REVIEW

JOURNAL OF **Applied Polymer** **WILEY**
SCIENCE

A review of research and application of polylactic acid composites

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Abstract

Polylactic acid (PLA) is a thermoplastic polyester that has received widespread attention for its environmentally friendly origin and excellent performance, and its potential to address the current problems of severe white pollution and scarcity of petroleum resources. This article focuses on the synthesis, modification, degradation and application of PLA. The main focus is on the modification of PLA with different environmentally friendly materials (natural organic materials, biodegradable polymers, inorganic minerals) in blends for defects such as the brittleness of PLA. In addition, the applications of PLA composites in the fields of construction, medical, packaging and oil and water separation are also introduced, which hopefully will provide some help to the promotion of PLA.

KEYWORDS
application, degradation, modification, PLA, synthesis

1 | INTRODUCTION

With good low thermal conductivity, insulation, flexibility and corrosion resistance, plastics have been widely used in industry, agriculture, medicine, construction and other fields. Representative plastics include polyethylene, polypropylene, polyethylene terephthalate, polyvinyl chloride, polystyrene and many others. As the use of plastics becomes more widespread, the demand for plastics is gradually increasing, which leads to an increase in the production of plastics year after year. It was reported that plastics production increased by about 13 million tons between 2015 and 2016.[1] 2018 world plastics production was 359 million tons, 11 million tonnes more than in 2017.[2] 2019 world plastics production will be about 368 million tons.[3]

The large amount of plastic produced will certainly bring about a huge pressure on the ecological environment, the pressure being mainly on the pollution of waste plastics. Current disposal and management systems for waste plastics are still inadequate, and under current plastic production and management practices, it is expected that 53 million metric tons of plastic will be released into the natural environment each year by
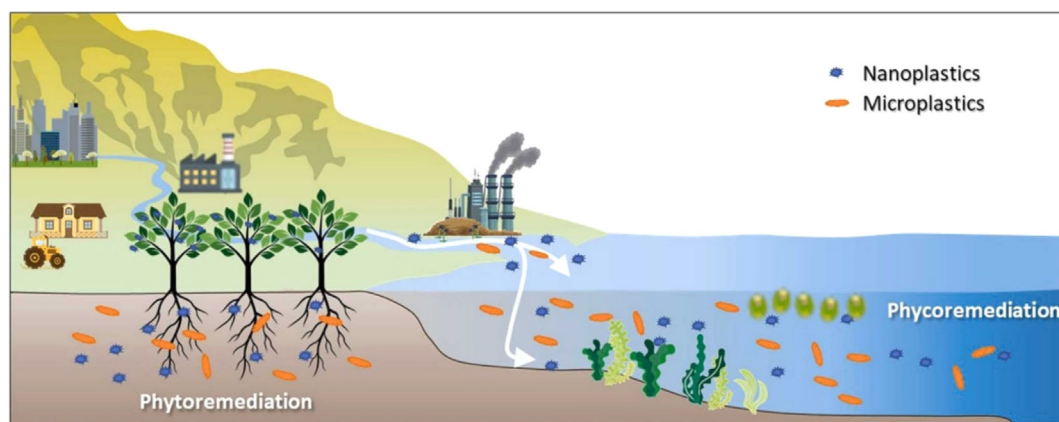
FIGURE 1 Interaction of microplastics with algae and plants. Reproduced with permission.[10] Copyright 2022, Elsevier [Color figure can be viewed at wileyonlinelibrary.com]

2030.[4] Waste plastics are already present in various areas such as rivers, lakes, oceans, the atmosphere and land. Plastic pollution poses a huge risk to the environment and human health. Apart from conventional plastic pollution, ordinary plastics may break down into smaller pieces in the natural environment due to ultraviolet radiation, physical forces and hydrolysis,[5,6] forming microplastics (<5 nm) and nanoplastics (<1 μm),[7] which are often more harmful. Microplastics have been integrated into the ecosystem cycle, and the interrelationship between microplastics already and plants is shown in Figure 1. Microplastics in the soil and water environment can contaminate each other through wastewater reuse, soil groundwater, and so on, and then cause harm through the shift of lower trophic level organisms to higher trophic level organisms, which eventually reach humans.[8] In addition, the surface of microplastics formed through weathering has more cracks and pores, which will also bring a large flow of oxygen-containing functional groups, such as carboxyl, hydroxyl and carbon groups, which largely enhance the adsorption capacity of microplastics.[9] And microplastics may form complex contamination if they adsorb some pollutants such as polycyclic aromatic hydrocarbons, polysulphonic acid mucopolysaccharides, etc.

There are currently four main ways to dispose of waste plastics, namely (1) sorting and recycling: old plastics are added to prepare for production, such as drums, tubs and building materials[11]; (2) chemical recycling: decomposing the recovered raw material monomers for reuse, but with high investment, technical and environmental requirements[12]; (3) incineration: for electricity and heat generation and (4) landfill: which increases the toxicity of the soil in the area near the landfill.[13,14]
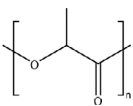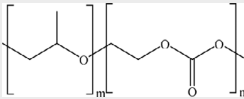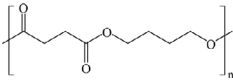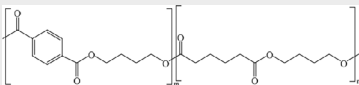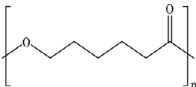
The problem of waste plastic disposal, however, has not been well addressed due to high disposal costs for more environmentally friendly disposal methods, lack of environmental awareness, and problems such as incineration and other rapid disposal methods that produce greenhouse gases and harmful emissions.[15] In summary, although plastics bring us a lot of convenience, there is an urgent need to address the problems of overuse of plastics production, improper disposal of waste plastics and the ubiquity of microplastics.

The development and use of biodegradable plastics may be one of the solutions to these problems. Biodegradable plastics are plastics that meet the needs of their use and can be degraded under natural conditions to be ecologically sound after use. Currently, biodegradable plastics are used in food packaging and tableware, foam packaging, compost bags, agricultural and horticultural planting products, paper coating and other packaging, among many other applications. Degradable plastics can be divided into two categories: plastics prepared from bio-based raw materials and biodegradable plastics prepared from fossil fuels.[16] The structure and properties of several biodegradable plastics are shown in Table 1.
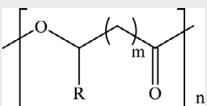
To achieve the goal of zero emissions by 2050, a significant increase in the development and use of renewable energy sources and a reduction in the use of fossil fuels in production and manufacturing processes will be required.[25] Among the above-mentioned degradable plastics, PLA has attracted the attention of many researchers due to the environmental friendliness of its source and its excellent properties. PLA is a thermoplastic aliphatic polyester material derived mainly from renewable materials such as maize, cassava, sugar cane and sugar beet. The end products of degradation are CO_2 and water (H_2O), which can be reabsorbed and used by plants, constituting a certain cycle,[26] as shown in the diagram in Figure 2. However, PLA has a number of performance

TABLE 1 Raw materials, structures and properties of several biodegradable plastics

| Designation	Main ingredients	Structure	Performance		Ref
Polylactic acid (PLA)	Lactic acid (LA)		Advantages	High transparency, high modulus/ strength, biocompatibility, biodegradability	[17]
			Limitations	Poor toughness, brittleness, high price, poor ductility	
Polypropylene carbonate (PPC)	Carbon dioxide and propylene oxide		Advantages	carbon dioxide(CO_2) fixation, good biocompatibility, fully degradable	[18]
			Limitations	Higher cost, poor tensile properties, poor thermal stability	
Poly (butylene succinate) (PBS)	Butanedioic acid and 1,4-butanediol		Advantages	Low cost, excellent mechanical properties, good flesh acidity, easy to process and shape	[19–21]
			Limitations	Excessive crystallinity, difficult to dye, poor heat resistance, easy to shrink at high temperatures	
Polycaprolactone-butylene terephthalate (PBAT)	Butylene glycol adipate and Butylene glycol terephthalate		Advantages	Good ductility, high elongation at break, good heat resistance and impact capacity, excellent biodegradability, good biocompatibility	[22]
			Limitations	Poor gas barrier properties, low crystallinity, low strength, low modulus	
Polycaprolactone (PCL)	Caprolactone		Advantages	Non-toxic, good biocompatibility, biodegradability, good biocompatibility, low cost, certain drug carrying capacity, shape memory	[23]
			Limitations	Insufficient mechanical strength	

(Continues)

TABLE 1 (Continued)

Designation	Main ingredients	Structure	Performance		Ref
Polyhydroxyalkanoates (PHA)	Hydroxy fatty acids		Advantages	Excellent thermal processability, biodegradability and biocompatibility	[24]
			Limitations	Expensive, poor thermoplasticity, narrow processing temperature range, poor thermal stability, high brittleness, unstable production quality	
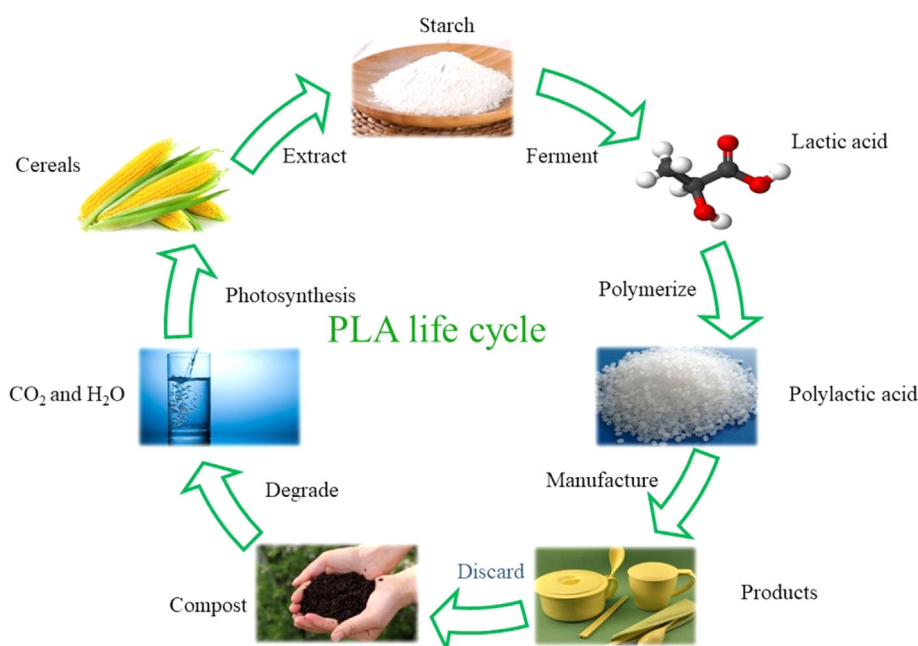
FIGURE 2 Schematic diagram of polylactic acid recycling [Color figure can be viewed at wileyonlinelibrary.com]

L-(+)-Lactic acid   D-(+)-Lactic acid
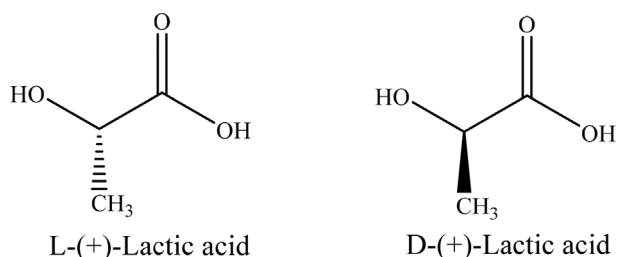
FIGURE 3 Two structures of lactic acid. Reproduced with permission.[35] Copyright 2016, Elsevier

drawbacks, such as excessive brittleness, which limit its large-scale application.

Based on a series of problems such as pollution caused by plastics, this paper focuses on the environmentally friendly material PLA and introduces the synthesis, degradation and modification of PLA as well as the application of PLA-based composites in the hope of providing guidance on the promotion of PLA.

