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Review

Recent advances in reinforced bioplastics for food packaging – A critical review

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ABSTRACT

Recently, diversifying the material, method, and application in food packaging has been massively developed to find more environment-friendly materials. However, the mechanical and barrier properties of the bioplastics are major hurdles to expansion in commercial realization. The compositional variation with the inclusion of different fillers could resolve the lacking performance of the bioplastic. This review summarizes the various reinforcement fillers and their effect on bioplastic development. In this review, we first discussed the status of bioplastics and their definition, advantages, and limitations regarding their performance in the food packaging application. Further, the overview of different fillers and development methods has been discussed thoroughly. The application of reinforced bioplastic for food packaging and its effect on food quality and shelf life are highlighted. The environmental issues, health concerns, and future perspectives of the reinforced bioplastic are also discussed at the end of the manuscript. Adding different fillers into the bioplastic improves physical, mechanical, barrier, and active properties, which render the required protective functions to replace conventional plastic for food packaging applications. Various fillers, such as natural and chemically synthesized, could be incorporated into the bioplastic, and their overall properties improve significantly for the food packaging application.

1. Introduction

Food packaging is a fundamental requirement for protection, containment, and convenience for packaged products. The synthetic polymer fits the requirement with characteristics of self-efficiency and simplicity. As a result, the production of conventional plastic reach nearly 359 million tons in 2018 [1,164]{, #3095}. The consumption surpassed the production in the same year as the other portions of plastics were also generated from 2010 to 2017 [2]. According to the latest information, nearly 45 % of the total plastics are utilized in the packaging market, while the remaining are used for manufacturing. Plastics are versatile and widely used for packaging and other applications due to their advantageous properties. For instance, plastics are lightweight, inexpensive, chemically inert, and have higher mechanical and barrier properties [3]. Other advantages of the materials are that they are easily thermal sealable, printable, and customizable into various shapes and sizes. However, their non-biodegradable and non-renewable nature overrides the properties due to the risk of environmental pollution. The usage cycle of packaging plastics is very short, and soon after usage, it is discarded. As per the latest report, only nine to 10 % of the total plastic is recycled [4]. Due to improper disposal management, the generated plastic waste ends up in an open ecosystem or is

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disposed of in a landfill or sea [5].

Recycling can be one of the approaches to reduce the impact of plastic pollution. This recycled plastic could meet the performance and essential quality requirements [6]. However, the recycling process requires thorough grading, washing, grinding, mixing, and extruding, which leads to prior degradation, limiting the advantages of the polymer recycling cycle [7]. Additionally, the acceptance and demand are still lower than virgin polymers due to the chemical impurities in different applications. Another approach could be the replacement of synthetic plastic with bioplastic. Consumers' awareness of conserving and protecting the environment has risen significantly to replace the largest possible portion of synthetic plastic with bioplastic. However, only 1 % of the total plastic produced is bioplastic, and the trend is continuously increasing in various industries. The adverse effect of synthetic plastic has also encouraged the use of bioplastic in different application areas, such as food packaging, medical, and pharmaceutical industries [8]. The surgical implants have been applied with the help of bioplastic as a scaffold and different drug delivery systems. In 2017, bioplastic production reached 2.06 million tons and was predicted to increase to >7.5 million tons in 2026 by 264 % [9].

