fiber and polymer composites. The thermal stability of natural fiber and polymer biocomposites has been thoroughly characterized using TG/DTG research. The lignocellulosic fibers are affected by temperature¹³⁴.

Yao et al.¹³⁵ selected ten natural fibers including wood, bamboo, agricultural residues and bast fibers to study thermal decomposition kinetics. The complete thermogravimetric degradation process of natural fibers at a rate of 2 °C/min is shown in Fig. 11a, b. A clear DTG peak with a noticeable shoulder (arrow) is formed due to cellulose thermal breakdown, which is generally the outcome of hemicellulose thermal decomposition in an inert environment. However, since the shoulder peaks of low-temperature hemicelluloses were overlapping with the cellulose main peaks, they were no longer visible in some situations as shown in Fig. 11b. The high-temperature (tails) is usually presented by the degradation of lignin as proven in Fig. 11a, b. Because natural fibers are lignocellulosic materials, the TG and DTG curves for various are very comparable, indicating that natural fibers have similar thermal breakdown characteristics.

Herrera-Kao et al.¹³⁶ studied the thermal degradation of three common biodegradable polyesters (polycaprolactone (PCL), polylactic acid (PLA) and polyhydroxybutyrate (PHB)) by using TGA. Degradation curves for each PCL and PLA showed that they have only one decomposition level. Whereas TGA of PHB displayed two degradation. The TGA mass loss curve and the related derivate curve

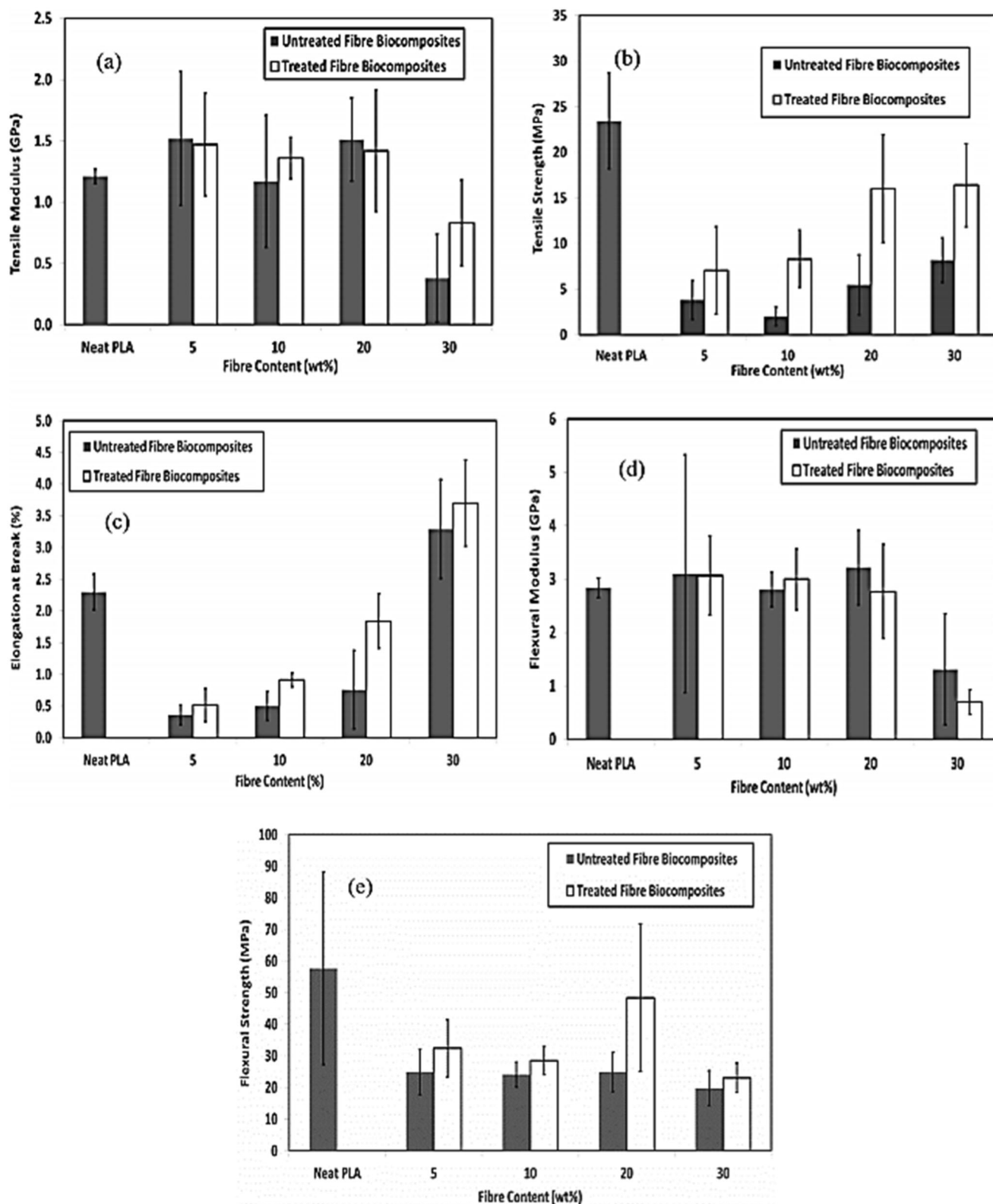


Fig. 10 Mechanical properties of PLA/coir fiber composites. The treated and untreated PLA/coir fiber composites flexural and tensile properties. **a** Tensile modulus, **b** tensile strength, **c** elongation at break, **d** flexural modulus, and **e** flexural strength^{1,32}.

(DTGA) for the examined biopolymers (PHB, PCL, and PLA) are shown in Fig. 11c, d. The maximum rate of breakdown temperature differs according to polymer type. However, both PCL and PLA showed only one phase of mass loss with an initiation temperature

higher than 300° C as displayed in Fig. 11c, d. This temperature was 430° C for PCL and 395° C for PLA. At 303° C and 410° C, PHB showed two different deterioration phases, the first stage is the major transition since it accounts for over 90% of the total mass loss.