2 | SYNTHESIS AND DEGRADATION OF PLA

PLA is currently a very widely used degradable plastic in the world, accounting for 25% of degradable plastics.[27] The main raw material for PLA is starch, a biodegradable thermoplastic aliphatic polyester obtained through the saccharification, fermentation and ring-opening polymerization of starch from renewable materials such as maize, cassava, sugar cane and sugar beet. PLA is not only green in origin, it also requires 25%–55% less fossil energy in its production than synthetic petroleum-based polymers.[28]
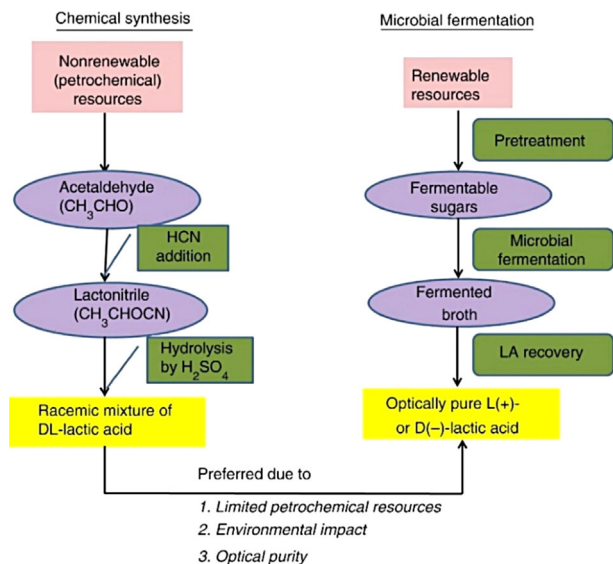
FIGURE 4 Methodology for producing LA. Reproduced with permission.[34] Copyright 2019, John Wiley and Sons [Color figure can be viewed at wileyonlinelibrary.com]

PLA offers many advantages such as complete degradability (PLA breaks down to CO_2 and H_2O in the natural environment); good biocompatibility (PLA is hydrolyzed in living organisms to LA, which can be further broken down by enzymes in the body); high transparency, modulus and strength comparable to many petroleum-based plastics. In terms of flame retardancy, pure PLA has an ultimate oxygen index of only 18%, a vertical combustion test rating of no grade, and is extremely flammable in air with severe molten droplets[29]; in terms of mechanical properties, PLA has a Young's modulus of about 4% at room temperature, a tensile strength between 50 and 70 MPa, an elongation at break of about 4%, and an impact strength close to 2.5 kJ/m².[30] The side group of PLA is methyl (—CH_3) which is spatially shielded and hydrophobic, which makes PLA highly water resistant.[31,32] However, the brittleness of PLA, poor toughness, low softening point, low melt strength and low crystallization rate during production, the above defects seriously limit its promotion and application. This section provides specific information on the synthesis and degradation of PLA based on its environmental friendliness.

2.1 | Synthesis of PLA

PLA is a long straight chain consisting of repeating LA, and there are two structures of LA monomers used to produce PLA: left-lactic acid (L-LA) and dextrolactic acid (D-LA),[33] the structure is shown in Figure 3. LA can be obtained either using chemical methods of manufacture or by biological fermentation, and the two processes for producing LA are shown in Figure 4. The chemical synthesis method is in the problem of producing a racemic mixture during the preparation process. The advantage of fermentation-produced LA is that it can produce optically pure L-LA or D-LA, and that the optical strength largely influences the physical properties of PLA.[34] Additionally, under acidic fermentation conditions, the microbial production of LA facilitates a reduction in production costs.

There are three main ways of preparing PLA, namely direct polymerization, azeotropic dehydration condensation and ring-opening polymerization. Direct polymerization (one-step method) is simple to operate, using LA direct polymerization, with a short process and little investment in equipment. However, this method requires strict control of the vacuum and temperature of the production process, as well as continuous removal of water as a by-product of the production process, and the molecular weight of the prepared PLA is not high. This method also requires a larger reactor to recover the solvent.[34] Mitsui Chemicals Co. used an azeotropic dehydration polycondensation method to prepare PLA, which resulted in an increase in the molecular weight of the PLA prepared. Dissociated water, which is clearly difficult to remove during production, can be removed by azeotropic distillation. However, this method involves the use of organic solvents, which may put pressure on the ecosystem and defeat the original purpose of PLA development, and therefore has not received much attention.[36] PLA can also be prepared by the polymerization of dimeric propylenes of LA by ring-opening polymerization (two-step process),[37] usually using a stannous octanoate based catalyst. This method results in a higher molecular weight PLA with a narrow molecular distribution and easier removal of the by-product water. But this method is an expensive raw material, resulting in high prices for PLA on the market. The different synthesis methods for PLA are shown in Figure 5.

2.2 | Degradation of PLA

Degradable polymers can generally be broken down into CO_2, methane (CH_4), H_2O, inorganic compounds or biomass materials. As a biodegradable polymer, degradability makes PLA ideal for environmental protection and can lead to degradation of PLA-based products during normal use, causing a range of problems. Therefore, the study of PLA degradation is of some practical significance for the application and promotion of PLA.

The most important environments for PLA degradation are soil, compost and seawater environments. In this
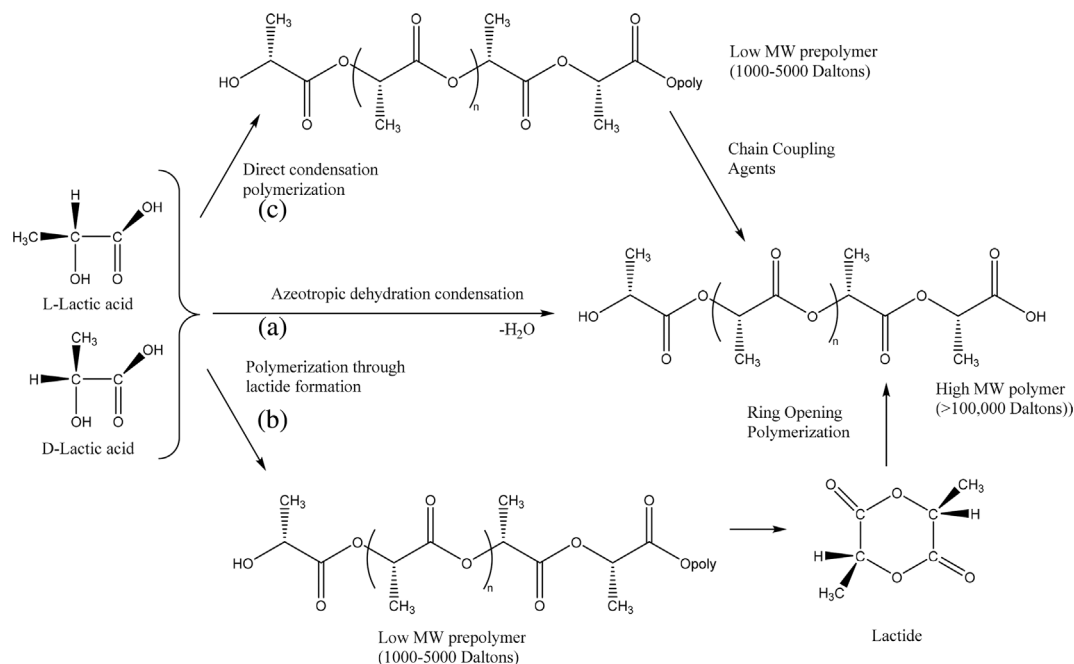
FIGURE 5 Polymerization route for the production of polylactic acid. Reproduced with permission.[38] Copyright 2010, Elsevier

environment, the main degradation of PLA occurs through water, biodegradation and enzymatic degradation.[39] Biodegradation is mainly based on the gradual degradation of PLA by various enzymes secreted by organisms and water.

Under natural and composting conditions, the carbon–oxygen bonds in the PLA backbone are first hydrolyzed and then further degraded to CO_2 and H_2O by the action of enzymes.[40] Due to the rigidity of the main chain of PLA molecular chain and the presence of methyl groups on the side chains, it is not easily decomposed by natural strains, which leads to its slow degradation rate.[41] Related experiments have shown that PLA can be biodegraded in 180 days under composting conditions.[42]
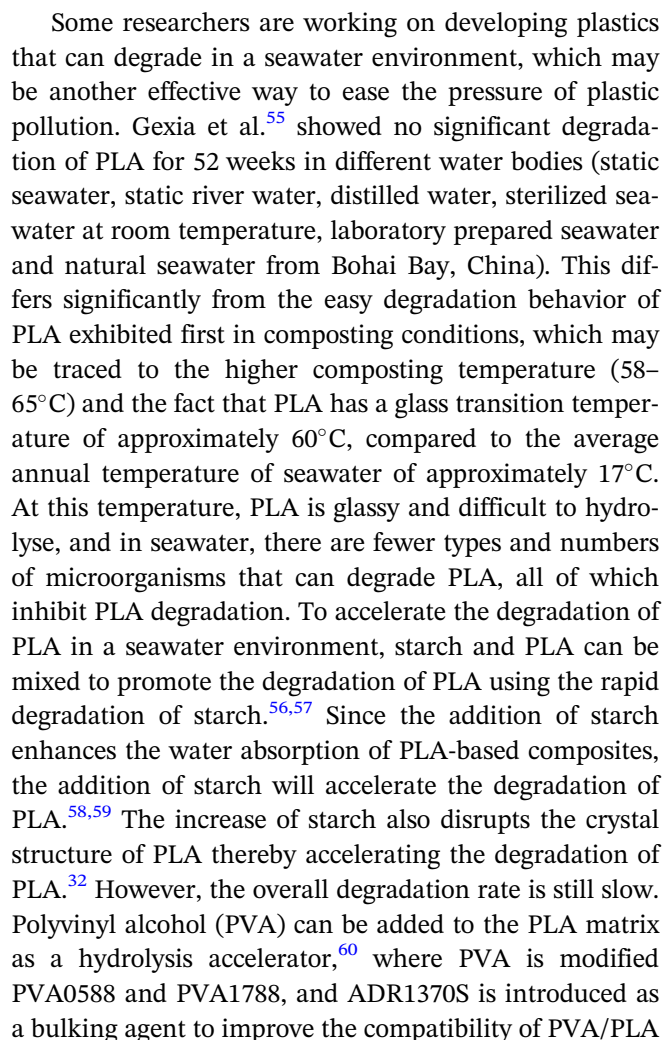
The process of biodegradation relies heavily on enzymes secreted by microorganisms. The process of biodegradation is roughly as follows: first, microorganisms secrete PLA depolymerase, then the depolymerase affects the lactone bonds of PLA to produce oligomers, dimers and monomers, and finally these low molecular weight compounds are taken up by microorganisms and broken down by intercellular enzymes into CO_2, H_2O and CH_4.[43] For example, the extracellular protease secreted by Bacillus megaterium, the extracellular protease attacks the ester bond on the polymer molecular chain and the PLA molecular chain breaks and degrades into small molecular polymers such as oligomers and LA, thus accelerating the degradation of PLA.[44] Microorganisms that can completely degrade PLA include Actinomyces,

Pseudonococcus, Pseudomonas, Bacillus and the fungus Tritirachium album.

Although PLA is a fatty polyester, the majority of known PLA degrading enzymes are protein-based. Enzymes commonly used to degrade PLA include proteinase K, streptomyces and pineapple protease.[45] There have been many attempts to enzymatically degrade PLA, including kutinases, lipases, proteases and esterases.[46–48] Hegyesi et al.[49] used lipase from Candida rugosa and proteinase K from Tritirachium album to try to degrade PLA/cellulose nanocrystal nanocomposites and found that proteinase K was very effective in catalyzing the degradation of PLA. But the use of enzymes to catalyze the degradation of PLA creates the problem of pH change. During the reaction, the LA formed by the degradation of PLA lowers the pH of the degradation medium (down to almost 4). When the pH is too low, this can affect the catalytic efficiency of the enzyme and can even lead to inactivity.

Microbial activity varies in different environments, which directly affects the amount of PLA-degrading enzymes produced by microorganisms. Pseudomonas geniculata WS3 is a PLA-degrading bacterium that can be isolated from sanitary landfills and it has been shown that nutrients or enzyme inducers can be used to enhance the degradable activity and enzyme production of the bacteria.[50] Pseudomonas geniculata WS3 bacteria can degrade PLA using proteases and PLA degrading enzymes, and such bacteria can also adhere to PLA surfaces to accelerate PLA degradation.[51] Combinations of

FIGURE 6 Polylactic acid thermal cracking mechanism. Adapted with permission.[61] Copyright 2022, Zhejiang University of Technology Magazine

bacteria and nutrient amendments can accelerate the degradation of PLA in the soil or composting process.[52] The inoculation of Pseudomonas geniculata WS3, fecal extracts or waste water extracts as a substrate in a basal salt medium and the addition of soy ketones as a nitrogen source stimulates microbial activity in the soil to accelerate the biodegradation of PLA. Soy ketones promote the production of enzymes (proteases) by microorganisms, which further activate the proteolytic activity of microbial ammonification to release ammonia. Ammonia and water react to form ammonium ions (NH^{4+}) and hydroxide ions (OH^-). NH^{4+} and OH^- attack the electron-deficient centre at the ester linkage, causing random chain cleavage of PLA,[53] which is involved in accelerating the biodegradation of PLA. NH^{4+} and OH^- also lead to an increase in pH and the onset of alkaline hydrolysis, whereby indirect hydrolysis of ammonia occurs. The degradation of PLA under submerged and soil-exposed burial conditions was effectively promoted under the combined use of Pseudomonas geniculata WS3 and soy ketones.