Bioplastic is growing rapidly in every sector, particularly in food packaging applications such as biodegradable plates, cups, film, sheets, and cutlery, continuously replacing synthetic plastic with bioplastic [10,165]. Protein and polysaccharides have been widely explored for their potential utilization in the design and development of bioplastic due to their thin film-forming ability and unique functional properties, including their optical, mechanical, O_2 and moisture barrier, antioxidant, and antimicrobial properties [11,12]. The development and characterizations of bioplastic have been observed, but the bioplastics still lack sufficient tensile strength and moisture-barrier functions, which causes limitations in widespread application [13]. The bioplastic must be reinforced with different additives to extend its limiting properties, which could improve its mechanical and barrier properties [14,15]. Bioplastics' mechanical and barrier properties must be improved to protect food from the outside atmosphere, which contains moisture and mechanical hazards. This could be done effectively by incorporating various reinforcement agents, such as fillers, compatibilizers, plasticizers, nanoparticles, etc. [16]. Introducing micro- and nano-reinforcing materials into the bioplastic matrix has been observed to improve their mechanical and barrier properties to compete with synthetic plastic in the polymer market. The reinforcing agents are dispersed into the continuous phase of the film-forming solution, thus forming a compact tortuous pathway to integrated network forming with modification and improvement in functional properties in the bioplastic matrix. The reinforcement filler agents may differ in size, shape, proportion, distribution, and chemical nature, which must be considered when applied to bioplastic fabrication [6]. Lignocellulosic or cellulosic materials from agro-waste, such as fibers from cotton, garlic straw, rice husk, wheat straw, sugarcane bagasse, and coffee silver skin, have been extensively used as reinforcing agents in plastic development [17]. Micro and nanoscale reinforcing agents greatly enhanced the mechanical strength and thermal resistivity when uniformly distributed throughout the bioplastic matrix. The crystalline structure led to the development of the dense hydrogen network to form a torturous pathway that restricts the diffusion of gaseous molecules from the polymer matrix when utilized in packaging. Inorganic nanofillers such as MgO, silicon dioxide, and aluminum nanoparticles (Al NPs) work as the filling agents in the composites that enhance the overall functional properties and, moreover, possess antimicrobial activity, which are added benefits to the polymer composite [18].

Fig. 1 represents the bibliometric mapping of the reinforced bioplastic development for the food packaging application. Mechanical properties are mostly mentioned in the bibliometric network, which shows that bioplastics still encounter problems with mechanical properties. Bioplastics made from renewable resources are receiving more attention as a result of the growing need for environmentally friendly packaging options. With the addition of natural fibers, nanoparticles, or other reinforcing agents, reinforced bioplastics provide a special chance to get around some of the drawbacks of conventional bioplastics. Comprehending the latest advancements in this domain is vital in evaluating the practicability and efficacy of reinforced bioplastics in mitigating the obstacles linked to traditional food packaging materials. Further, the development process, impact on the composites, and different approaches to improving the performance of reinforced bioplastic for food packaging have been thoroughly overviewed.

2. Definition of reinforced bioplastics

Bioplastics are polymers produced from natural or renewable sources and can be both biodegradable and non-biodegradable. Bioplastic is not merely a single material; it comprises a whole group of materials with various properties and applications. According to European Bioplastics, any material is considered bioplastic if it is biobased, biodegradable, or features both properties [19]. Biobased refers to the material or products either fully or partly derived from biomass and biomass utilized to produce bioplastic, from potato, barley, banana, corn, sugarcane, etc. [20]. Biodegradation depends upon many factors, including environmental conditions such as temperature, moisture, humidity, material, and its application purpose [21]. The difference in biobased and biodegradability can be effectively understood by Fig. 2, which separates them based on their characteristics. The fabricated biopolymers cannot provide sufficient functional properties that could compete with conventional plastic in terms of barrier, mechanical, thermal stability, and flexibility. This could be solved by incorporating fillers or additives, which could disperse or dissolve in the bioplastic formulation and help fabricate enhanced mechanical and barrier bioplastic [22–24].

The common practice of reinforcement of the fillers into the packaging films is blending the filler with a biopolymer solution, where the filler is some polymer or material in the form of powder or solution that could be added to the bioplastic solution to overcome the shortcoming properties of the bioplastic. Adding fillers into the bioplastic formulation gives several options as different suitable materials that are better in mechanical properties and strong in reinforcement are available [25]. However, fillers in loose condition cannot perform the task directly for any purpose, so they must be blended or mixed with some polymer before forming the packaging films. So, these fillers or reinforce agents should be available in either particle or fiber form and have a high aspect ratio with efficient stress transfer potentiality [26]. Recent research and developments have better understood the bioplastic fabrication strategy to enhance the lacking properties. There are various reinforcement agents to gain comparable physicomechanical and barrier properties to replace synthetic polymers in various industries, particularly food packaging applications [25]. Utilizing the nanofiller in the bioplastic could greatly enhance the overall performance. The advances of different reinforcement agents have a wide variety in broad classes such as clays, organic, inorganic, and carbon nanostructured. The organic nanofiller such as also includes biopolymers such as chitosan and cellulose; at the same time, inorganic nanofiller is either metal (silver, gold) or metal oxides (ZnO, TiO_2) [27]. Sustainability in the development of reinforced bioplastic is one of the major concerns when selecting a reinforcement agent. Using suitable fillers could improve the degradability of the bioplastic, but it also depends upon the filler's biodegradability directly. So, biobased fillers are preferred over inorganic fillers to incorporate due to their sustainability concern of reducing the carbon footprint, preserving petrochemical resources, less cytotoxicity, and being less harmful to the environment and human beings if disposed to open areas [28]. The production of reinforced bioplastic involves various biopolymers and fillers, which are compatible or incompatible. The compatibility of the reinforcement fillers is also a major factor in selecting the fillers to incorporate in the bioplastic, which affects the overall performance and the sustainability of the packaging material. Blending two or more incompatible biopolymers