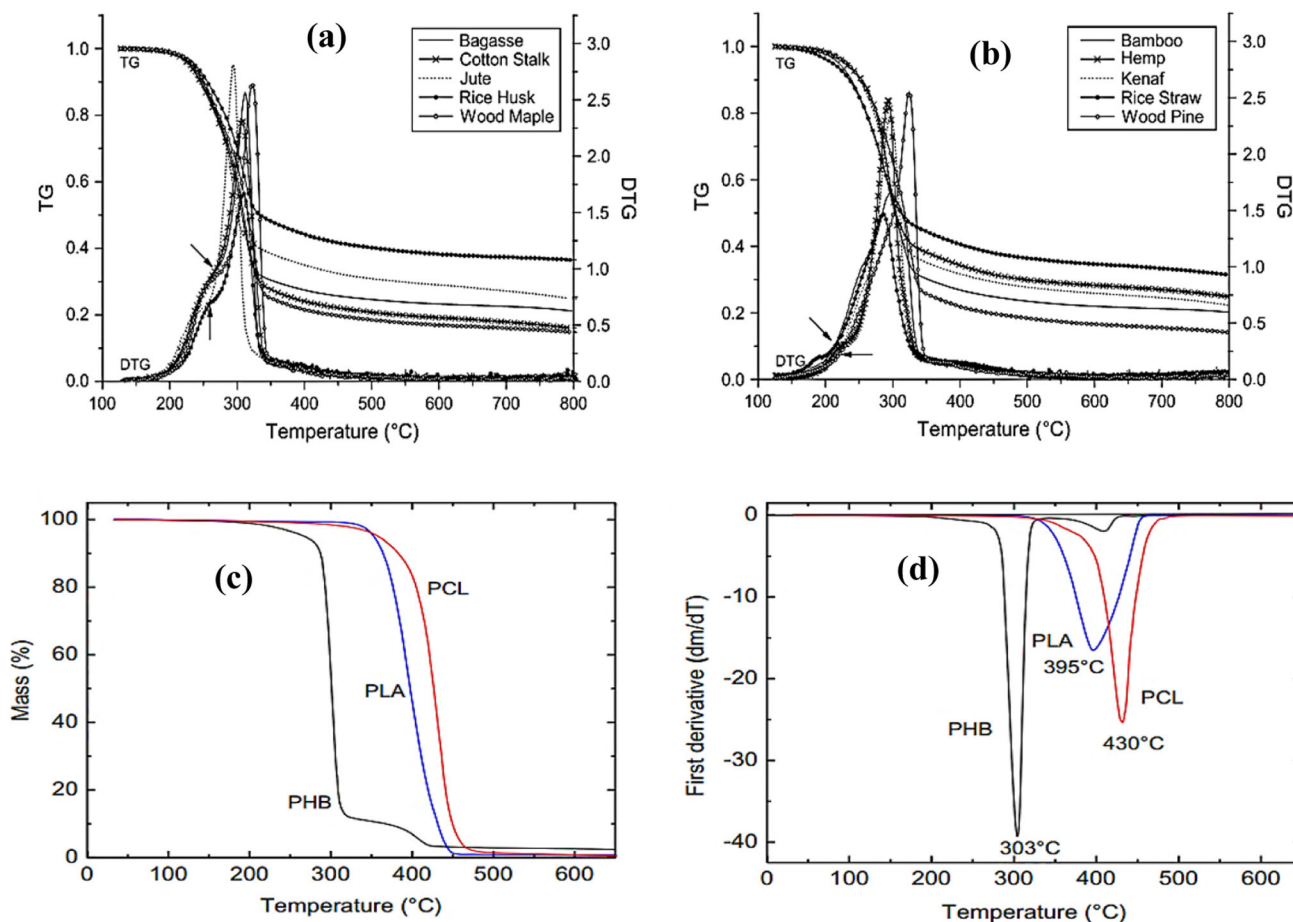


Fig. 11 Thermogravimetric decomposition of natural fibers and biodegradable polymers. **a** Natural fibers with obvious hemicellulose shoulders and **b** not obvious hemicellulose shoulders¹³⁵ **c** TGA for PCL, PLA, and PHB **d** DTGA for PCL, PLA, and PHB¹³⁶.

Dynamic mechanical analysis

Dynamic mechanical analysis (DMA) is used on advanced materials with a rheometer to examine their viscoelastic behavior based on storage modulus (G'), loss modulus (G'') and damping factor ($\tan \Delta$). Also, DMA is applied on the advanced composite material to understand the glass transition temperature (T_g)¹³⁷. Manral et al.¹³⁸ studied the viscoelastic behavior of neat PLA and natural fibers reinforcing PLA by using DMA at different temperature range 30–100 °C. Samples were exposed to twisting load during DMA test.

Storage modulus (G'). The storage modulus of a composite material is a measure of storage energy in terms of elasticity when subjected to cyclic load. The quantity of energy absorbed in step with the cycle during oscillatory load on the material is known as G' ¹³⁹. Figure 12a displays the storage modulus fluctuation with relation to temperature at a steady cycle loading frequency of 1 Hz. Results confirmed that polymer reinforced with natural fibers improved the G' or the stiffness of polymer composites. Storage modulus curves showed that flax/PLA has the highest value by obtaining the value of 2524 MPa when compared to other composites¹³⁸. The high G' value indicates that flax/PLA composites are stiffer. When the temperature increases, the value of G' decreases for all composites. As the temperature increased, the storage modulus of the composite decreased due to a decrease in stiffness. The decrease in G' refers to viscous behavior of polymer in addition to low stored energy¹³⁸.

Loss modulus (G''). The loss modulus (G'') and storage modulus (G') are interrelated. When polymer shows viscous behavior, it has the

potential to dissipate energy in the form of heat due to intermolecular friction among molecules under loading. Further energy could be dissipated by stiff materials and showing higher G'' value. Thus, increasing fiber loading in the presence of biodegradable polymer such as PLA led to increase in friction force and energy dissipation as shown in Fig. 12b 18. Flax fiber reinforcement enhanced the stiffness of neat PLA and resulted in a greater loss modulus. It was reported that flax/PLA composites have higher strength than other natural fiber/PLA biocomposites¹³⁸.

Damping factor ($\tan \Delta$). The ratio between the loss modulus and the storage modulus is defined as $\tan \Delta$. The ratio refers to the damping behavior of the substance with different temperatures. Figure 12c shows the damping behavior variation with respect to temperature. Neat PLA reinforced with the fibers reduces its damping behavior. The $\tan \Delta$ value of neat PLA is found to be higher than all developed composites. It is found that Jute/PLA has a lower damping factor value (0.65). The glass transition temperature (T_g) of material also determined using damping factor curve where the polymer mobility is about to begin. From Fig. 12c, it was found that the damping curve showed that flax/PLA has the highest value of (T_g) of 68.68 °C compared to other biocomposites. This proves that the flax/PLA requires a higher temperature to start molecules mobility¹³⁸.