The combination of current production and applications has led to the inevitable presence of plastics in the oceans, and not only does the litter in the oceans put significant pressure on marine ecosystems, but marine plastic pollution is far more difficult to manage than land-based plastic pollution. The main mode of degradation of PLA in the marine environment is hydrolysis, which involves chain breakage of the amorphous part of the ester bond followed by catalysis by carboxyl groups. Hydrolytic degradation depends on the amount of water absorbed, the diffusion coefficient of the polymer inner chain fragments, the solubility of the degradation products and other conditions.[54]

Some researchers are working on developing plastics that can degrade in a seawater environment, which may be another effective way to ease the pressure of plastic pollution. Gexia et al.[55] showed no significant degradation of PLA for 52 weeks in different water bodies (static seawater, static river water, distilled water, sterilized seawater at room temperature, laboratory prepared seawater and natural seawater from Bohai Bay, China). This differs significantly from the easy degradation behavior of PLA exhibited first in composting conditions, which may be traced to the higher composting temperature (58–65°C) and the fact that PLA has a glass transition temperature of approximately 60°C, compared to the average annual temperature of seawater of approximately 17°C. At this temperature, PLA is glassy and difficult to hydrolyse, and in seawater, there are fewer types and numbers of microorganisms that can degrade PLA, all of which inhibit PLA degradation. To accelerate the degradation of PLA in a seawater environment, starch and PLA can be mixed to promote the degradation of PLA using the rapid degradation of starch.[56,57] Since the addition of starch enhances the water absorption of PLA-based composites, the addition of starch will accelerate the degradation of PLA.[58,59] The increase of starch also disrupts the crystal structure of PLA thereby accelerating the degradation of PLA.[32] However, the overall degradation rate is still slow. Polyvinyl alcohol (PVA) can be added to the PLA matrix as a hydrolysis accelerator,[60] where PVA is modified PVA0588 and PVA1788, and ADR1370S is introduced as a bulking agent to improve the compatibility of PVA/PLA

composites.PVA addition undergoes dissolution and swelling, which facilitates microbial contact with PLA in water and thus accelerates the degradation of PLA.

PLA can be thermally degraded during thermal processing, the thermal decomposition temperature range of PLA is 320–420°C. The thermal cracking mechanism of PLA is random fracture and intramolecular ester exchange to produce propylene glycol esters, cyclic oligomers and linear oligomers. Thermal cracking allows the recovery of propyleneglycerides at an optimum recovery temperature of 350–400°C.[61] The thermal cracking process is shown in Figure 6.

To investigate the process of PLA abiotic degradation factors (composting temperature) and the effect of algal biomass on PLA degradation, Kalita et al.[27] used a twin-screw extruder and sheet casting unit to make continuous films of PLA and PLA/algal biomass composites, and then prepared PLA and PLA/algal foam sheets using CO_2 as the foaming agent. Under non-biodegradable conditions (composting temperature of $58 \pm 2°C$), the chain breaks that occur in PLA-based biomass composites do not produce enough oligomers to provide microbial assimilation, resulting in slow degradation. In contrast, with the addition of 5 wt% de-oiled algal biomass to the PLA-based composite, the algal biomass acts as a nitrogen source to promote microbial growth, resulting in accelerated degradation.

Qin et al.[62] investigated the degradation of PLA under conventional weathering conditions and the adsorption behavior between PLA and tetracycline (TC). Under conventional weathering conditions, the surface of PLA will be cracked with cavities after weathering, the oxygen-containing functional groups will increase and the contact angle will be reduced to 77.94%, which is easier for PLA to adsorb and bind to TC, and the adsorption capacity of PLA to TC is enhanced by about 3.5 times. In contrast, under seawater weathering conditions, the change in PLA surface is not significant.

Compounding of PLA with other materials can also have an effect on the degradation of PLA. Jute fibers can significantly enhance the mechanical properties of PLA and retain the biodegradability of the composite, which can also accelerate the degradation of PLA. Chen et al.[63] in order to investigate the mechanism of enzymatic degradation of PLA/jute fiber composites. PLA/jute fiber composites were subjected to enzyme solutions containing proteinase K, cellulase, ligninase and pectinase for 35 days for degradation experiments. It was concluded that the addition of jute accelerated the degradation of PLA and that three stages of enzymatic degradation of jute/PLA composites were obtained. The first stage degrades mainly the amorphous PLA molecules; the second stage degrades mainly the fiber-matrix interface and the PLA crystalline region; in the third stage, the jute fibers begin to degrade and the PLA starts to disintegrate after the enzymatic breakdown of the crystalline region.

Hemp fibers are cheap, low density and non-abrasive to processing equipment, and have been increasingly used as a reinforcing material in the automotive, furniture and construction industries.[64,65] In order to investigate the adverse degradation effects of hemp fibers on PLA properties, Mazzanti et al.[66] prepared PLA/hemp composites using a melt blending method (twin-screw extruder) to investigate the effect of a small amount of short hemp fiber filling on the PLA degradation process. Fibers that have been dried and mercerised (including alkali treatment) were subjected to degradation analysis in seawater at room temperature. Overall, the degradation of PLA was accelerated by small amounts of hemp fibers. It was also found that drying the hemp fibers prior to composite preparation reduced the degradation of PLA by the fibers.

Lima et al.[67] prepared PLA/mango seed pomace/clay composites that degraded at a higher rate in water than the samples did in soil due to the coupling of water and microbial degradation. Comparing the degradation rates of PLA/peel/peel samples and PLA/peel, the PLA-peel-peel samples degraded faster as both mango peel and mango kernel contributed to the degradation.

By studying degradation methods such as water degradation, enzymatic degradation, biodegradation and thermal cracking, it is possible to understand the degradation conditions of PLA in order to prevent possible degradation of PLA products during production and use and to enhance their service life. It is also possible to accelerate the degradation of PLA materials at waste and accelerate their life cycle cycle cycle.

3 | PLA BLEND MODIFICATION

The main focus of research into the modification of a material is to overcome its deficiencies or enhance its properties (including its basic mechanical properties, thermal properties and functionalities such as flame retardancy, antibacterial, etc.) by physical or chemical means. PLA commonly used modification methods are blending treatment, adding nucleation agents, thermal annealing treatment, etc. In combination with the high price of PLA, among the many modification methods, material blending can be mixed with a certain amount of cheaper materials, which can reduce the price of PLA products to some extent. In order to maintain the advantages of PLA degradability, the choice of environmentally friendly materials for the co-blending modification of PLA is a feasible method. This section focuses on three
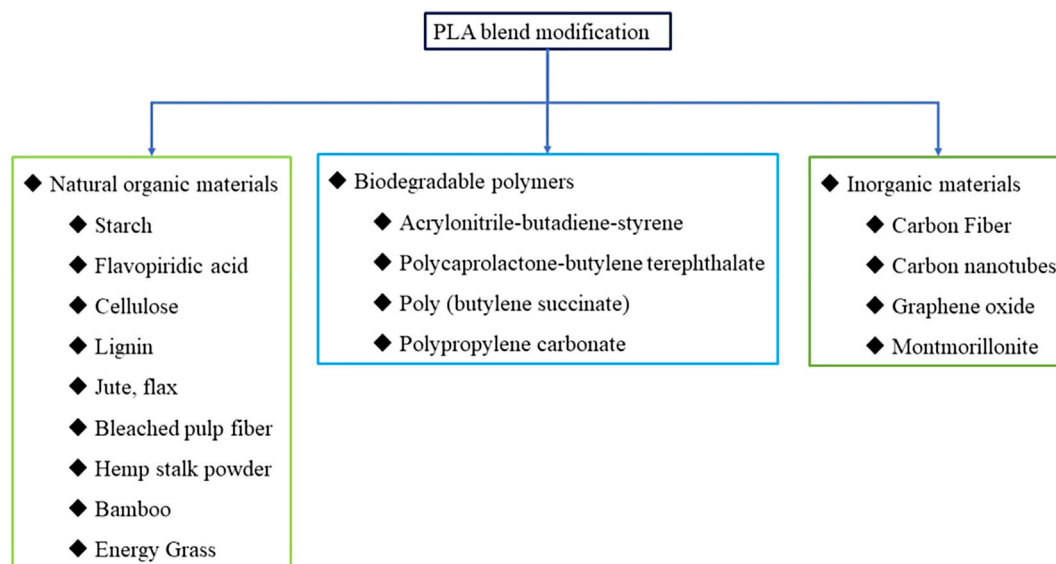
FIGURE 7 Polylactic acid blended materials [Color figure can be viewed at wileyonlinelibrary.com]

aspects of co-blending modification, including natural materials, degradable polymers and inorganic materials. The main materials presented are shown in Figure 7.

3.1 | Natural organic materials

Natural organic materials are themselves renewable resources, often derived from plant tissue, and can enhance the performance of composite materials to a large extent. Natural organic materials can eventually be degraded by micro-organisms and do not put pressure on the environment.

Starch is an ideal biomass filler, widely found in the roots, stems, fruits and other tissues of various plants, and has the advantages of being widely available, inexpensive and completely biodegradable.[68] Therefore, some researchers have also taken starch to improve the properties of PLA. However, starch has a crystalline structure and the melting temperature is greater than its thermal decomposition temperature, so it is usually necessary to break the crystal structure of the starch to make it thermoplastic,[69] thus obtaining thermoplastic starch (TPS). On the other hand, starch is a hydrophilic material, while PLA is somewhat hydrophobic, so directly making the two materials into composites, the mechanical properties of the blends will instead be worse and more brittle.[70] Therefore, the focus of research on PLA/starch blends is on the mechanical properties and compatibility of the composites.

Oligo(lactic acid)-grafted starch (OLA-g-starch) is composed of oligo(lactic acid) and starch and is compatible with PLA and TPS respectively, therefore OLA-g-starch can be used as an environmentally friendly compatibiliser for PLA/TPS blends. By adding OLA-g-starch, the TPS phase exhibited a smaller size and better distribution, suggesting that OLA-g-starch improved the compatibility between the TPS dispersed phase and the PLA matrix. The ductility, water resistance, gas barrier, melt flow and thermal stability of PLA/TPS were further enhanced.[71] Li et al.[72] used chemically modified grafting of furfuryl glycidyl ether (FGE) to successfully graft FGE onto starch molecules using maleic anhydride bridging to produce TPS with excellent melt processing properties. The contact angle was increased from about 30° to 65°, which showed excellent hydrophobic properties, thus significantly improving the interfacial compatibility of the PLA/FGE grafted starch (FGE-g-St) composite. The tensile strength, flexural strength and elongation at break of the PLA/FGE-g-St composite were significantly improved compared to the PLA/starch composite.

The amount of TPS added will have an effect on the composite. When the content of TPS is too high, TPS will appear to agglomerate[73] and the dispersion is reduced, so when doing modification treatments on PLA/TPS blends, a third component can be added as a plasticizer (glycerol, formamide, lipids, etc.), inorganic filler (glass fiber, calcium carbonate, silica, etc.), block or graft blends (PCL-g-starch), the functional additives such as reactants or crosslinkers (isonitrile esters).[74]

Flavopiridic acid (FA) is a green, natural polymer with excellent properties of interfacial activity, hydrophilicity, cation exchange, complexation and adsorption and dispersion. Due to the presence of oxygen-containing functional groups in FA, grafting of PLA with FA is possible. Duan et al.[40] grafted FA onto the backbone of PLA

and then prepared PLA/PLA-FA blends using a melt blending method. It was confirmed by XPS spectroscopy that the O=C/O—C of PLA-FA was significantly increased compared to that of pure FA. The hydroxyl group in PLA was dehydrated and condensed by the carboxyl group in FA, and the relative contents of —CH$_2$ and —CH$_3$ in the composites were also reduced, confirming the successful grafting of FA into PLA. PLA-FA enhanced the crystallization rate of PLA composites, and also played a better role in plasticization, enhancing the the toughness of the composites was enhanced.

Lignocellulose is a common renewable resource whose components are mainly cellulose, hemicellulose and lignin. Cellulose is a major component of some natural fibers and agricultural by-products. Cellulose typically enhances the barrier properties, thermal insulation and mechanical properties of composites.[75] Cellulose nanowhiskers (CNW) have good mechanical properties and can be used to reinforce biopolymers. However, CNW has a polar surface and is difficult to disperse uniformly in non-polar media. Poor compatibility can lead to non-uniform dispersion of natural fibers in the PLA matrix and poor mechanical properties. Therefore, the use of additives such as chemical coupling agents or compatibilisers and hybridization with stronger materials may be an effective means of reinforcing PLA.[76] Moran et al.[77] prepared PLA/CNW nanocomposites by twin-screw extrusion and injection molding techniques using glycerin polyglycidyl ether (PL) as a plasticizer/volumizing agent, after dispersing PLA in PL. PL reduces the brittleness of PLA and prevents the agglomeration of CNW. The mechanical properties of PLA/CNW nanocomposites are better than those of pure PLA. The mechanical properties of the PLA/CNW nanocomposites are better than those of pure PLA, and the best mechanical properties are achieved at lower CNW content.

Microcrystalline cellulose (MCC) is obtained by hydrolysis of natural cellulose and contains a large number of hydroxyl groups on its surface.[78] The addition of MCC to PLA matrix can improve the mechanical properties of the composites on the one hand, and on the other hand, MCC can participate in the carbon formation process of the composites and improve the flame retardant properties of the composites. However, the large amount of hydroxyl groups on the surface of MCC leads to its poor compatibility with PLA and prone to agglomeration.[79] To this end, Ren et al.[80] used the silane coupling agent 3-aminopropyltriethoxysilane (KH550) to modify MCC in order to improve the flame retardant properties of PLA, and formed a compound flame retardant with modified MCC and ammonium polyphosphate (APP), and melt-blended the compound flame retardant with PLA, which effectively improved the thermal stability and flame retardancy of PLA composites. At a modified APP addition of 3% and an APP addition of 7%, the PLA based composite achieved an limiting oxygen index of 35.3% and passed V-0 in UL-94 testing. However, the tensile and impact strengths of PLA composites are reduced compared to pure PLA.