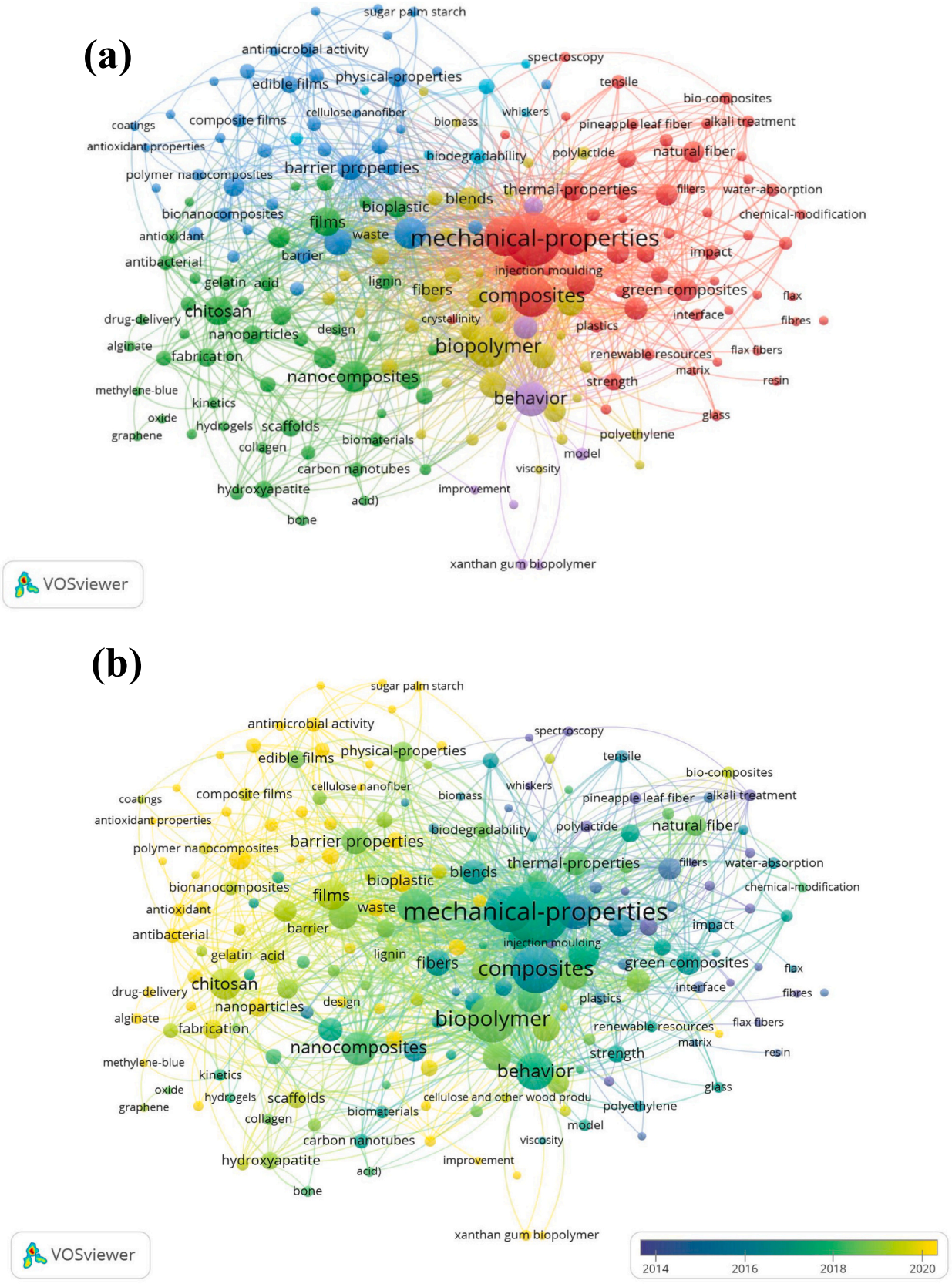

Fig. 1. A bibliometric network map of scientific research on reinforced bioplastics during the previous five years through VOS viewer is gathered data retrieved through Scopus and Web of Science with the 'reinforced bioplastic' and 'reinforced biopolymer' from 2014 to 2020. Clusters (A) indicate the co-occurrence of the keywords reinforced bioplastic and reinforced biopolymers, and an overlay visualization (B) Specifies a period of the occurrence of the keyword from 2014 (blue) to 2020 (yellow).

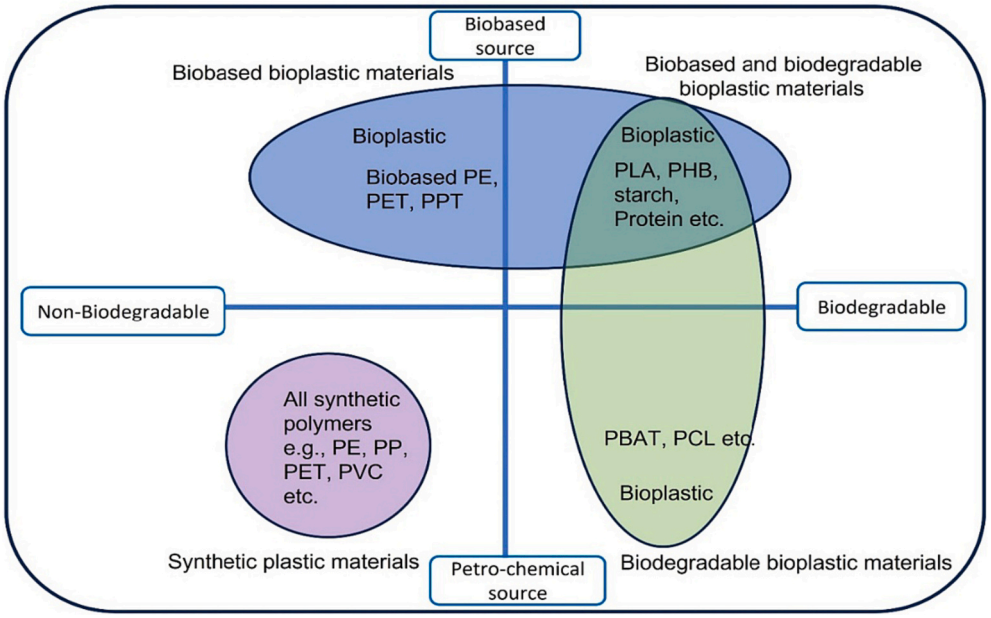

Fig. 2. Classification of bioplastic concerning synthetic, biobased, biodegradable polymers (fossil fuel and biobased) according to European bioplastic (Lindström & Österberg, 2020).

and the filler results in poor interfacial adhesion, consequently leading to worse physicochemical properties of the bioplastic. Here, the addition of compatibilizer into the polymer formulation is necessary to blend the composite properly and could be beneficial to overcome the combability issue of the reinforced bioplastic. However, adding many components could affect the matrix properties later, and it can cause major performance deficiency in the reinforced bioplastic products [14].

These issues are related to the lack of a deeper understanding of the reinforcement filler and the biopolymers. Numerous combinations of various materials may be used to create reinforced bioplastics; thus, additional research on the interfacial characteristics of bioplastics is needed to understand how different reinforcing materials interact with bioplastics. Ongoing research and development are required to explore further possibilities for reinforced bioplastic applications in many