Acoustic properties

Natural fiber-reinforced materials have advantages such as better acoustic properties. Because of the great potential of

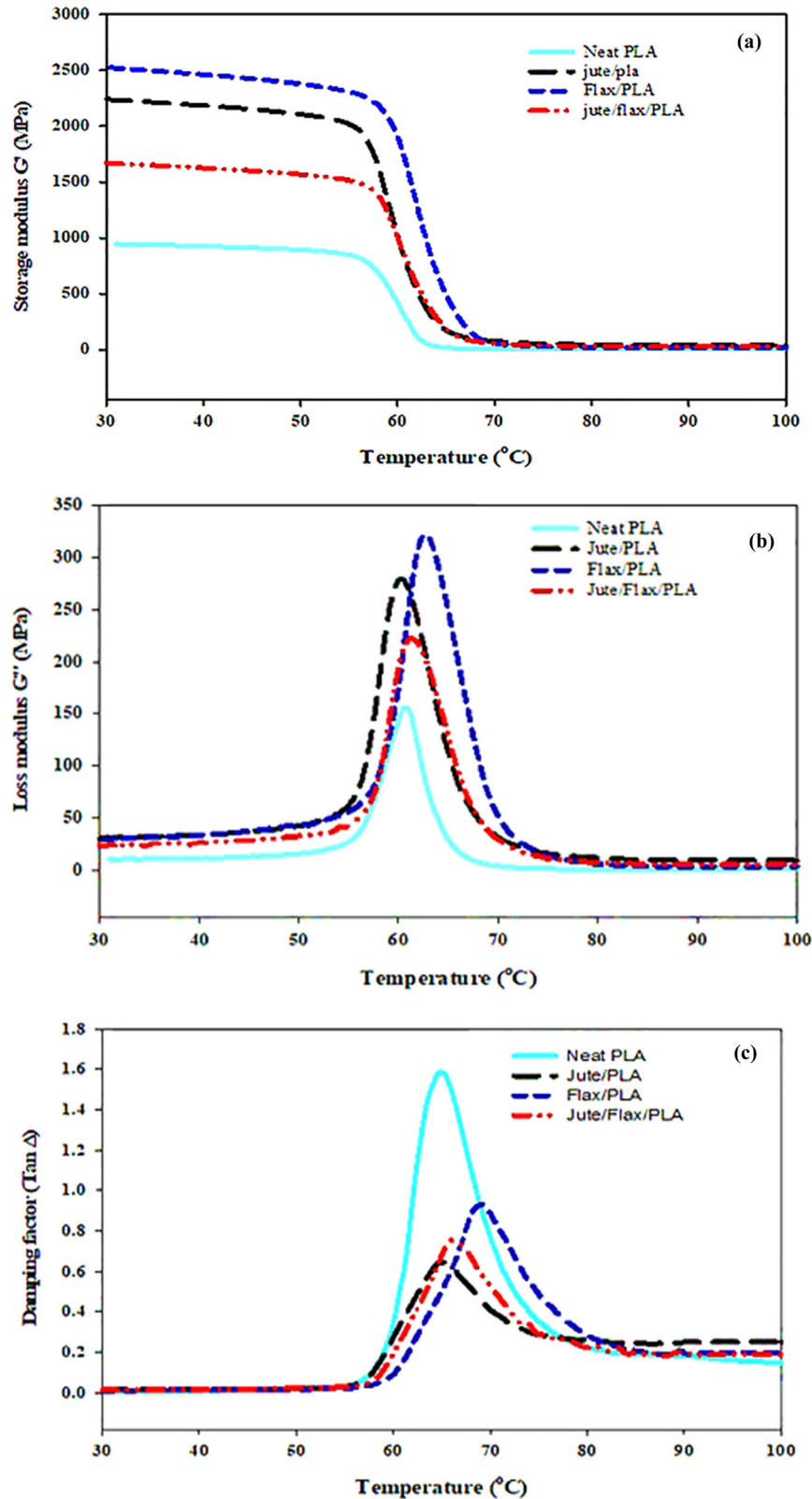


Fig. 12 Dynamic mechanical analysis. **a** Storage modulus. **b** Loss modulus **c** damping factor¹³⁸.

natural fibers to absorb sound, several studies were reported in the literature showing the possibility of using natural fiber rather than synthetic fiber for sound acoustic absorbers. Natural fibers as sound-absorbing materials have an essential consideration in the fabrication of sound-absorbing materials for buildings in

today's environment. Most natural fibers function as porous absorbers when it comes to sound absorption. Porosity appears to be one of the factors that define the absorption properties of any fibrous material. Also, density and thickness of fiber are other factors. The porous material with sound absorption is

Table 5. Some natural fibers acoustical absorption coefficient.

Fibers	Absorption Coefficient	Frequency (Hz)	Fiber properties	Reference
Cotton	0.50	500	Diameter, 13.5 μm	142
Kenaf	0.74	500	Thickness, 50 mm	141
Bagasse	0.13	500	Thickness, 20 mm	143
Ramie	0.60	800	Thickness, 40 mm	144
Corn	0.16	500	Thickness, 20 mm	143
Jute	0.20	500	Diameter, 81.2 μm	142
Flax	0.40	500	Diameter, 21.8 μm	142
Hemp	0.60	500	Thickness, 40 mm	141
Coir	0.84	500	Thickness, 35 mm	141
Sisal	0.1	500	Diameter, 213 μm	142

characterized by a very weak absorption coefficient in the small frequency range but with better acoustic properties in the high-frequency range¹¹⁸.

Natural fibers have good acoustic properties and are utilized in various applications such as vehicles, soundboards, and thermal and acoustic panels¹⁴⁰. Values of acoustic absorption coefficients of selected natural fibers are listed in Table 5^{141–144}. Chin et al.¹⁴⁵ have made a PLA as a polymer matrix reinforced with kenaf fiber. They found that when the kenaf fiber content is 30%, the maximum sound absorption coefficient 0.987 appears at 1521 Hz frequency, and the mechanical properties also improved¹⁴¹.

Surface morphology

Polymers and fiber testing and analysis using surface morphology is one of the areas that need a unique method for creating fully prepared samples. The fibers are tested before and after various processes, including fracture surface. Results are accurate and reliable. Surface analysis can measure dispersed phase at all scales, from micro to nano scale. Scanning electron microscopy provides information on the interaction between matrix and dispersed phase¹⁴⁶.