The aromatic structure of lignin, which results in a high charring capacity (50%–60%), also means that lignin may improve the flame retardancy of thermoplastics.[81,82] Podkościelna et al.[83] prepared PLA/lignin composites using a melt blending method as a means of improving the thermal properties and flammability of PLA-based composites. The effect of natural flame retardant additives on the flammability of PLA-based composites was assessed by adding the natural flame retardant silicon dioxide (SiO$_2$). Analysis of the thermal properties revealed that the initial thermal decomposition temperature (T_{onest}) of pure PLA was 131.2°C. After the addition of kraft lignin, sulphonated lignin and SiO$_2$, the T_{onest} was increased to 136.2, 151.5 and 150.3°C respectively, reducing the flammability of the composites.

Jute and flax are plant fibers that are easily extracted from plants. Among them, jute has good mechanical and thermal properties. Flax fibers have low density, high strength, high toughness and high stiffness. PLA/jute, PLA/linen and PLA/jute/linen composites were prepared by a hot pressing process. Comparing the tensile strength of the three composites, the tensile strength of PLA/linen composite is the best and the tensile strength of PLA/jute composite is inferior among the three composites as the tensile strength of flax is higher than that of jute. In terms of impact resistance, the PLA/jute/linen composite had the best impact resistance.[84]

In addition to plant fibers, to further reduce costs, PLA-based wood-plastic composites can be prepared by crushing wood, straw, bamboo, and so on with PLA. Wood plastic composites (WPC) are made by blending wood flour, thermoplastics and other process additives through a base, injection or compression molding process, using PLA as the plastic matrix in the WPC, which can make the prepared WPC completely degradable. With the shortage of wood resources, the wood flour in WPCs can also be derived from sawdust, straw flour, and so on from agroforestry residues.[85,86]

The current research on wood fiber/PLA composites does not include high fiber additions to fully demonstrate the cost advantages of fiber-filled composites. Therefore, She et al.[87] prepared pulp fiber/PLA composites from bleached pulp fibers with higher content (10–50 wt%) by melt blending. Two different methods, roller mill extrusion and dense refining, were chosen to prepare the composites during the melting process, and the effects of the different preparation processes on the mechanical and

thermal properties of the composites were compared. The results show that the tensile properties and elastic modulus of the composites are improved compared to those of pure PLA at 50 wt% pulp fiber content due to the more uniform distribution of the pulp fibers in the pulp fiber/ PLA composites prepared by the dense refining method. However, the thermal properties of the composite are adversely affected by the low thermal degradation temperature of the pulp fibers themselves.

The crystallinity of a composite has a significant impact on the properties of the material. Pérez-Fonseca et al.[88] prepared PLA biocomposites using two different woods, white ash and tzalam, by compression and rotational molding techniques in order to improve the crystallinity and impact strength of PLA/wood particles. Annealing at 100°C for 40 min improved the crystallinity of the composites by 60% and compensated for the loss of energy storage modulus of the composites without annealing treatment. In addition, the dimensional stability, thermal stability and impact strength of the composites were all improved compared to the non-annealed composites.

Farrokhpayam et al.[89] prepared PLA/cold mountain wood chips and PLA/pulp fiber composites to investigate the water absorption, mechanical properties and morphology of the composites. In terms of physical and mechanical properties, the reinforcement effect of wood flour was better than that of pulp fiber, and the mechanical properties of the composites could be enhanced by increasing the filler content.

Several researchers have also investigated whether the length and geometry of the wood has an effect on the properties of WPC.[90–92] Madyan et al.[93] found that there may be some errors in the dimensions screened using conventional filter sieves, combining image processing and analytical techniques to analyze the true dimensions of wood fibers (WF) in composites. Most fibers were found to have lengths and widths greater than the previous mesh sizes. The use of sieve size results as the fiber geometry for the production of WPC is somewhat misleading. The dimensions of the WF change during processing. Fibers with high aspect ratios tend to become entangled during processing and are therefore more likely to break rather than be pulled out of the polymer during manufacture.

As wood resources are currently tight, the wide range of planting, fast-growing and inexpensive hemp fibers go to replace WF. The cores of the hemp stalks account for 70% of the total mass of hemp, so hemp stalk powder (HP) has the potential to replace wood powder in WPC. From the point of view of reducing the production cost of WPC, on the one hand, some straw powder, which is cheaper than wood flour, can be selected, and on the other hand, the content of polymer can be reduced to a certain extent while maintaining the minimum usable properties of WPC. Related studies[94,95] have shown that the tensile properties of composites are worst at 50% added mass fraction, the higher the wood flour content, the mechanical properties of WPC will rise, but too high a wood flour content will lead to a decrease in the mechanical properties of WPC instead. Jing et al.[96] used a twin-screw extruder to prepare HP/PLA composites. The bending strength and bending modulus of 50-HP/ PLA prepared by KH550-modified HP did reach the maximum, which was 61.2% and 17.2% higher than that of the unmodified HP/PLA, respectively.

Compared to wood, bamboo has the advantages of rapid growth, high yield, early maturity and excellent mechanical properties of bamboo fibers,[97] so WPC can be prepared from PLA and bamboo fibers. The high cellulose and lignin content of bamboo fibers and the low ash content affecting interfacial bonding result in better overall mechanical properties. Bamboo fiber/PLA composites can be prepared by means of melt injection molding.[98] In addition to the injection molding method, bamboo fiber/ PLA composites can also be prepared using melt hot pressing methods. Some researchers have investigated the processing conditions during the preparation and production process in order to optimize the processing conditions. In the preparation of bamboo fiber/PLA composites using the hot pressing process, three factors, namely pressure, temperature and time, were addressed and it was concluded that the pressure had a significant effect on the tensile strength of the composites.[99] Studies have also targeted modified bamboo fibers to enhance the overall properties of the composites.[100]

Some perennial herbs also have great potential. Silver grasses can be ground into a powder and blended with PLA to enhance the performance of PLA. There is a wide variety of energy grasses with clear advantages (high yield, high adaptability, cold resistance, pest resistance, etc.). Besides being used as a solid fuel, it can now also be used as a source of woody fiber for WPC.[101] Ma et al.[102] crushed perennial herbaceous plants such as reed, switchgrass, silvergrass and pennisetum by means of a ball mill. The above materials were well dispersed in the PLA matrix after melt blending with PLA. The pre-treatment of energy grass is a combination of physical and chemical methods (ball milling followed by acid treatment, alkali treatment and steam blasting treatment). The ball milling process reduces the particle size of the energy grass to a large extent, resulting in rougher fiber edges and enhanced interfacial adhesion. The ball milling process also destroys the crystalline zone and increases the non-crystalline zone, thus affecting the properties of the composite. Comparing the four

different types of energy grasses, the PLA/silver grass composites showed better mechanical properties with tensile strength, Young's modulus and impact strength of 23.1 MPa, 652.9 MPa and 38.67 kJ/m^2, respectively. The composites treated with acid showed better mechanical properties with tensile strength of 26.94 MPa, Young's modulus of 720.36 MPa and impact strength of 40 kJ/m^2.

The natural fibers themselves are susceptible to fungal and mold infestation, which not only reduces the mechanical properties of the composite, but also affects the aesthetics. Borysiuk et al.[103] studied the effect of two different fillers, coniferous wood chips and coniferous bark, on the growth of molds in PLA-based WPC. The effect of *Aspergillus niger* on the composites was studied and it was found that the bark blend was more susceptible to corrosion by *A. niger*. It was also found that the composites prepared using smaller particles were less antimicrobial.

In order to provide PLA-based WPC with antimicrobial properties, metallic antimicrobial agents can be prepared using a number of metal nanoparticles to enhance the antimicrobial properties of the composite. Yang et al.[104] used micron-sized copper-zinc alloy particles to reinforce particleboard wood flour/PLA composites. The copper-zinc alloy particles enhanced the antimicrobial properties and gloss of the composites while the mechanical properties and thermal stability of the composites were significantly improved. The copper-zinc alloy has a golden luster that will give the composite material a unique color and shine, thus broadening its application prospects in the home, artwork, toys and other areas.

Although WF can increase the stiffness and strength of thermoplastics, they can also make the composite less tough. To reduce the negative effects caused by WF, Petchwattana et al.[105] used acrylic core-shell rubber (CSR) particles to toughen and modify PLA. The impact resistance of the composite is enhanced by the good interfacial adhesion of the two materials and the good dispersion of CSR particles in PLA, with a reduction in the tensile modulus and strength of the composite. With the addition of rubber wood shavings, the formation of hydrogen bonds between the hydroxyl groups in the rubber wood and the ester, carboxyl or carbonyl groups in the PLA results in another increase in the tensile modulus and strength of the composite.

Many natural polymeric materials also have reuse value in terms of contaminants or wastes generated during processing. One such type of waste is wood leachate (WL) from medium density fiberboard, which will continue to be produced by the wood-based industry in the future and which can be a serious environmental hazard if released directly into the environment as an acid. It is therefore relevant to utilize this waste. The main substances in WL are lignin, phenol and tannin, so they can be used as fillers for fusion with PLA. The solids in WL are extracted by heat treatment and then crushed to obtain a powder for melt blending with PLA. The lignin in WL can play an adhesive role and improve the mechanical properties of the composite. The phenol and tannin in WL can play an antibacterial role.[106]

3.2 | Biodegradable polymers

Blending PLA with some tougher polymers is also a common method to improve the toughness of PLA. For example, the use of Acrylonitrile-butadiene-styrene copolymer combined with the compatibilizer styrene–acrylonitrile-glycidyl methacrylate copolymer can substantially improve the toughness of PLA[107]; the use of elastomeric glycidyl methacrylate or maleic anhydride functionalized ethylene-acrylate based elastomers for melt blending with PLA and chemical interactions between PLA end groups and epoxide or maleic anhydride functional groups of the elastomer, which enhance the flexibility and processability of PLA films[108]; the use of thermoplastic polyurethane (PU) to toughen PLA without sacrificing the necessary mechanical strength at the same time.[109] It is worth noting that although the above polymers are good at toughening PLA and improving its performance, they are toxic or contaminating, so some researchers have turned their attention to reinforcing PLA with biodegradable polymers. Commonly used biodegradable polymers include PBAT, PBS, PPC, and so on. Direct mechanical blending of these polymers with PLA can improve their impact strength.[110–112]

PBAT is a fully biodegradable aliphatic and aromatic copolyester made from the copolymerisation of polybutylene adipate and polybutylene terephthalate. PBAT has excellent heat resistance, good ductility, toughness, high elongation at break, good hydrophilicity and water resistance, but low tensile strength and modulus of elasticity. PBAT is blended with PLA to improve the tensile strength of PLA.[113] However, the molecular chain segment structures of PBAT and PLA are very different, resulting in poor mechanical properties of the blends.[114] It can be modified by bulking through bulking agent blending, block copolymer bulking blending, starch blending. PBAT/PLA/calcium carbonate ($CaCO_3$) composite films were prepared by melt blending and blown film forming techniques using low cost $CaCO_3$. The addition of $CaCO_3$ enhanced the compatibility between PBAT and PLA, improved the thermal stability, tensile properties, complex viscosity, energy storage modulus and loss modulus of the composites and reduced the crystallinity of the composites.[115]

As PLA also thermally degrades at higher temperatures, the addition of magnesium oxide (MgO) to PLA/PBAT composites can reduce the thermal degradation temperature and activation energy of the composites.[115] Using the solution blending method to prepare PLA/PBAT/MgO composites, the addition of MgO can enhance the initial thermal decomposition temperature of PLA/PBAT/MgO composites.[84]

During melt extrusion of PLA/PBAT composites, melt flow rate, softening temperature and other factors affect the processing fluidity of the composite, which in turn affects the base shape of the blend. PLA/PBAT/PBS blended composites are prepared by adding PBS to PLA/PBAT composites. The PBS in the composites improves the compatibility of the PLA/PBAT composites. The impact strength, melt flow rate, softening temperature and elongation at break of the PLA/PBAT/PBS blended composites decreased significantly during the increase of PBS content. When the PBS content reached 25%, the fluid rate of the PLA/PBAT/PBS blends decreased by 4.5% and the softening temperature by 7.8%, improving the processing flow and facilitating the extrusion setting of the blends.[116]

PBS is relatively inexpensive, has excellent mechanical properties and good heat resistance, so PBS can also be used in isolation to reinforce PLA. both PLA and PBS are hydrophobic materials and have no polarity on their surfaces. In order to improve the interfacial compatibility of WPC made from these two materials, Ge et al.[117] used two bulking agents (Maleic anhydride graft PLA(MAPLA) and Silane γ-(2,3-epoxypropoxy)propytrimethoxysilane) to enhance the compatibility of the composites. PLA/PBS/straw powder composites are prepared by using injection molding techniques. And the best mechanical properties (strongest tensile and flexural strength) of the composites were achieved when the content of MAPLA was less than 5%. The reason for this is that the functional groups of maleic anhydride react with the large number of hydroxyl groups on the surface of the straw powder to form hydrogen bonds, which enhance the bond between the straw powder and the plastic.