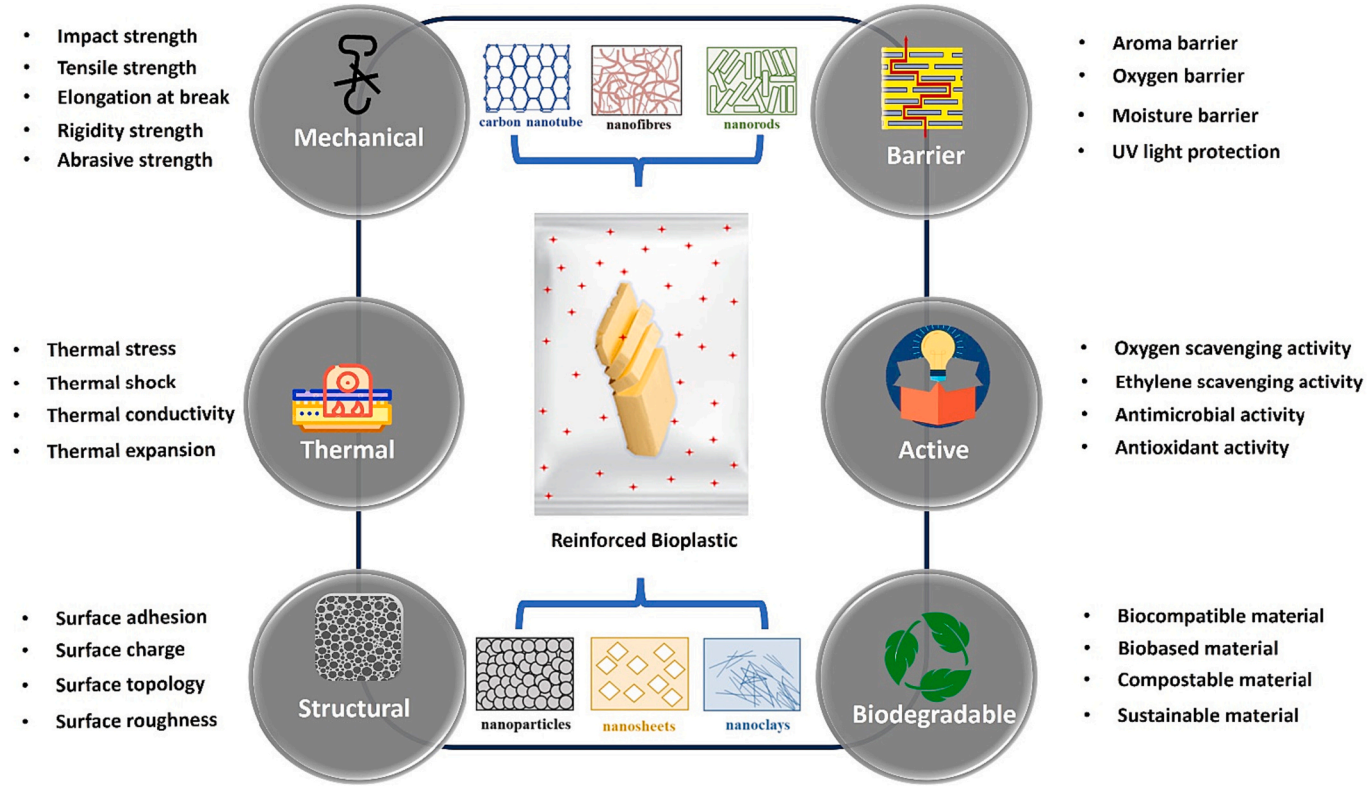

Fig. 3. The effect of several nanofillers reinforcement in the bioplastic with respect to the physical, mechanical, barrier, thermal, morphological, active functions, and biodegradability.

sectors. The application of different bioplastic and reinforcement fillers is shown in Fig. 3, which shows the effect of reinforcement filler on the bioplastic packaging materials.

3. Reinforced bioplastic production methods

Incorporating different fillers into the bioplastic composite had several challenges, such as their thermal stability, weak interfacial force between fillers and film matrix, solubility, and dispersion behavior of the filler. In filler-reinforced bio composite, a polymer matrix holds the filler strongly, which helps to distribute the load evenly, and fillers work as primary load-bearing substances [24]. To work well with the composite, both phases must be well-bonded. Another issue with the filler and bioplastic is that they have distinct chemical structures, and the filler dispersion is important for composite fabrication. The filler's chemical nature affects the compatibility with the polymer matrix. Various filters have undergone different surface treatments to overcome the interfacial issues and offer their smooth blending property with the polymer, such as physical, chemical, and biological, as illustrated in Fig. 4 [14]. The methods commonly used for composite fabrication are discussed below.