In order to study the PLA and natural fiber biocomposites, Xia et al.¹⁴⁷ used alkaline treatment, maleic anhydride (MA) grafting treatment, aminopropyl triethoxysilane treatment, and corona discharge treatment to modify the surface of flax fibers^{17,148}. Lignin, hemicelluloses, waxes, and oils were successfully removed from the fiber surface using an alkali treatment. The fiber surface hydrophobic character was increased. Morphological changes resulting from the various treatment methods are shown in Fig. 13a–e¹⁴⁷. It was noticed that the alkaline treatment increased the surface roughness leading to more contact points and an improvement in the mechanical interaction between PLA and fiber as shown in Fig. 13a. Maleic anhydride (MA) and silane act as coupling agents. Figure 13d, e shows the surface of the fibers coated with a layer of MA and silane where increased the interaction between the natural fiber and PLA^{17,147}.

Fracture surface of neat PLA and PLA/flax composites were studied by Xia et al. Figure 14a, b shows that the neat PLA fracture surface is smooth compared with the PLA/flax biocomposites. This refers to brittle fracture. After tensile test, flax fibers were pulled-out from the PLA matrix, leaving a substantial number of fragments¹⁴⁷. Also, the remaining fibers showed smooth and clean surface and does not contain any layers of polymer matrix. Furthermore, there are numerous holes in the PLA matrix/fiber interfaces, resulting from a relatively poor interface strength between the untreated fiber and PLA^{147–149}. The tough fracture morphology is significantly different from the neat PLA and PLA/untreated fiber composite fracture as shown in

Fig. 14c–e. Inside the matrix, fibers are well dispersed, and between the fiber and matrix there are no gaps. The remaining of flax fibers were coated with PLA matrix, indicating that the treated flax fibers and PLA have better interfacial force. The results are in a good agreement with toughness results that are obtained by mechanical testing.

Spectroscopic characterization

The functional groups of polymer composites that might be responsible for the polymer/natural fiber interaction are determined using effective approach of Fourier transform infrared spectroscopy (FTIR)^{150–153}. The FTIR analysis peaks for various untreated natural fibers are listed in Table 6^{53,154–157}. The spectroscopic properties of treated sisal fibers by benzylated were studied. It was that peaks at 1250 cm^{-1} and 1363 cm^{-1} which are assigned to lignin and hemicellulose. They were successfully removed in the treatment process of sisal fiber. Mofokeng et al.¹⁵⁸ studied the interaction of random and oriented PLA/sisal fiber composite using FTIR analysis as shown in Fig. 15. Peaks at 1780–1680 cm^{-1} are assigned to C=O stretches, 3600–3000 cm^{-1} corresponding to O–H stretches, and 1180 cm^{-1} referred to C–O–C stretches. Due to the existence of free OH groups in the fiber, the presence of the O–H bonds in the composites became more apparent and wider as the fiber loading increases. They also found that hydroxyl groups developed a new peak at 1650 cm^{-1} , which is assigned to the absorbed water molecules⁵³.

Using infrared spectra, Xia et al.¹⁴⁵ investigated the untreated flax (UF), alkali-treated fiber, corona-treated fiber, MA grafted fiber, and silane-treated fiber. The samples were analyzed with a spectral resolution of 2 cm^{-1} and a spectrum of 4000 cm^{-1} to 500 cm^{-1} . The FTIR spectra of treated and untreated flax fibers are shown in Fig. 16. The components of flax fiber are cellulose, hemicelluloses, and lignin. These components can be identified using FTIR spectra. C–O and C–O–C looked stretched at 1170 and 1030 cm^{-1} , respectively. The –C=O stretching of aliphatic carboxylic acid and ketone groups in the cellulose chain showed the peak at 1654 cm^{-1} . The absorption of –C–H in flax fiber cellulose showed a peak at around 2900 cm^{-1} . The largest peak was the –OH stretching of D-pyran glucose in the cellulose chain at 3400 cm^{-1} . It was noticed that the band at 1735 cm^{-1} (–C=O) stretching of aldehyde carbonyl is assigned to lignin and hemicellulose. Figure 16b shows that the band at 1735 cm^{-1} was vanished in the alkali-treated fiber indicating that the lignin and hemicellulose of flax fiber were removed by NaOH solution treatment. Due to the alkali treatment, the band at 1735 cm^{-1} was not visible in the MA-grafted treated fiber, corona-treated fiber, and silane-treated fiber spectra, as shown in Fig. 16c–e¹⁴⁷.