PPC is an aliphatic polycarbonate formed by the polymerization of CO_2 and propylene oxide in the presence of a catalyst. However, PPC is an amorphous polymer with poor thermal stability, which severely limits its industrialization. Blending PPC with PLA can enhance the toughness of PLA while overcoming the disadvantages of PPC's poor thermal stability.

The PPC and PLA blends were in a partially compatible state with poor compatibility. Zhang et al.[118] used tert-butyldimethylchlorosilane to modify alkali lignin (AL) with methylsilane and melt-blended modified lignin (SAL) and PLA/PPC composites into films. The addition of SAL improved the crystallization ability of PLA. The mechanical and oxygen barrier properties of the PLA/PPC/SAL composite packaging film were optimized when the amount of SAL was 1.5%. Compared to the PLA/PPC/AL composite, the tensile strength of the PLA/PPC/SAL composite packaging film increased by 6.9% and the oxygen barrier capacity increased by 33.7%; the water vapor transmission coefficient of the composite packaging film reached its lowest value at 1% SAL addition.

PLA also has potential as a shape-memory material, and Ji et al.[119] prepared a PLA-PCL-PLA triblock polymer by melt-condensation of the degradable polymers PLA and PCL. The PLA-PCL triblock copolymer is then chain expanded using hexamethylene diisocyanate and 1,4-butanediol to obtain an environmentally friendly polylactic acid-based thermoplastic elastomer. The PU structure was introduced into the PLA chain segment. The polylactic acid thermoplastic elastomer (PLAE) is soft and tough, and the PLAE can be blended with PLA to obtain a PLA-based shape memory material with good shape memory and a recovery rate of about 80%.

3.3 | Inorganic materials

The inorganic filler has excellent weather resistance, heat resistance and degradability, and so on. After blending with biodegradable plastics, the resulting products have greater advantages in terms of effect and marketing.[120]

Tian et al.[121] used a continuous fiber reinforced thermoplastic composite (CFRTPC) preparation process based on 3D printing and fusion deposition molding (FDM) technology to prepare PLA/carbon fiber composites using carbon fiber as the reinforcing phase and PLA as the matrix. When the carbon fiber content was at 27%, the composite achieved a flexural strength of 335 MPa and a modulus of 30 GPa. Li et al.[122] also used the FDM technique to prepare PLA/graphene composites. The main focus was on the influence of factors that may affect the tensile properties of PLA/graphene when 3D printing (filling density, filling pattern, printing method and layer height). The results show that the above parameters affect the tensile properties of the composites in the order of filling density, filling pattern, printing method, layer height, and the best printing parameters process for PLA/graphene was derived as filling density 70%, filling pattern linear, printing method lie-flat and layer height 0.25 mm. The addition of carbon fiber (CF) to PLA/PPC blends can enhance the mechanical and thermal properties. Hazer et al.[123] prepared CF/PLA/PPC composites using extrusion process and injection molding technique.

The addition of CF enhanced the mechanical and thermal properties of PLA/PPC and the ratio of CF to PLA/PPC was 50:50 The reinforcement effect of the composites was better than that of the composites with a 90:10 ratio of PLA/PPC.

Carbon nanotubes (CNT) have excellent mechanical strength, high electrical conductivity and good thermal conductivity. CNT can be blended with polymers to improve the mechanical, thermal and electromagnetic properties of polymers.[124] However, CNT has a high aspect ratio and specific surface area, with high surface energy, and is very prone to agglomeration and entanglement in the polymer. Polyvinyl butyral (PVB) is a condensation product of PVA and n-butyraldehyde under acid catalysis, which has good light transmission, better degradability, good impact resistance and can be used as a modifier for degradable materials.[125,126] Zhang et al.[127] used PLA as the matrix, single-walled carbon nanotubes (SWCNT) as the reinforcement and PVB as the interfacial compatibilizer to prepare SWCNT and PVB synergistically modified PLA composites using twin-screw extruder melt blending. When 10% PVB and 0.5% SWCNT were added to the PLA matrix, the tensile strength, elongation at break and impact strength of the SWCNT/PVB/PLA composites were increased by 20.7%, 121.8% and 262.6%, respectively, compared to pure PLA. The electrical conductivity of the composites was improved by six orders of magnitude compared to pure PLA. In addition, the addition of SWCNT and PVB can effectively improve the heat deflection temperature and thermal conductivity of the PLA blends.

Due to the poor dispersion of CNT in polymers and poor interfacial adhesion, treatment of CNT is required prior to the modification of PLA using CNT. Zhang et al.[128] used (sodium dodecylbenzene sulfonate) to modify the surface of multi-walled CNT, and then fused levopolylactic acid (PLLA) with modified CNT by melt blending, which ultimately reduced the conductivity over penetration threshold of the composite and improved the thermal stability. CNT can also be used to enhance composites with PLA matrix. Urtekin et al.[129] used the bridging effect of CNT to enhance the interaction between PLA and polycarbonate (PC) before. By combining multi-walled CNT containing different functional groups Non-functionalized (MWCNT), hydroxyl-functionalized (MWCNT-OH), carboxyl-functionalized (MWCNT-COOH), and polycarbonate-grafted multi-walled carbon nanotube (PC-g-MWCNT) were compounded with PC/PLA in a twin-screw extruder, resulting in good grafting of PC onto MWCNT and improved T_g of the composites. The elongation at break, tensile strength and modulus of the composites reached their maximum values after the addition of 5 wt% MWCNT-OH to PC/PLA.

Despite its high modulus of elasticity and tensile strength, PLA has high brittleness and low elongation at break. To solve this problem, common methods of strengthening PLA include plasticization, copolymerization, blending with flexible materials and the addition of nano-fillers. Nanofillers have high specific surface area, high surface energy and functionalization[130] and can be used to improve the strength and toughness of PLA. Graphene is considered to be a revolutionary material of the future in 2D materials and is commonly used to enhance the properties of polymers. However, good dispersion and interfacial compatibility of the filler between the substrates is required if more desirable performance enhancements are to be achieved.[131] Li et al.[132] prepared graphene oxide (GO) grafted with PLA reinforced PLA (GO-g-PLA) using a combination of solution blending and molding techniques. The elongation at break and tensile strength of GO-g-PLA nanocomposites were improved by 114.3% and 105.7%, respectively, compared to pure PLA, but the ductility improvement effect was only slightly higher at 150% or so. Further exploration is required to further improve the performance of PLA-based nanocomposites. Polyethylene glycol (PEG) is a non-toxic, biocompatible, readily soluble in water and various organic solvents,[133] and an effective plasticising material for PLA.[134] However, the enhancement of PLA with PEG alone is limited, reducing the tensile strength of PLA by only about 50%.[135] It is possible to surface modify GO with PEG, however the grafting of GO to PEG occurs at the carboxyl group of GO, while the hydroxyl and epoxy groups are distributed on the surface.[136] For this reason, Williamson synthesis can be used to convert some of the hydroxyl groups of GO into carboxyl groups, thereby increasing the reactive position of GO toward PEG.[137] Niu et al.[138] used carboxygraphene oxide (GC) grafted with PEG (GC-g-PEG) for melt blending with PLA. The good dispersion and interfacial compatibility of GC in the PLA matrix resulted in PLA/GC nanocomposites with higher crystallinity, thermal stability and mechanical strength, and with the addition of 0.3 wt% of GC-g The elongation at break of PLA/GC nanocomposites was enhanced by a factor of 7 compared to pure PLA under the addition of 0.3 wt% GC-g-PEG.

Montmorillonite (MMT) is an inorganic nanomaterial with a typical layered structure, rich in energy storage, low in price and high in rigidity. The addition of MMT to the PLA matrix can enhance the strength, toughness, biodegradability and heat resistance of PLA composites.[139] However, the melting low viscosity droplets of PLA can spread fire rapidly and the fire risk is high. Therefore it is of some practical importance to enhance the flame retardancy of PLA. However, adding MMT to the PLA matrix
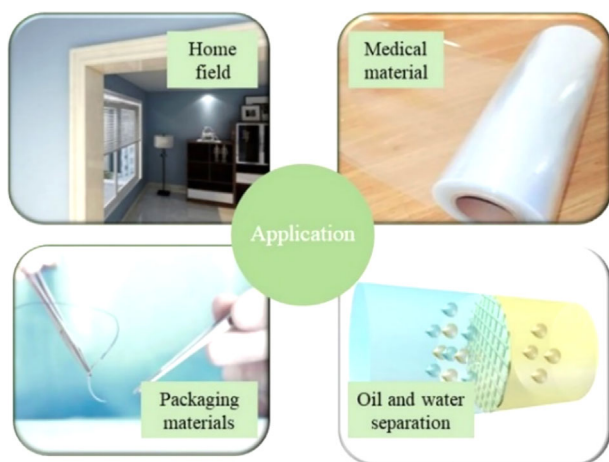
FIGURE 8 Application of polylactic acid composites [Color figure can be viewed at wileyonlinelibrary.com]

alone does not achieve the desired flame retardant effect, so a flame retardant needs to be added to the PLA-based composite. The halogen-free flame retardant Crystal form II ammonium polyphosphate (APP-II) can be added to PLA matrix in synergy with calcium-based MMT to effectively improve the flame retardancy of PLA composites. In the case of PLA/Crystal form APP-II /calcium MMT composites, only 3 wt% of the nano compound is required to achieve a UL-94 vertical combustion rating of V-0 and an ultimate oxygen index of 28.0%. At the same time, the composite retains good mechanical properties.[140] In addition, the use of flame retardants can also lead to excellent flame retardancy of PLA. Yemisci et al.[141] added APP, boron phosphate and tri-phenyl phosphate as flame retardants to PLA, respectively, where tri-phenyl phosphate was more uniformly dispersed in PLA and the flame retardant effect was the best among the three. MMT was compounded with the above flame retardants to achieve synergistic flame retardant effect.

Some plasticisers can also enhance the toughness of PLA/TPS composites. Mao et al.[142] used sodium-based MMT modified with chitosan polyammonium salt (HACC) first, followed by the use of an environmentally friendly plasticizer, tributyl acetyl citrate, and modified MMT or organic MMT or sodium-based MMT added to PLA/TPC composites to prepare the composites using melt extrusion. By comparing the reinforcing effect of three different types of MMT, on PLA/TPS composites, it is found that HACC-MMT has the strongest reinforcing effect. The reason for this is the good dispersion and compatibility of HACC-MMT in PLA/TPS, which enhances the overall performance of the material. During the melt extrusion process, TPS and PLA molecular chains can be inserted between the Si—O tetrahedral and Al—O

octahedral layers of MMT due to shear force, which is beneficial to the reinforced material.

Sharma et al.[143] explored the effects of silica nanoparticles and alumina nanoparticles on composites using a fused filament fabrication 4D printing technique to fabricate PLA-based WPC. The hardness tensile and flexural strengths of the composites were increased by 40%, 25% and 3.3% respectively with the addition of 2 wt% alumina nanoparticles. With the addition of 2 wt% silica nanoparticles, the hardness, compressive and flexural strengths of the composites were increased by 60%, 55% and 10%, respectively. The addition of both materials enhanced the thermal stability and reduced the maximum shape recovery of the composites. The nano-silica showed better performance in terms of mechanical properties, thermal properties and maximum shape recovery of the composites.

4 | APPLICATIONS OF PLA COMPOSITES

The application of PLA has become more and more widespread today and is widely used in medicine, biotechnology, agriculture, forestry, animal husbandry, petroleum, chemicals, building materials, environmental protection, packaging, electronic and electrical products and transportation.[144] This section focuses on the four areas of application as shown in Figure 8.

4.1 | Building and home furnishing

PLA-based WPC can be used in the same way as other WPCs in flooring, household and construction applications, which require PLA-based WPCs that are impact resistant, tough and light enough to resist cracking and fracture during use.

Qin et al.[145] improved the tensile properties of the composite by blending WF with PLA, increasing the tensile properties from 23.1 to 30.3 MPa. However, the density of WPC did not decrease significantly. To further achieve the lightweight goal, Wang et al.[146] used azodicarbonamide (AC) as a foaming agent in PLA/WF composites to make the composites lighter, improve the cell structure, enhance the mechanical strength and improve the material cost effectiveness. This enabled the WF/PLA composites to be better used in architectural or automotive decoration applications.

PLA-based WPC can be combined with 3D printing technology, which allows the shape and color of composite materials to be used without the limitations of traditional molds. In order to enrich the functionality of WPC,

Yang et al.[147] used $Sr_2MgSi_2O_7:Eu^{2+}$, Dy^{3+} (SMSED) phosphor, chipboard wood powder, and PLA as raw materials to prepare composites that could be used for 3D printing by melt extrusion. In addition, the phosphor was modified with KH550, which forms a dense protective film on the surface of the phosphor and significantly enhances the mechanical and thermal stability of the WPC, as SMSED dissolves easily in water and thus increases the pH. Based on this material, a safety warning model that emits blue light in dark conditions was prepared.