3.1. Solution casting

Solution casting is the oldest and most convenient method to develop packaging film and was discovered in the 19th century by Eastman Kodak to make plastic films. It is a versatile process to fabricate thin films or sheets at small scale or laboratory experiments. The polymer solutions of different polymers are dissolved in suitable solvents, either aqueous or non-aqueous volatile solvents, occasionally reinforcing various additives such as micro or nanosized materials before being cast on the flat surface. Then, the solvent phase is evaporated with different drying methods, and the dried thin film is casted-off from the substrate [29]. Cassava starch-based composite was developed using crude kaolin clay through solution casting. The reinforcement of the fillers diminishes the solubility and water diffusivity for the control film. The hydric kaolin caused the free volume in the starch macromolecule composite matrix due to the interaction with clay platelets. The microbial proliferation was increased due to the incorporation of clay particles, which improved biodegradability properties [30]. In another study, yam starch-based bio nanocomposite was developed via solution casting with the reinforcement of bentonite at 0.5 to 1.5 %*w*/w. The micrographs through SEM analysis showed uniform bentonite dispersion in the composite. The FTIR analysis was observed and depicted that the bentonite inclusion in the composite had strengthened the O—H and Si-O-Si bond, resulting in higher mechanical strength. The high bentonite percent bioplastic was found to be the maximum soil degradation rate, which might be due to the silicate layer of bentonite in the bioplastic matrix. The intercalated bentonite in the bioplastic matrix improves the composite's barrier property to moisture and O_2 [31]. In another study, polylactic acid was blended with polyhydroxy butyrate (PHB), and its blends were reinforced with cellulose nanofibrils to evaluate and compare their physicomechanical properties. The mechanical strength of the polylactic acid (PLA) and PHB was significantly higher than that of the reinforced biopolymer. This could be due to the better interaction of PLA and PHB, as shown in the SEM analysis. In contrast, the cellulose nanofibers (CNFs)-inforced composite found cracked space [32].

3.2. Melt extrusion

The raw material is transformed into a substance with a distinct structure and textural qualities throughout the extrusion process. The combination of diverse forces in the extrusion, such as shear, temperature, and pressure, causes the raw material's melting at low/high