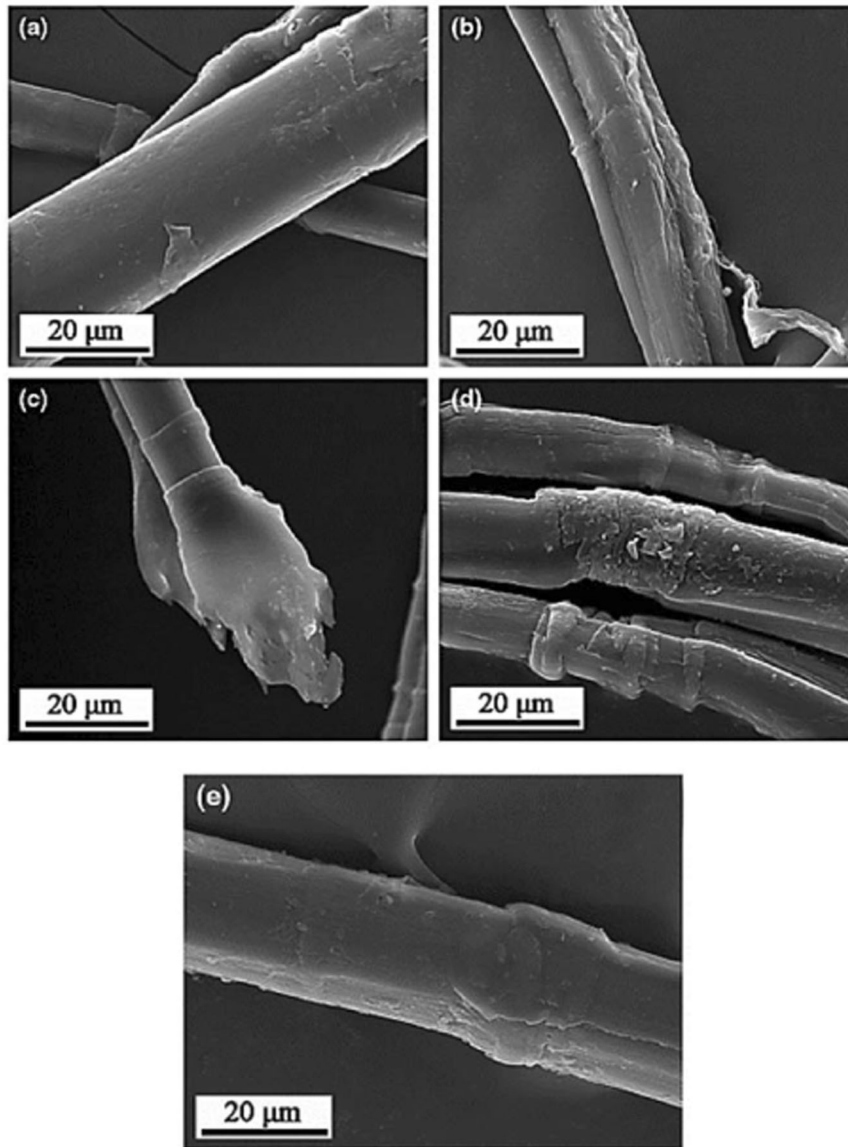


Fig. 13 Morphological changes using various treatment methods. SEM pictures of **a** untreated fiber, **b** alkali-treated fiber, **c** coronatreated fiber, **d** MA-grafted fiber, and **e** silane-treated fiber¹⁴⁷.

THEORETICAL MODELING OF MECHANICAL PROPERTIES OF BIOCOMPOSITES

Several studies were conducted to explore if natural fiber composites are applicable for structural and non-structural applications. The tensile properties of synthesized composites are frequently measured using a variety of mechanical tests. Young's modulus and strength of discontinuous and continuous fiber composites were reported using micromechanical models. The fundamental rule of mixtures is often used to calculate Young's modulus and strength of reinforced composites by unidirectional continuous fiber in longitudinal and transverse loading. However, rule of Mixtures model is no longer obeyed if the fibers are discontinuous. The tensile characteristics of discontinuous fiber composites are influenced by the fiber's length, dispersibility, orientation, and the interfacial force between the matrix and natural fiber. The orientation of the fibers and stress transfer are strongly correlated with tensile properties of polymer composites. Several models were developed to estimate composite tensile characteristics while

accounting for the aforementioned conditions. Nevertheless, there is a wide range of results¹⁵⁹.

The rule of Mixtures model was modified for fibers parallel to the loading direction by including Kelly and Tyson. Various ways for predicting young's modulus and strength of non-continuous fiber composites were applied. It is based totally on the distribution of stress of the dispersed fibers in the plastic matrix. It is supposed that shear forces on the fiber-matrix interface shift the applied load to the fibers, as defined in the Cox model. Shear stress at the interface is expected to keep constant until the fiber strain equals the matrix strain where the average fiber stress and the shear stress at the interface will be zero. In this model, the critical fiber length (L_c) is defined as the average fibers' length relative to the fiber length, which is known as the shortest possible length to carry the load. It could be obtained by experimentation or micromechanics, like a model suggested by Bowyer-Bader. The fiber fragmentation test and a pull-out test are the most popular techniques used to determine the critical fiber length, which could be obtained

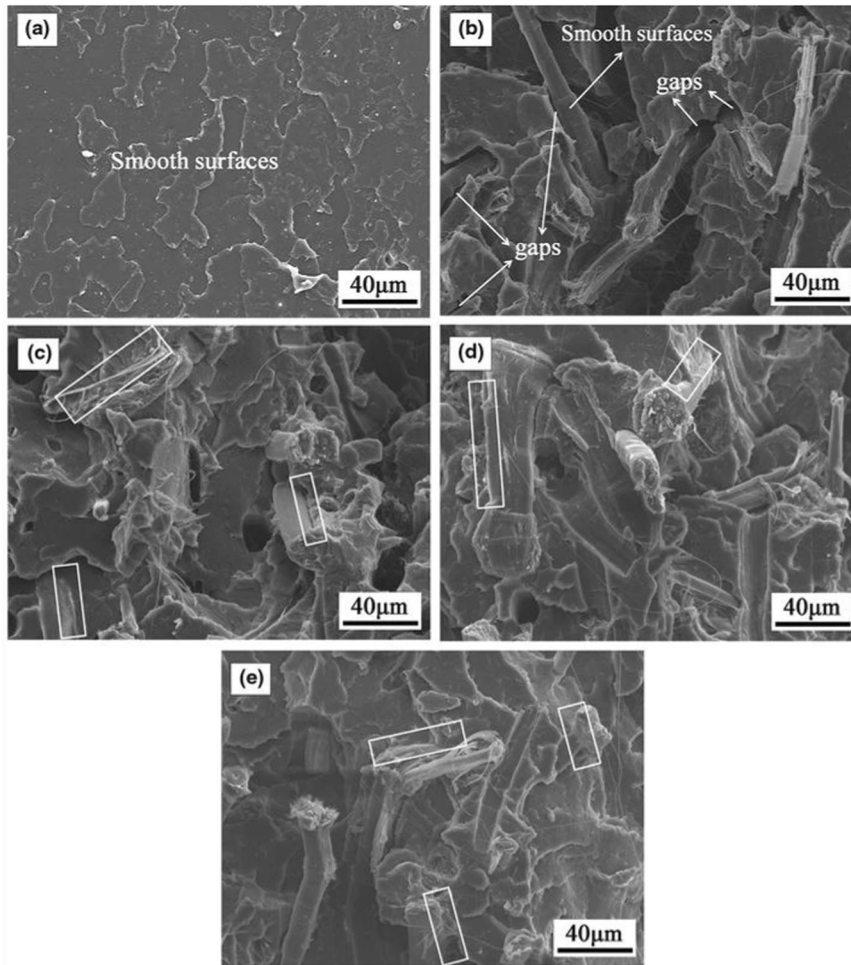


Fig. 14 Surface morphology of PLA fracture surface. SEM pictures of impact fracture surface of **a** neat PLA, **b** PLA/untreated, **c** PLA/alkali-treated, **d** PLA/MA-grafted, and **e** PLA/silane-treated composites¹⁴⁷.

Table 6. FTIR peaks of different untreated natural fibers (in cm^{-1}).

Fiber	-OH stretching	C=O stretching	C-H stretching	C-O stretching acetyl group in hemicellulose	C-vibration of lignin and	CH ₂ Symmetric bonding	Bending vibration of C-H and C-O groups of aromatic rings in polysaccharides	Ref
Sisal	3434	1742	—	1249	—	2923–2896	—	152
Kenaf	3397	—	2902–2918	—	—	1416	1036	53
Banana	3600–3100	—	2910	—	—	1416	1036	53
Jute	3427	1739	2922	1440–1530	—	—	—	153
Hemp	3410	1732	2916	1247	—	1425	—	154
Coir	3330	1725	2910	1235	—	1420	—	155

using below equation:

$$L_c = \frac{\sigma_f D}{2\tau} \quad (13)$$

Where D is the average fiber diameter and τ is the interfacial shear strength.