4.2 | Medical materials

An important reason why PLA can be used as a medical material is due to its good biocompatibility. the intermediate product of PLA metabolism in the human body is LA, which can be metabolized by the body and is non-toxic and non-hazardous. PLA can be used in medical materials for bone stents, wound dressings, masks, medical sutures, pharmaceutical slow release, surgical gowns, absorbable vascular stents, heart valves, nappies, physiological hygiene products and other applications.

4.2.1 | Medical stent

In the field of medicine, bone defects have always been a major challenge in reconstructive surgery. For the treatment of fractures, the current autograft treatment has many disadvantages (e.g., limited available tissue, pain, donor site pathology, etc.) and therefore there is an urgent need for the development and utilization of engineered implants or scaffolds. Currently there are three main types of commercial stents: bare-metal stents (BMS), drug-eluting stents (DES) and biodegradable stents (BS). BMS and DES can leave permanent residues in the body that may affect subsequent treatment and recovery. Bioresorbable stents, on the other hand, provide both mechanical support and drug delivery, and can be completely absorbed by the body, allowing for better vascular remodeling and reducing the triggering of persistent inflammation.[148]

The ideal bone scaffold should have the following properties: suitable mechanical properties, controlled biodegradability, appropriate osteoinductive capacity and drug release capacity, thus improving the simulated environment for cell adhesion and value addition.[149] Samadian et al.[150] proposed a scaffold related to PLA to promote bone regeneration. A PLA/PCL/gelatin nanorubber/taurine composite scaffold was prepared by combining electrospinning and 3D printing techniques. The scaffold has a porosity of up to 90% and the porous

structure ensures its hydrolytic conditions. This composite promotes bone regeneration with suitable porosity, acceptable hydrophilicity, mass loss, mechanical properties, cell and blood compatibility.

Hydroxyapatite (HA) is a biologically active, non-toxic and osteoinductive ceramic. Combining it with biocompatible PLA therefore has a better level of bioactivity and regenerative potential for bone tissue engineering.[151] Bernardo et al.[152] prepared PLA/HA composite porous scaffolds based on FDM and 3D printing techniques. The combination of PLA and HA overcame the hydrophobicity of PLA and also enhanced the mechanical properties of the composite, stimulating osteoinduction and osseointegration of the scaffold.HA neutralized the acidification effect of PLA degradation and this material did not exhibit immunostimulatory properties. PLA/HA composites were effective in inducing human interstitial osteogenesis even in the absence of classical osteogenic stimulation. The PLA/HA composite can effectively induce osteogenic differentiation of human mesenchymal stem cells even in the absence of classical osteogenic stimulation.

Both polymethylene-trimethylene carbonate (PTMC) and PLLA polymers are biocompatible and biodegradable, and the combination of PLLA and PTCM can significantly improve the mechanical properties of the material and minimize the inflammatory response to the wound. He et al.[153] prepared a microsphere scaffold for drug release using chitosan (CS)-coated PTMC/PLLA/oleic acid modified hydroxyapatite (OA-HA)/vancomycin hydrochloride (VH), which has excellent biocompatibility. The incorporation of PLLA, OA-HA, and VH in the PTCM microspheres not only slows down the biodegradability of the scaffold, but also enhances the mechanical properties and surface activity of the microsphere scaffold. The CS coating stimulates osteoblast adhesion prior to OA-HA infiltration and this PTCM/PLLA/OA-HA/VH microsphere scaffold has promise for use in bone tissue engineering.

In bone tissue engineering and regenerative surgery, there is a need to focus on infection control and materials with antimicrobial activity and osteoinductive properties. Therefore, Orafa et al.[154] wrapped amoxicillin on a laponite nanoplate and then immobilized it on the surface of electrospun PLA nanorubber to meet these requirements. This scaffold's showed good inhibition of *Staphylococcus aureus* and *Escherichia coli* and the release of amoxicillin was divided into a burst release for the first 48 h and a sustained release for 21 days.

4.2.2 | Medical dressing

When the human epidermis is damaged, it is often necessary to apply a dressing to the wound surface to give it a

suitable environment for repair. PLA/bacterial cellulose/CS multistage dressings prepared using electrostatic spinning technology have good water absorption (>6790% water absorption) and moisture vapor permeability (>10,000 g/[m d]), biocompatibility, volumetric stability and comfort.[155] In the process of wound healing, Hajikhani et al.[156] first combined cefazolin as an antibiotic with collagen and loaded it onto a co-axial electrospun fiber scaffold with PLA as the core material and Polyvinylpyrrolidone as the shell material, ultimately achieving both drug release, accelerated healing and prevention of contamination. For exuding wounds, in addition to the efficient antibacterial effect, it should also have a fast-drying structure with high liquid absorption capacity. Therefore, PLA, PU, and silver nanoparticles (AgNP) were prepared as hollow PLA/PU/AgNP nanofiber trauma dressings, and such dressings can effectively inhibit *E. coli*, *Pseudomonas aeruginosa*, and *S. aureus*.[157] Besides, for dressings that need to be waterproof, PLA/PU nonwoven mats can be prepared by electrostatic spinning technique with POSS Amic Acid Isobutyl as capacitor. This material has ultra-high hydrophobicity and can be applied for dressing surface coating.[158]

4.2.3 | Mask

With the outbreak of the new crown, wearing masks in public has become the norm in most countries. The use of PLA for disposable masks has great degradation advantages, overcoming the problem of microplastics that tend to be produced during the degradation of ordinary disposable mask fabrics. Degradation studies on three-layer PLA masks revealed that PLA masks degrade at a faster rate under alkaline conditions.[159] Of the three layers, only the middle layer underwent the major degradation of random chain breakage and cleavage of the enol or enol ester chain ends due to the thin middle layer of the mask, while the outer and inner layers were less affected. After 8 weeks of degradation under alkaline conditions, the masks do not reach a state of complete degradation, but the small dissolved fragments can be completely mineralized to CO_2. PLA masks can be 3D printed using fused deposition modeling techniques, but masks prepared using this method may have a high porosity, which can affect the protective function of the mask. Some researchers have therefore investigated the suitability of PLA for viral protection.[160] The decontamination efficiency of conventional chemical disinfectants (96% ethanol, 70% isopropyl alcohol and commercially available disinfectants containing 0.85% sodium hypochlorite) on PLA surfaces was measured and confirmed that the PLA protective masks are compact. Adequate

protection against microbial corresponding particles, including viruses, is possible. For mask applications, PLA can also be used as a mask filter. Nanoscale bio-based PLA fibers are made by electrostatic spinning of very dilute solutions. Filters prepared from such fibers have a high filtration efficiency with a low pressure drop (104 Pa), as well as a more durable particulate matter filtration efficiency and biodegradability.[161]

4.2.4 | Medical suture

Medical sutures made from PLA are non-toxic, biocompatible, biodegradable and high-strength. PLA gradually decomposes into CO_2 and H_2O during the wound healing process, which will avoid the pain associated with wound removal.[162,163] Therefore, PLA sutures have been used in many types of surgery (e.g., facial, neck, circumcision, colostomy, etc).[164] However, different types of wounds require different healing times and the degradation rate of conventional PLA sutures is fixed and there is no way to control the rate and duration of degradation. The degradation rate can be regulated by adjusting the diameter of the suture, copolymerizing or blending with some polymer materials such as PEG and polyglycolic acid, and blending with some nanomaterials (such as nano-SiO_2 and nano-MMT), but these methods are mainly used to shorten the degradation time. Based on this, Liu et al.[165] compounded CNT with PLA sutures, and the resulting composite extended the degradation time of PLA sutures. The complete degradation time of CNT/PLA sutures was extended from 49 weeks to 63 to 73 weeks for pure PLA sutures, effectively extending their strength effective time in degradation.

4.3 | Packaging

PLA has great potential in the packaging sector. However, due to its inherent performance deficiency of being too brittle, PLA needs to be laminated with other materials to obtain a good composite with excellent properties. In food packaging, anti-bacterial properties are often more important.

Many metal oxides (ZnO, MgO, TiO_2) can be compounded with PLA to obtain bacteriostatic composite films. PLA compounded with zinc oxide nanoparticles and applied to the surface of the wrapper using a coating machine, the wrapper will have a good freshness retention effect.[166] The addition of zinc oxide nanoparticles does not change the crystal structure of PLA, but causes disorder in the ordering pattern of PLA polymer chains,

enhancing the mechanical properties, antibacterial properties and oil resistance of the packaging.

Composite packaging materials can be prepared by laminating thymol or carvacrol in β-cyclodextrins (β-CDs) and then laminating them with PLA.[167] PLA/β-CD-thymol packaging can be used as a packaging material to preserve the color, weight and phenolic content of berries (blackberries and raspberries). A comparison of commercially available commercial clamshell and PLA packaging materials can extend the shelf life of blackberries and raspberries by up to 1 week at 4°C.

Antimicrobial packaging can be prepared by incorporating cinnamaldehyde or tea polyphenols into PLA, PBAT and starch blends by extrusion techniques.[168] PLA/PBAT films loaded with cinnamon phenols inhibit the growth of *E. coli* and *S. aureus* at 4°C and have good barrier properties to prevent water evaporation, ensure texture characteristics and ensure the quality and safety of meat analogues.

Using para-phenylenediamine as a cross-linking agent, para-phe-nylenediamine (p-SB) compounds can be synthesized via vanillin. PLA can react with p-SB to reduce the brittleness of PLA and enhance antimicrobial activity.[169] The resulting composites have hydrogen bonds between PLA and SB, resulting in enhanced toughness, improved tensile properties and hydrophobicity, and improved barrier properties.

In addition, the high price of PLA largely limits its use in packaging applications. To overcome this disadvantage, PLA can be blended with other biopolymers to enhance the performance of PLA while also reducing its cost. For example, organic clay is used as a reinforcing material and the layer spacing between the clay sheet crystals is enhanced by using antibacterial natural rosin to reinforce the organic clay.[170] By blending the modified clay with PLA/PBAT, the resulting packaging material can be used in food packaging and biofilm applications.[171]

4.4 | Oil water separation material

In recent years, pollution caused by crude oil spills and industrial effluent discharges can not only cause damage to the ecological environment, but may also pose a certain danger to humans. Therefore, it is urgent to develop an oil–water separation material.

Xing et al.[172] prepared a PLA porous membrane structure based on FDM 3D printing technology with reference to the properties of lotus leaves, and then used PS nanospheres for chemical etching and decorative effects to enhance the micron-scale and nanoscale roughness of the porous membrane, respectively, ultimately achieving superhydrophobicity. The oil–water separation efficiency is up to 99.4% when the pores of the PLA porous membrane reach 250 μm. They are also degradable and do not cause pollution.

Aerogels with ultra-low density and ultra-high specific surface area, hydrophobic and lipophilic aerogels possess excellent oil–water selectivity and adsorption properties, showing great potential in oil–water separation.[173] Aerogels can also be prepared using PLA, but pure PLA aerogel oils have poor adsorption capacity, adsorption selectivity and mechanical properties.[174] The use of metal–organic skeletons as fillers is one of the important methods to enhance the performance of porous materials. Among the metal–organic skeletons, zeolite imidazole skeletons (ZIF-8) have been widely studied[175–177] and have the advantages of low cost, simple preparation, large specific surface area and adjustable pore size. The ZIE-8@PLA composite aerogel is prepared by physical blending and water-assisted thermally induced phase separation by introducing ZIF-8 into the PLA matrix. The aerogel has excellent hydrophobicity, high oil absorption efficiency and good durability. The aerogel has excellent hydrophobicity, high oil absorption efficiency and good durability. The aerogel maintains a high adsorption capacity even after repeated use.

5 | CONCLUSION

PLA is a biodegradable polymer that is beginning to replace some traditional plastics. There are various ways to prepare PLA, and the properties of the PLA produced can vary. However, PLA itself has certain drawbacks and needs to be modified to improve its performance. A more environmentally friendly form of modification is the use of environmentally friendly materials blended with PLA, which can also be used to reduce the price of PLA products if an inexpensive blend is used. The properties of PLA composites modified by blending are often substantially improved, and depending on the difference between the properties of the PLA itself and the blended material, PLA composites can be used in a number of applications.

DATA AVAILABILITY STATEMENT

Data sharing is not applicable to this article as no new data were created or analyzed in this study.