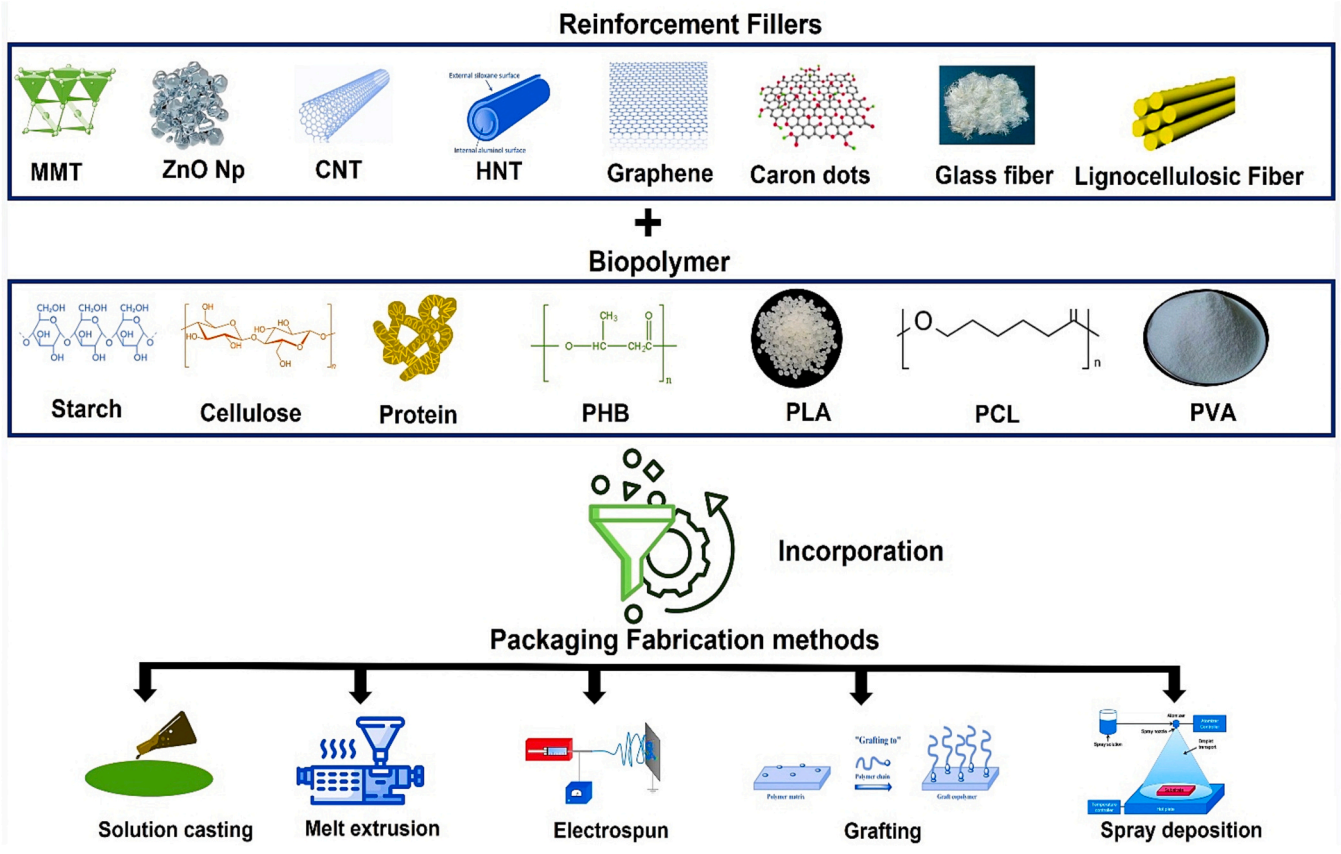

Fig. 4. Different methods of filler reinforcement into the bioplastic.

moisture content to expand and gelatinize. Extrusion released during expansion forms the liquid phase containing biopolymers, which leads to the construction of the molded structure of the film or package container [33]. The polycaprolactone (PCL) was reinforced with macaiba fiber (MF) at 10 to 20 % through the melting extrusion process. It was added naturally and modified chemically with maleic anhydride (MA), sodium hydroxide (NaOH), and methacryloxypropyltrimethoxysilane (silane). The biocomposite's mechanical strength and thermal stability were the lowest with higher MF and the chemical treatment MA. The hydrophobicity of the developed film decreased after adding the MF to the PCL as it caused more surface interaction with the water and eased the water molecules' entrance [34]. In another study, PLA and polybutylene succinate (PBS) blends were reinforced with the mineral filler of calcium carbonate up to 30 %. The results showed that the higher concentration of Calcium carbonate attains a greater crystalline fraction, which further decelerates the water absorption. The composite's thermoforming ability has a high stretching ratio to the thermoforming process due to higher thermal diffusivity promoted by calcium carbonate, which leads to the lowered thermal and mechanical energy needed during the product's manufacturing [35].

Moreover, the filler reinforcement caused an increased melting flow index and made it more precise in the thermoforming development process of the containment during the molding process [36]. In another study, a twin-extrusion process was applied to develop the reinforced thermoplastic starch for the food packaging application with the reinforcement of bamboo pulp nanofiber. The tensile strength of the film attends at 5.07 MPa with the nanofiber. The water vapor transmission rate was decreased, which is desirable for the food packaging application, and 80 % biodegradability has been shown within 60 days [37].

3.3. Electrospinning

The assembly of two highly sophisticated methods, electrospray and spinning, is made using electrospinning (electro + spinning). When a high electric field is applied to the melted fluid or solution from the die tip, it also acts as an electrode due to high field energy. The resulting formation coming out as an ejected charged jet from the die tip toward the counter electrode leads to continuous fiber formation [38]. The production of the thinnest fibers, possibly up to the nanometer range, with large surface area, superior mechanical properties, and the capability to produce three-dimensional (3D) renders the ease of functionalization for various applications. Electrospinning has often been utilized in biomedical and tissue engineering, using natural bioplastics such as protein, polysaccharides, and lipidic formations [39]. Biopolymers are preferred for active and intelligent food packaging due to their functional properties, ability to carry active agents, and controlled release. The active and intelligent agents from different natural sources are thermally sensitive and get evaporated while drying the incorporated film due to its high volatility. Electrospinning can be an option to overcome this problem with electrospun fiber encapsulation. The above factors increased interest in using electrospun fiber in food and packaging industries [40]. Electrospun fibers are applicable in food industries in many ways, such as reinforcement agents for eco-friendly packaging, emulating elements for artificial foods, scaffolding for cell cultures, encapsulation of enzymes, vitamins, and antimicrobials, etc. [41].