It is clear that the decrease in critical fiber length makes an improvement to the interfacial shear strength. If the fiber length is equal to L_c (critical fiber), the ultimate fiber tensile stress is achieved only at the center of the fiber. If the fiber length exceeds L_c , reinforcement of fiber becomes more efficient and ultimate tensile stress within the fibers can be achieved along a greater

length. And finally, if the fiber length is less than (subcritical fiber), it would ultimately pull-out from the matrix.

Efendy et al.¹⁵⁹ investigated the tensile properties of a PLA composite reinforced with harakeke and hemp mats of fiber. They compared experimental results with predicted values using different models. Results are displayed in Fig. 17a, b. For Hirsch, it is found that, when the fitting parameter $x=0.13$, it gave a better correlation with the experimental strengths. When the fiber content increased up to 30 wt%, Modified Cox models were shown to demonstrate logical agreement with experimental tensile strength as shown in Fig. 17a, b. The fitted, predicted, and experimental young's modulus of the two fiber polymer

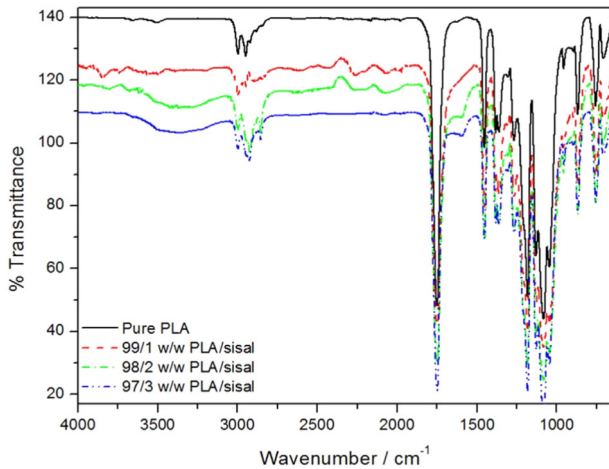


Fig. 15 FTIR analysis of PLA/sisal fiber composites. FTIR spectra of random-oriented sisal fiber/PLA composite manufactured by a technique of injection molding¹⁵⁸.

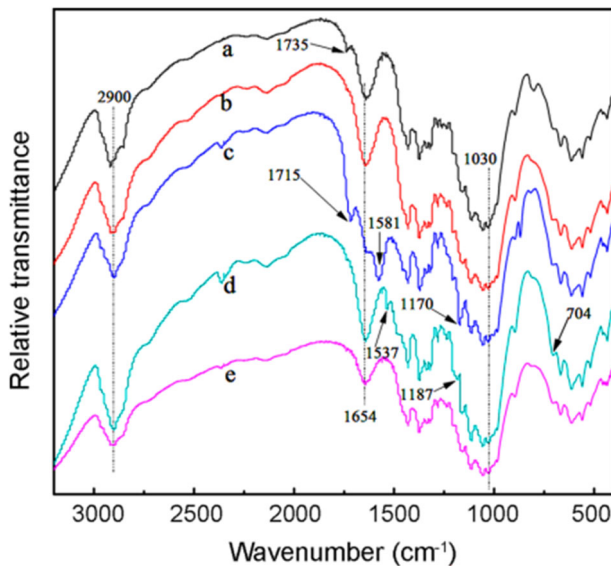


Fig. 16 FTIR spectra of (a) untreated fiber, (b) alkali-treated fiber, (c) MA grafted fiber, (d) corona-treated fiber, and (e) silane-treated fiber¹⁴⁷.

composites with different fiber loading are shown in Fig. 17c, d. The modified Halpin-Tsai and Cox models, which take into consideration the interconnection integrity between the polymer matrix and the fibers are the best models for predicting Young's modulus at fiber loading up to 25% by weight¹⁵⁹.

FUTURE PERSPECTIVE OF BIODEGRADABLE POLYMER

The need to develop sustainable alternatives to oil-based polymers has sparked a lot of study. Biodegradable polymers are also appealing materials for biomedical applications due to their unique physicochemical, biological, and degrading features. They help in reducing greenhouse gas emissions and fossil resource depletion^{160,161}. Currently, biopolymers are being used and produced in small quantities around the world. They are used mainly for food packaging and bioplastic industries. In the future, studies need to be conducted to improve the polymeric properties of these biodegradable materials by combining different types of polymers in varying ratios to see how they affect the physical

and chemical properties of biomaterials. The medical industry will benefit greatly since biopolymer-based bio-implants and drug carriers' agents are now being developed. More advancements in the future might result in a revolution in medical implants in terms of economic effectiveness.

The United Nations General Assembly approved 17 sustainable development goals (SDGs) to be implemented by 2030. The SDGs objectives reflect a comprehensive approach to achieving a society that is both healthy and sustainable. Bioplastic production establishes a foundation for environmentally friendly product development. Most of sustainable development goals are made possible by the plastic industry and polymers, which are widespread and adaptable materials¹⁶². Materials science helps to achieve a variety of goals, according to report maps of the most recent sustainability research and activities namely; SDG 2: zero hunger, SDG 3: good health and well-being, SDG 6: clean water and sanitation, SDG 7: affordable and clean energy, SDG (9): industry, innovation, and infrastructure, SDG 11: sustainable cities and communities, SDG 12: responsible consumption and production, SDG 13: climate action, and SDG 14: life below water are among the 17 SDGs¹⁶². The carbon footprint of both the raw and final product is minimized since bioplastics are made from monomers derived from agricultural waste. Through its sourcing, manufacture, marketing, consumption, and removal, the recyclable characteristic of bioplastics assures that the commodity is commercially, socially, and ecologically acceptable. As a result, consumers' lives are enhanced, and the community as whole benefits¹⁶³. Energy, waste disposal, and operating costs may all be reduced by adopting sustainable materials¹⁶⁴. It is clear that biopolymers will open up new possibilities in the effort to establish a better environment devoid of hazardous substances and products.

FORENSIC ENGINEERING OF ADVANCED BIODEGRADABLE POLYMER SYSTEMS

The study of failure in polymer products is the focus of traditional forensic polymer engineering. This branch of science deals with the breaking of plastic items, as well as any other reason for the product's inability to function or meet its standards¹⁶⁵. Ex-post analyses of classic polymeric materials or their thermoplastic composites have been the focus of most forensic polymer engineering case studies¹⁶⁶.