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REFERENCES

[1] O. Kehinde, O. Ramonu, K. Babaremu, L. Justin, *Heliyon* **2020**, *6*, e05131.
[2] Z. Menglei, Y. Zeng, Z. Jingnan, W. Yan, M. Xiaolei, G. Jian, *J. Resour. Ecol.* **2022**, *13*, 428.
[3] Q. Chang, D. Zhu, L. Hu, H. Kim, Y. Liu, L. Cai, *Sci. Total Environ.* **2022**, *822*, 153235.
[4] B. Borrelle, J. Ringma, L. Law, C. Monnahan, L. Lebreton, A. McGivern, E. Murphy, J. Jambeck, H. Leonard, G. A. Hilleary, *Science* **2020**, *369*, 1515.
[5] S. Pazos, T. Maiztegui, C. Colautti, H. Paracampo, N. Gómez, *Mar. Pollut. Bull.* **2017**, *122*, 85.
[6] J. Moore, *Environ. Res.* **2008**, *108*, 131.
[7] B. Hartmann, T. Huffer, C. Thompson, M. Hassellöv, A. Verschoor, E. Daugaard, S. Rist, T. Karlsson, N. Brennholt, M. Cole, *Environ. Sci. Technol.* **2019**, *53*, 1039.
[8] W. Pan, L. Luo, L. Ao, H. Xie, L. Pu, S. Wang, *Appl. Chem. Ind.* **2022**, *51*, 1508.
[9] L. Gao, D. Fu, J. Zhao, W. Wu, L. Peng, *Mar. Pollut. Bull.* **2021**, *169*, 112480.
[10] E. Karalija, M. Carbó, A. Coppi, I. Colzi, M. Dainelli, M. Gasparovic, T. Grebenc, C. Gonnelli, V. Papadakis, S. Pilić, N. Šibanc, L. Valledor, A. Poma, F. Martinelli, *J. Hazard. Mater.* **2022**, *438*, 129450.
[11] H. Du, S. Lu, J. Sun, S. Song, *China Plast.* **2021**, *35*, 37.
[12] F. Jackulin, P. Senthil, V. Nguyen, F. Carolin, J. Lakshmipriya, *Environ. Chem. Lett.* **2022**, *20*, 223.
[13] X. Luan, W. Liu, Z. Cui, Y. Liu, Y. Chen, S. Lu, Y. Wang, *Resour. Sci.* **2020**, *42*, 372.
[14] K. Kalita, A. Sarmah, M. Bhasney, A. Kalamdhad, V. Katiyar, *Environ. Challenges.* **2021**, *3*, 100030.
[15] Q. Zheng, *Polym. Bull.* **2022**, *4*, 1.
[16] R. Altman, *Science (New York, N.Y.)* **2021**, *373*, 47.
[17] Y. Chen, L. Geever, J. Killion, J. Lyons, C. Higginbotham, D. Devine, *Polym.-Plast. Technol. Eng.* **2016**, *55*, 1057.
[18] X. Li, L. Meng, Y. Zhang, Z. Qin, L. Meng, C. Li, M. Liu, *Polymers-Basel* **2022**, *14*, 2159.
[19] C. Yu, *Degree Thesis*, China University of Geosciences, Beijing **2011**, p. 5.
[20] L. Li, *Degree Thesis*, Zhejiang Sci-Tech University, Hangzhou **2021**, p. 4.
[21] Y. Han, *Degree Thesis*, East China University of Science and Technology, Shanghai **2013**, p. 4.
[22] Y. Zhang, *Degree Thesis*, Changchun University of Technology, Changchun **2022**, p. 6.
[23] Y. Pang, *Degree Thesis*, Guangdong University of Technology, Guangzhou **2021**, p. 5.
[24] Z. Liu, *Appl. Chem. Ind.* **2022**, *51*, 2113.
[25] J. Cook, *Miner. Econ.* **2022**, *35*, 1.
[26] R. Wang, H. Zhang, H. Sun, S. Duan, Z. Qin, H. Li, F. Zhu, Y. Zhang, *China Plast.* **2022**, *36*, 158.
[27] N. Kalita, N. Damare, D. Hazarika, P. Bhagabati, K. V. Atiyar, *Environ. Challenges* **2021**, *3*, 100067.
[28] O. Avinc, A. Khoddami, *Fibre Chem.* **2009**, *41*, 391.
[29] M. Li, Y. Chen, *China Plast.* **2022**, *36*, 102.
[30] J. Raquez, Y. Habibi, M. Murariu, P. Dubois, *Prog. Polym. Sci.* **2013**, *38*, 1504.
[31] N. Rosli, I. Ahmad, F. Anuar, I. Abdullah, *J. Clean. Prod.* **2018**, *198*, 987.
[32] M. Akrami, I. Ghasemi, H. Azizi, M. Karrabi, M. Seyedabadi, *Carbohydr. Polym.* **2016**, *144*, 254.
[33] Y. Ding, B. Lu, J. Ji, *Prog. Chem. (Beijing, China)* **2020**, *32*, 738.
[34] M. Singhvi, S. Zinjarde, D. Gokhale, *J. Appl. Microbiol.* **2019**, *127*, 1612.
[35] E. Castro-Aguirre, F. Iniguez-Franco, H. Samsudin, X. Fang, R. Auras, *Adv. Drug Deliv. Rev.* **2016**, *107*, 333.
[36] K. Stefaniak, A. Masek, *Materials* **2021**, *14*, 5254.
[37] A. Lasprilla, G. Martinez, B. Lunelli, A. Jardini, R. Filho, *Biotechnol. Adv.* **2012**, *30*, 321.
[38] M. Murariu, P. Dubois, *Adv. Drug Deliv. Rev.* **2016**, *107*, 17.
[39] N. Kalita, M. Nagar, C. Mudenur, A. Kalamdhad, V. Katiyar, *Polym. Test.* **2019**, *76*, 522.
[40] K. Duan, W. Zhen, *Polym.-Plast. Technol. Mater.* **2019**, *58*, 1.
[41] M. Yang, P. Xu, *Contemp. Chem. & Ind.* **2010**, *39*, 353.
[42] A. Hosni, J. Pittman, G. Robson, *Waste Manag.* **2019**, *97*, 105.
[43] A. Richert, G. Dbrowska, *Int. J. Biol. Macromol.* **2021**, *176*, 226.
[44] W. Jia, F. Gao, H. Zhou, F. Qian, Y. Lv, *Plast. Sci. Technol.* **2022**, *50*, 68.
[45] M. Watanabe, F. Kawai, S. Tsuboi, S. Nakatsu, H. Ohara, *Macromol. Theory Simul.* **2007**, *16*, 619.
[46] K. Masaki, N. Kamini, H. Ikeda, H. Iefuji, *Appl. Environ. Microbiol.* **2005**, *71*, 7548.
[47] H. Pranamuda, A. Tsuchii, Y. Tokiwa, *Macromol. Biosci.* **2001**, *1*, 25.
[48] Y. Tokiwa, B. Calabia, *Appl. Microbiol. Biotechnol.* **2006**, *72*, 244.
[49] N. Hegyesi, Y. Zhang, A. Kohári, P. Polyák, B. Pukánszky, *Ind. Crop. Prod.* **2019**, *141*, 111799.
[50] P. Lotrakul, S. Dharmsthiti, *World J. Microbiol. Biotechnol.* **1997**, *13*, 163.
[51] S. da Silva, E. Hinkel, T. Lisboa, V. Selistre, A. da Silva, L. da Silva, D. Faccin, N. Cardozo, *Polym. Test.* **2020**, *91*, 106732.

[52] Y. Boonluksiri, B. Prapagdee, N. Sombatsompop, *Polym. Degrad. Stab.* **2021**, *188*, 109562.

[53] A. Mittal, R. Soni, K. Dutt, S. Singh, *J. Hazard. Mater.* **2010**, *178*, 390.

[54] M. Elsawy, K. Kim, J. Park, A. Deep, *Renew. Sust. Energ. Rev.* **2017**, *79*, 1346.

[55] W. Gexia, H. Dan, Z. Wei, J. Junhui, *J. Funct. Polym.* **2020**, *33*, 492.

[56] X. Chen, L. J. Wang, H. Shi, Y. L. Shi, *Polym. Polym. Compos.* **2011**, *19*, 559.

[57] A. Guzman-Sielicka, H. Janik, P. Sielicki, *J. Polym. Environ.* **2013**, *21*, 802.

[58] I. Taiatele, T. Dal Bosco, P. Faria-Tischer, A. Bilck, F. Yamashita, J. Bertozzi, R. Michels, S. Mali, *J. Polym. Environ.* **2019**, *27*, 2577.

[59] O. Wilfred, H. Tai, R. Marriott, Q. Liu, V. Tverezovskiy, S. Curling, Z. Fan, W. Wang, *Int. J. Med. Nano Res.* **2018**, *1*, 1.

[60] D. Huang, Z. Hu, T. Liu, B. Lu, Z. Zhen, G. Wang, J. Ji, *e-Polym.* **2020**, *20*, 759.

[61] R. Yao, X. Li, H. Yang, N. Li, H. Chen, *J. Zhejiang Sci-Tech Univ.* **2022**, *47*, 1.

[62] Q. Qin, Y. Yang, C. Yang, L. Zhang, H. Yin, F. Yu, J. Ma, *Sci. Total Environ.* **2022**, *842*, 156775.

[63] Z. Chen, D. Ding, T. Yu, W. Yang, Q. Li, Y. Li, *Compos. Commun.* **2022**, *33*, 101227.

[64] A. Mohanty, M. Misra, L. Drzal, *Natural fibers, biopolymers, and biocomposites*, CRC Press, Boca Raton, FL **2005**.

[65] D. Nath, I. Oluwarotimi, Z. Zhang, B. Asa, W. Euan, M. Jean-Eudes, B. Johnny, *Compos. Part A* **2018**, *113*, 350.

[66] V. Mazzanti, M. Luna, R. Pariante, F. Mollica, G. Filippone, *Compos. Part A* **2020**, *137*, 105990.

[67] E. Lima, R. Nunes Oliveira, A. Middea, I. Miranda de Castro, M. Mattos, T. Neves, L. da Costa, R. Neumann, M. Tavares, *J. Nat. Fibers* **2022**, *19*, 1783.

[68] Z. Wen, Z. Xin, M. Hongpeng, X. Jie, G. Bin, L. Panxin, *Prog. Chem.* **1972**, *2020*, 33.

[69] W. Zhou, X. Zhang, H. Ma, X. Xu, P. Li, *Prog. Chem. (Beijing, China)* **2021**, *33*, 1972.

[70] J. Zhang, X. Sun, *Biomacromolecules* **2004**, *5*, 1446.

[71] N. Noivoil, R. Yoksan, *Int. J. Biol. Macromol.* **2020**, *160*, 506.

[72] P. Li, N. Shao, D. Lu, G. Lu, Z. Tang, *Polym. Mater. Sci. Eng.* **2022**, *38*, 56.

[73] A. Nazrin, S. Sapuan, M. Zuhri, *Polymers-Basel.* **2020**, *12*, 2216.

[74] Z. Kou, *Suliao* **2020**, *49*, 115.

[75] A. Fernanda, M. Tavares, *Polym. Test.* **2015**, *47*, 92.

[76] R. Ilyas, M. Zuhri, H. Aisyah, M. Asyraf, S. Hassan, E. Zainudin, S. Sapuan, S. Sharma, S. Bangar, R. Jumaidin, *Polymers-Basel* **2022**, *14*, 202.

[77] J. Moran, L. Ludueña, V. Phuong, P. Cinelli, A. Lazzeri, V. Alvarez, *Polym. Polym. Compos.* **2016**, *24*, 341.

[78] R. Kharismi, H. Suryadi, *J. Young Pharm.* **2018**, *10*, S79.

[79] B. Dogu, C. Kaynak, *Cellulose* **2016**, *23*, 1.

[80] J. Ren, J. Cao, L. Li, W. Zhao, J. Du, Y. Zhao, *Plast. Sci. Technol.* **2022**, *50*, 31.

[81] L. Liu, M. Qian, P. Song, G. Huang, Y. Yu, S. Fu, *ACS Sustain. Chem. Eng.* **2016**, *4*, 2422.

[82] W. Xing, H. Yuan, P. Zhang, H. Yang, L. Song, Y. Hu, *J. Polym. Res.* **2013**, *20*, 1.

[83] B. Podkościelna, M. Gargol, M. Goliszek, T. Klepka, O. Sevastyanova, *Polym. Test.* **2022**, *111*, 107622.

[84] S. Xiang, L. Feng, X. Bian, G. Li, X. Chen, *Polym. Test.* **2020**, *81*, 106211.

[85] L. Teuber, V. Osburg, W. Toporowski, H. Militz, A. Krause, *J. Clean. Prod.* **2016**, *110*, 9.

[86] Q. Wu, K. Chi, Y. Wu, S. Lee, *Mater. Des.* **2014**, *60*, 334.

[87] Y. She, Y. Fu, Q. Zhu, S. Wu, X. Xu, *Acta Mater. Compos. Sin.* **2022**, *39*, 1.

[88] A. Pérez-Fonseca, V. Ramírez-Herrera, F. Fuentes-Talavera, D. Rodrigue, J. Silva-Guzmán, J. Robledo-Ortíz, *Maderas-Cienc. Tecnol.* **2021**, *23*, 1.

[89] S. Farrokhpayam, M. Shahabi, B. Sheshkal, R. Gargari, *J. Indian Acad. Wood Sci.* **2021**, *18*, 20.

[90] S. Chaudemanche, A. Perrot, S. Pimbert, T. Lecompte, F. Faure, *Constr. Build. Mater.* **2018**, *162*, 543.

[91] A. Thumm, A. Dickson, *Compos. Part A* **2013**, *46*, 45.

[92] X. Hao, H. Zhou, B. Mu, L. Chen, R. Ou, *Compos. Part B* **2020**, *185*, 107778.

[93] O. Madyan, Y. Wang, J. Corker, Y. Zhou, M. Fan, *Compos. Part A* **2020**, *133*, 105871.

[94] M. Dalu, A. Temiz, E. Altuntaş, G. Demirel, M. Aslan, *Polym. Test.* **2019**, *76*, 376.