One of the major drawbacks of biodegradable polymers derived from renewable sources is their rapid rate of degradation due to the hydrophilic nature and, in certain circumstances, low mechanical properties, especially in water environment. Despite their drawbacks, biodegradable polymers have a number of benefits due to the fact that they are made from a variety of plant components¹⁶⁷. Bio-polyesters have recently gained prominence as a result of their biodegradability and possible medical uses. Specific aspects such as biodegradation methods, biocompatibility, processing conditions, and prospective uses in medicine, environmental protection, and agro-chemistry have received a lot of attention. The bio-safety of such advanced biodegradable polymers as well as the nano-safety of their composite continues to be negligible. This new perspective focuses on predicting, evaluating and indicating possible issues associated with advanced polymer usage. The approach created by forensic engineering of advanced polymeric materials (FEAPM) would be used to analyze associations between polymeric materials' structures, characteristics, and behaviors before, during, and after actual applications. This should aid in the development of new sophisticated polymeric materials, reducing the number of product flaws that occur during manufacture and use. The economic and societal effect of FEAMP investigations is projected to be significant, allowing for the identification of safe advanced polymers in a future sustainable society. Failure modes are well

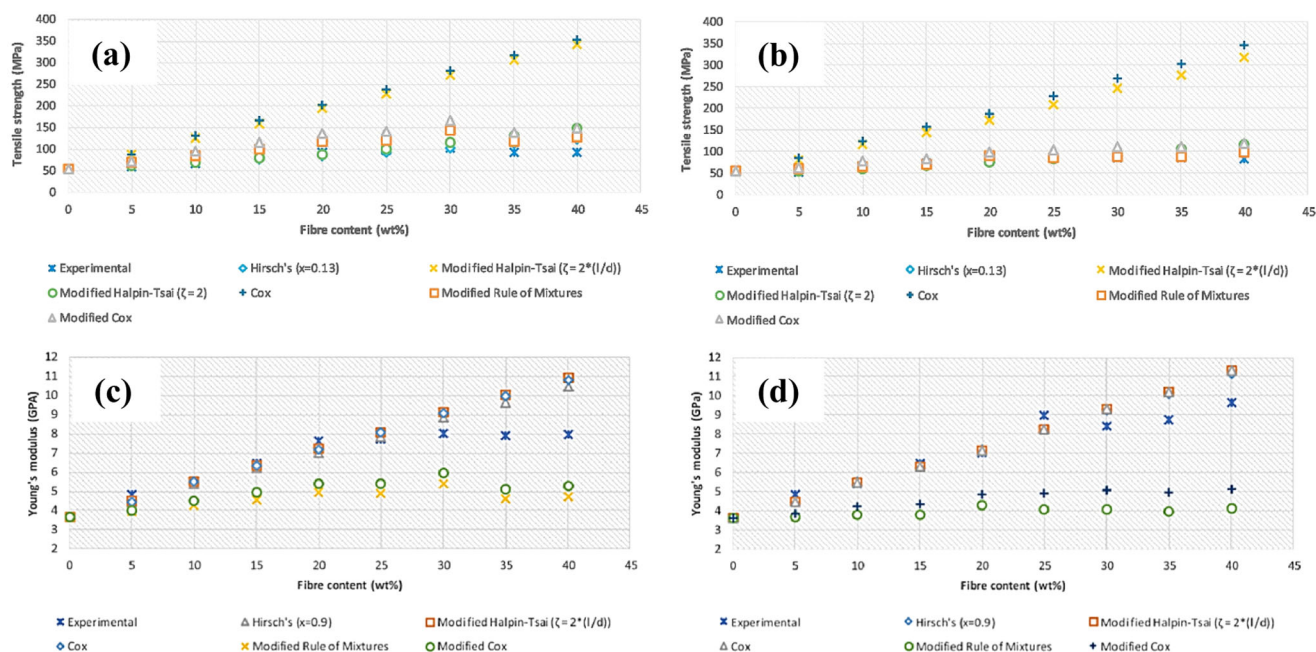


Fig. 17 Mechanical properties of PLA/natural fiber biocomposites. Variation of actual and predicted **a** tensile strength of PLA/harakeke Biocomposites, **b** tensile strength of PLA/hemp fiber biocomposites, **c** Young's moduli of PLA/harakeke biocomposites, and **d** Young's moduli of PLA/hemp fiber¹⁵⁹.

reported for several applications and laboratories scales. These studies do not only study failures but also work on reducing the possibility of their causes.

Mechanical failure

Mechanical failure is easy to be realized if the loads and dimensions of a product are known at specific time over its life¹⁶⁸. Fracture is one of the most prevalent failure modes¹⁶⁹. It could take place under various conditions such as overload, creep, stress relaxation, fatigue, and wear^{170–172}.

Loading patterns. Tension, compression, bending, shear, torsion, and impact are all examples of how the polymer is exposed to stress. Loading patterns are commonly result from one or more factors.

Load path. When evaluating loading patterns, it is notably that forces must be coupled in a chain via a product to produce a path. When there are many distinct components along a path, the load takes on diverse shapes as it travels along with it. The compressive local load on the flat surface would decrease from the point of contact until it reaches zero. To counteract the compressive force, a tension force would be generated to the surface.

Stress concentrations. They are localized form deviations caused by the product's stress lines being pressed together and amplified. Cracks in or at the edges of bodies, holes in flat sheets, voids inside materials, corners and fillets, change in the profile of shafts, and screw threads are just a few simple examples of stress concentration.

Chemical attack

If mechanical failure occurred in a complex process, a chemical attack on a product would begin as a complicated problem. The fact that materials may be attacked in variable ways reflects the complex process. Polymers might be exposed to chemicals in service, beginning with the atmospheric air. When the product is

loaded or has in-built forces that can be alleviated through fracture development, the situation becomes much more significant. Although hydrolysis is a prominent degradation process for one type of polymer, oxidation may be the most prevalent kind of assault in its many varied incarnations.

Oxidation. The main active element is oxygen (O_2) in the air (around 21% by volume), although there are numerous additional chemicals that breakdown polymers as well (degraded many thermoplastics). Ozone gas is one of the most powerful oxidizing agents. Because oxidizing chemicals are ubiquitous in our environment, oxidation must always be considered a potential attack vector.