[95] C. Wang, Z. Ren, L. Wu, Z. Dai, C. Lu, *Plast. Sci. Technol.* **2018**, *46*, 65.

[96] M. Jing, X. Han, C. Wang, H. Wu, L. Wang, N. Wang, *China Plast. Ind.* **2022**, *50*, 99.

[97] D. Wu, X. Wei, L. Yao, L. Li, Z. Wu, L. Yu, *China Forest Prod Industry* **2022**, *59*, 48.

[98] J. Zhang, C. He, H. Tang, J. Fu, M. Wang, *Eng. Plast. Appl.* **2016**, *44*, 12.

[99] X. Zheng, L. Shao, X. Jin, C. Zhu, W. Tian, *Adv. Text. Technol.* **2022**, *31*, 1.

[100] Y. Ma, S. Qian, L. Hu, J. Qian, C. Fontanillo Lopez, L. Xu, *Polym. Compos.* **2019**, *40*, 3012.

[101] K. Henke, S. Treml, *Eur. J. Wood Wood Prod.* **2013**, *71*, 139.

[102] S. Ma, L. Kou, X. Zhang, T. Tan, *Cellulose* **2020**, *27*, 2669.

[103] P. Borysiuk, K. Krzysztof, A. Alicja, A. Radosław, B. Izabela, R. Katarzyna, N. Małgorzata, B. Piotr, *Polymers-Basel* **2021**, *14*, 157.

[104] F. Yang, J. Zeng, H. Long, J. Xiao, X. Dong, *Polymers-Basel* **2020**, *12*, 621.

[105] N. Petchwattana, S. Covavisaruch, *J. Bionic Eng.* **2014**, *11*, 630.

[106] M. Shahavi, P. Selakjani, M. Abatari, P. Antov, V. Savov, *Polymers-Basel* **2022**, *14*, 1227.

[107] L. Ma, J. Ding, S. Yu, D. Guo, L. Zhou, *Plast. Sci. Technol.* **2022**, *50*, 9.

[108] K. Dogan, S. Gumus, A. Aytac, G. Ozkoc, *Fibers Polym.* **2013**, *14*, 1422.

[109] H. Hong, J. Wei, Y. Yuan, P. Chen, J. Wang, X. Qu, S. Liu, *J. Appl. Polym. Sci.* **2011**, *121*, 855.

[110] P. Cao, F. Liu, H. Wang, *Plast. Sci. Technol.* **2021**, *49*, 51.

[111] Y. Li, X. Meng, X. Cui, T. Wang, Y. Shi, L. Liu, L. Song, *Plast. Sci. Technol.* **2021**, *49*, 7.

[112] H. Zhao, S. Hu, X. Xia, Y. Zhang, *Eng. Plast. Appl.* **2021**, *49*, 131.

[113] H. Chiu, S. Huang, Y. Chen, M. Kuo, T. Chiang, C. Chang, Y. Wang, *Int. J. Polym. Sci.* **2013**, *2013*, 951696.

[114] X. Wang, S. Peng, H. Chen, X. Yu, X. Zhao, *Compos. Part B* **2019**, *173*, 107028.

[115] Y. Zhang, A. Huang, W. Zhang, L. Gao, Z. Chen, R. Xu, *Eng. Plast. Appl.* **2019**, *47*, 158.

[116] H. Li, R. Duan, G. Wen, J. Feng, D. Li, J. Liu, *Suliao* **2022**, *51*, 29.

[117] Z. Ge, X. Li, D. Si, *IOP Conf. Ser: Mater. Sci. Eng.* **2020**, *892*, 012018.

[118] N. Zhang, M. Zhao, Z. Zhang, Y. Chen, *Suliao Gongye* **2022**, *50*, 154.

[119] F. Ji, J. Li, Y. Weng, J. Ren, *Mater. Res. Express* **2019**, *7*, 015315.

[120] C. Hu, Y. Wang, G. Zhu, M. Lv, Y. Ru, S. Zhang, D. Gao, J. Xiao, *Plast. Sci. Technol.* **2022**, *50*, 83.

[121] X. Tian, T. Liu, C. Yang, Q. Wang, D. Li, *Compos. Part A* **2016**, *88*, 198.

[122] Y. Li, Y. Gao, Z. Tu, *Suliao* **2022**, *51*, 141.

[123] S. Hazer, M. Coban, A. Aytac, *J. Appl. Polym. Sci.* **2018**, *135*, 46881.

[124] Y. Abiko, T. Hayasaki, S. Hirayama, A. Almarasy, A. Fujimori, *Polym. Bull.* **2021**, *78*, 1585.

[125] A. Zanjanijam, S. Hakim, H. Azizi, *RSC Adv.* **2016**, *6*, 44673.

[126] L. Wang, W. Ma, S. Xiao, *Acta Polym. Sin.* **2014**, *2*, 255.

[127] F. Zhang, Y. Zhu, X. Du, Y. He, T. He, S. Jia, *China Plast. Ind.* **2022**, *50*, 77.

[128] Q. Zhang, H. Quan, J. Liu, D. Gao, S. Zhang, *China Synth. Resin Plast.* **2022**, *39*, 5.

[129] G. Urtekin, A. Aytac, *J. Polym. Res.* **2021**, *28*, 1.

[130] C. Hui, J. Pietrasik, M. Schmitt, C. Mahoney, J. Choi, M. Bockstaller, K. Matyjaszewski, *Chem. Mater.* **2014**, *26*, 745.

[131] Y. Fu, L. Liu, J. Zhang, *ACS Appl. Mater. Interfaces* **2014**, *6*, 14069.

[132] W. Li, Z. Xu, L. Chen, M. Shan, X. Tian, C. Yang, H. Lv, X. Qian, *Chem. Eng. J.* **2014**, *237*, 29.

[133] N. Mao, H. Jeong, T. Nguyen, T. Nguyen, T. Do, C. Thuc, P. Perré, S. Ko, H. Kim, D. Tran, *Compos. Part B* **2019**, *161*, 651.

[134] J. Rabanel, J. Faivre, S. Tehrani, A. Lalloz, P. Hildgen, X. Banquy, *ACS Appl. Mater. Interfaces* **2015**, *7*, 10374.

[135] C. Liu, S. Ye, J. Feng, *Compos. Sci. Technol.* **2017**, *144*, 215.

[136] D. Dreyer, S. Park, C. Bielawski, R. Ruoff, *Chem. Soc. Rev.* **2010**, *39*, 228.

[137] H. Zhou, H. Zhai, *J. Mater. Sci. Mater. Electron.* **2018**, *29*, 7893.

[138] M. Niu, H. Wang, J. Li, H. Chen, J. Chen, R. Soc, *Open Sci.* **2020**, *7*, 192154.

[139] W. Pinnavaia, *Appl. Clay Sci.* **1999**, *15*, 11.

[140] Y. Liu, Y. Liu, R. Yang, *J. Fire Sci.* **2021**, *39*, 495.

[141] F. Yemisci, S. Yesil, A. Aytac, *Fire Mater.* **2017**, *41*, 964.

[142] W. Mao, J. Du, T. Liu, *New Chem. Mater.* **2021**, *49*, 178.

[143] A. Sharma, A. Singholi, *Mater. Werkst.* **2021**, *52*, 635.

[144] C. Lantano, I. Alfieri, A. Cavazza, C. Corradini, A. Lorenzi, N. Zucchetto, A. Montenero, *Food Chem.* **2014**, *165*, 342.

[145] L. Qin, J. Qiu, M. Liu, S. Ding, L. Shao, S. Lü, G. Zhang, Y. Zhao, X. Fu, *Chem. Eng. J.* **2011**, *166*, 772.

[146] B. Wang, Z. Qi, X. Chen, C. Sun, W. Yao, H. Zheng, M. Liu, W. Li, A. Qin, H. Tan, *Int. J. Biol. Macromol.* **2022**, *217*, 792.

[147] F. Yang, X. Guo, Z. Zeng, J. Xiao, X. Dong, *Polym. Compos.* **2021**, *42*, 3125.

[148] E. Hasanpur, A. Ghazavizadeh, A. Sadeghi, M. Haboussi, *J. Mech. Behav. Biomed. Mater.* **2021**, *124*, 104768.

[149] S. Hsu, K. Hung, C. Chen, *J. Mater. Chem. B* **2016**, *4*, 7493.

[150] H. Samadian, S. Farzamfar, A. Vaez, A. Ehterami, M. Salehi, *Sci. Rep.* **2020**, *10*, 13366.

[151] M. Bernardo, B. da Silva, L. Mattoso, *J. Compos. Mater.* **2021**, *55*, 2289.

[152] M. Bernardo, B. da Silva, A. Hamouda, M. de Toledo, C. Schalla, S. Rütten, R. Goetzke, H. Luiz, M. Zenke, A. Sechi, *Sci. Rep.* **2022**, *12*, 1.

[153] J. He, X. Hu, J. Cao, Y. Zhang, L. Zhang, *Carbohydr. Polym.* **2021**, *253*, 117198.

[154] Z. Orafa, H. Bakhshi, S. Arab-Ahmadi, S. I. rani, *Sci. Rep.* **2022**, *12*, 1.

[155] J. Zhu, C. Zhang, R. Mei, X. Yu, L. Wang, H. Yu, *J. Wuyi Univ. (Natural Science Edition)* **2022**, *36*, 20.

[156] M. Hajikhani, Z. Emam-Djomeh, G. Askari, *Int. J. Biol. Macromol.* **2021**, *172*, 143.

[157] S. Yilmaz, A. Aytac, *J. Polym. Res.* **2022**, *29*, 473. [Correction added on December 23, 2022, after first online publication: Reference 157 has been updated]

[158] S. Yilmaz, A. Aytac, *J. Ind. Text.* **2022**, *51*, 8428S.

[159] Y. Xiang, A. Sw, A. Ccjy, C. Jlb, P. Xi, J. Lu, A. Kx, A. Zl, C. Xfb, Z. Qiang, *Sci. Total Environ.* **2021**, *807*, 151084.

[160] E. Vaňková, P. Kaparová, J. Khun, A. Machková, V. Scholtz, *PeerJ* **2020**, *8*, e10259.

[161] L. Wang, Y. Gao, J. Xiong, W. Shao, C. Cui, N. Sun, Y. Zhang, S. Chang, P. Han, F. Liu, *J. Colloid Interface Sci.* **2022**, *606*, 961.

[162] M. Güleçyüz, A. Mazur, C. Schröder, C. Braun, A. Ficklscherer, B. Roßbach, P. Müller, M. Pietschmann, *Arthroscopy* **2015**, *31*, 1134.

[163] B. Huh, B. Kim, S. Kim, C. Park, S. Lee, K. Kim, C. Yeong, Y. Choy, *Mater. Sci. Eng. C* **2017**, *79*, 209.

[164] M. Schrumpf, A. Lee, A. Weiland, *J. Hand Surg. Am.* **2011**, *36*, 1769.

[165] S. Liu, G. Wu, X. Chen, X. Zhang, P. Wang, *Polymers-Basel* **2019**, *11*, 1015.

[166] M. Ghozali, S. Fahmiati, E. Triwulandari, W. Restu, D. Farhan, M. Wulansari, W. Fatriasari, *Polym.-Plast. Technol. Mater.* **2020**, *59*, 1332.

[167] F. Velázquez, N. García-Caldera, P. Rosa, D. Martínez-Romero, J. Gabaldon, *Polymers-Basel* **2021**, *13*, 1399.

[168] L. Wang, J. Xu, M. Zhang, H. Zheng, L. Li, *Food Chem.* **2022**, *380*, 132022.

[169] S. Natesan, J. Samuel, A. Srinivasan, *Polym. Bull.* **2022**, *79*, 4627.

[170] S. Gumus, G. Ozkoc, A. Aytac, *J. Appl. Polym. Sci.* **2012**, *123*, 2837.

[171] H. Moustafa, N. Kissi, A. Abou-Kandil, M. Abdel-Aziz, A. Dufresne, *ACS Appl. Mater. Interfaces* **2017**, *9*, 20132.

[172] R. Xing, B. Yang, R. Huang, W. Qi, R. Su, B. Binks, Z. He, *Langmuir* **2019**, *35*, 12799.

[173] A. Tuteja, W. Choi, M. Ma, J. Mabry, S. Mazzella, G. Rutledge, G. McKinley, R. Cohen, *Science* **2007**, *318*, 1618.

[174] X. Wang, Y. Pan, H. Yuan, M. Su, C. Shao, C. Liu, Z. Guo, C. Shen, X. Liu, *Chin. Chem. Lett.* **2020**, *31*, 365.

[175] L. Ma, C. Abney, W. Lin, *Chem. Soc. Rev.* **2009**, *38*, 1248.

[176] L. Murray, M. Dincă, J. Long, *Chem. Soc. Rev.* **2009**, *38*, 1294.

[177] J. Li, R. Kuppler, H. Zhou, *Chem. Soc. Rev.* **2009**, *38*, 1477.

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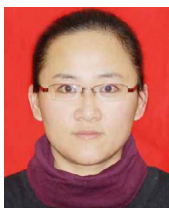
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