Hydrolysis. Alkali and acids break down polymers by hydrolysis, where the polymer chain is broken by the cleavage of functional groups that bind the chains together. Dramatic decrease in the molecular weight is found as the chains are reduced in length at each stage. Water have a negative impact on most polymers due to its widespread presence. Hydrolysis step of polymers are direct or indirect process. When high temperatures enhance the risks of hydrolysis during processing or shaping, the issue appears. Because most processing temperatures exceed 100°C , any liquid water in the feedstock will evaporate and cause undesirable bubbles in the final product.

Ultraviolet radiation. Exposure to UV radiation from the sun is a typical cause of failure in many polymers. Polymers containing double bonds or other absorbing functional groups are the most vulnerable to UV deterioration. UV damage can cause cracking and the creation of a damaged layer on the surface, as well as the bleaching of pigments, a process known as whitening.

Stress corrosion cracking

Stress corrosion cracking (SCC) is a common failure mechanism caused by a chemical assault on polymers. Microcracks can be caused by trace concentrations of strong chemicals, which further increase slowly over applied stresses or by an issue known as

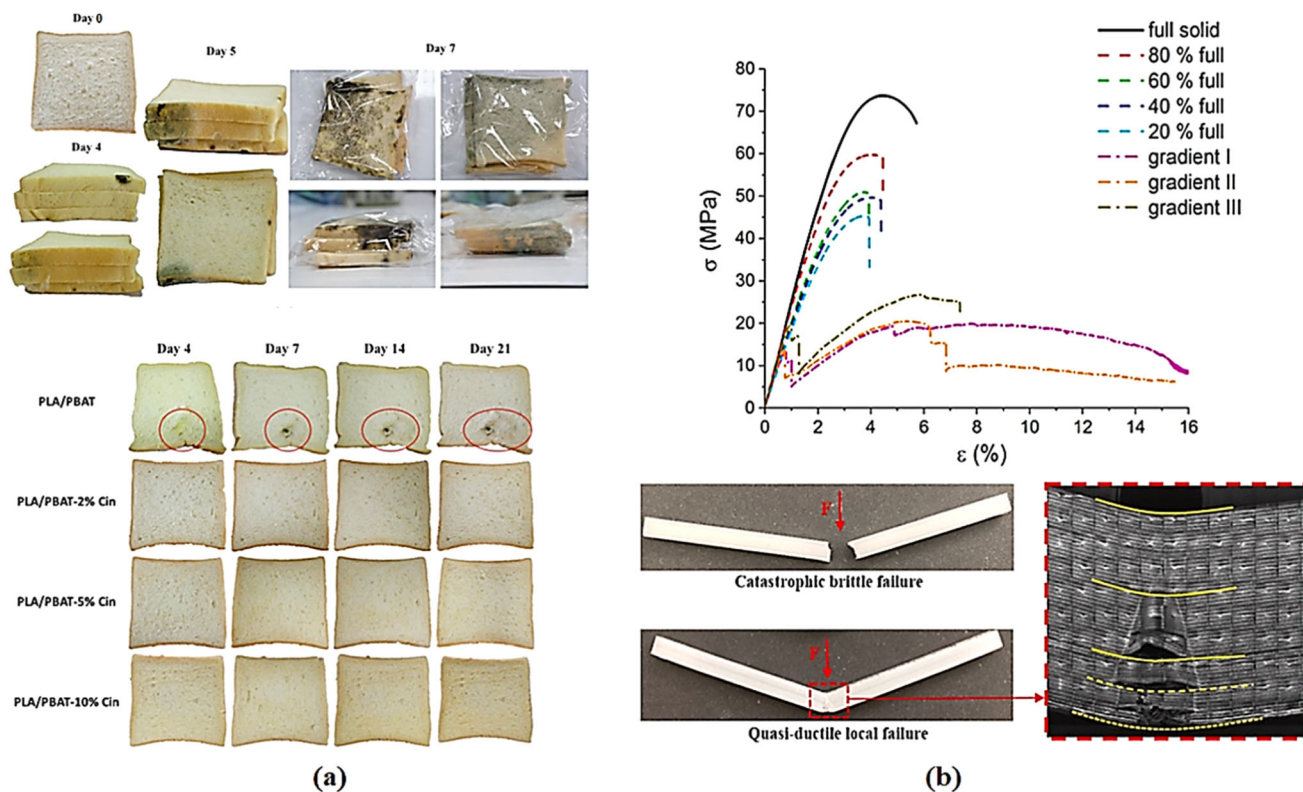


Fig. 18 Forensic engineering of biodegradable polymers. **a** Appearance of packaged bread stored in (1) PP and (2) PLA and PBAT blend films with different concentrations of trans-cinnamaldehyde, and **b** stress–strain curves of PLA samples with various porosity and 3D printed design deformed in three-point bending (top). Comparison of failure modes of specimens^{174,175}.

frozen-in strain. Other types of SCC in polymers include attacks by oxygen, ozone, and chlorine, which need a low stress or strain threshold for crack formation¹⁷³.

Environmental stress cracking (ESC)

Among the most common prevalent causes of abrupt brittle breaking of thermoplastic polymers is environmental stress cracking (ESC). The chemical composition of the polymer, bonding, crystallinity, surface roughness, molar mass, and residual stress are all variables that impact the rate of ESC. There is no long-term chemical alteration, although the symptoms are similar to those of SCC. Srisa et al.¹⁷⁴ studied antifungal bioplastic films, developed based on PLA and poly(butylene adipate-co-terephthalate) (PBAT) blends with incorporated trans-cinnamaldehyde. Mold was discovered on bread that were preserved in ordinary PP films. According to PP's forensic engineering, additional storage resulted in the development of fungal growth over the entire loaf on day 4 as shown in Fig. 18, 1 a. Mold mycelium expansion was successfully reduced by PLA/PBAT films and microbiological growth was not detected in any of the films containing trans-cinnamaldehyde (Fig. 18a). This is because the bacteria were successfully suppressed by large levels of trans-cinnamaldehyde release.

Juraj Svatik et al.¹⁷⁵ studied the mechanical strength and toughness of neat PLA and PLA bamboo biocomposites. It was found that the solid specimen and uniform porosity specimens were brittle and was broken up catastrophically. However, the gradient porosity specimens were quasi-ductile with no catastrophic breakdown and much greater strain at break as shown in Fig. 18b.

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon request.

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A.S.: Conceptualization, Methodology, Writing—original draft, Writing—review & editing. F.A.: Methodology, supervision, review & editing. A.A.A.H.: Writing—review & editing. M.B.: Conceptualization, Supervision, Writing—review & editing.

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COMPETING INTERESTS

The authors declare no competing interests.

ADDITIONAL INFORMATION

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