Pectin was dissolved in water and combined with a small amount of polyethylene oxide 2000 (PEO2000) for electrospinning. It produced extremely thin fibers with a high pectin content; nevertheless, because of its low heat stability, annealing produced weak, porous, and black films. Glycerol and polyethylene glycol 900 (PEG900) were added to improve things. Strong, transparent films were obtained by pairing dichloromethane washing with optimal annealing at 150 °C for one minute at a load of 12 kN. Using electrospinning, these films of pectin were used as an interlayer between two layers of poly(3-hydroxybutyrate-*co*-3-hydroxyvalerate) (PHBV). The end product was a multilayer film that was entirely biobased and biodegradable and had improved limonene and water vapor barrier qualities [42]. Electrospun nanofibers are better in mechanical properties. They can aid the biopolymers in strengthening the functional properties of the packaging films as reinforcing fillers.

3.4. Coating deposition

The coating is applied to the thin film or substrate polymer using a continuous layer deposition. These coatings work as a functional layer or a layer to incorporate active ingredients. Several well-known techniques include spraying, dipping, casting, chemical vapor deposition, physical vapor deposition, and screen-printing coating. Spray When active compounds are blasted through a high-pressure nozzle and deposited onto the surface of a thin film to produce a coating, the process is known as coating or deposition. Dip coating involves immersing the thin film in the coating solution and allowing it to dry. Cast coating, composed of active substances dissolved in a suitable solvent, is applied to the film surface and allowed to dry until the solvent completely disappears. The coating technique is influenced by the adsorbing agents' and substrate's physical and chemical makeup [43]. The titanium dioxide with and without dopamine was sprayed on the poly (vinyl butyral) (PVB) composite with electro-spraying. Using non-coated TiO_2 particles, nanoindentation tests reveal that the final hierarchical composite material has a hardness of 0.75 0.04 GPa and Young's modulus of 29.5 1.0 GPa. Compared to composites with uncoated TiO_2, the ones with polydopamine-coated TiO_2 particles exhibit increases in hardness and Young's modulus of around 40 % and 25 %, respectively [44]. A different method of incorporation of fillers into bioplastic development and their impact has been thoroughly listed in Table 1.

3.5. Compression and injection molding

A thermoplastic polymer is heated in a cylindrical chamber during the injection molding process for the creation of biopolymer composites, which encourages material flow. The molten substance is pumped into a mold with temperature and pressure carefully regulated. The material is pressed into the cold mold by hydraulic pressures that are assisted by a plunger and ram. A reciprocating, revolving screw then compresses and melts the material. When the screw moves backward, indicating that the cycle is ready to begin, the formed biopolymer composite is taken out of the mold [45]. Compared to solution casting, compression molding provides a faster forming process, which makes it an intriguing technology for producing biocomposite products. In addition, this molding method is among the least expensive when compared to other methods like injection molding, etc. When molding using pricey materials, this method reduces waste and is economical. Compared to injection molding, compression molding is more suited for natural fiber-reinforced polymer biocomposites because it can produce complicated components at high pressure and in huge quantities [46]. Injection molding was used to combine sodium alginate with starch, a biopolymer with poor mechanical strength and significant water absorption, in cassava starch. The final composite showed good phase compatibility, which resulted in improved mechanical characteristics [47]. Natural polymers like proteins and polysaccharides are widely used because of their many functional characteristics and intermolecular bonding. Film fabrication is the main application for this approach; in particular, chitosan films crosslinked with citric acid were created effectively [48].

4. Reinforced materials to improve the bioplastics' properties

The reinforcement of the bioplastic is done with the nanofillers, such as organic and inorganic, as categorized in Fig. 5, to improve the overall performance of the composites. The Fillers are generally inexpensive, which increases the cost-effectiveness of filler-reinforced biocomposites. The creation of newer polymeric materials with more fascinating

Table 1
The reinforced bioplastic for food packaging application with different types of filler and their effect on the mechanical, barrier, morphological, and biodegradability.

Bioplastic base matrix	Reinforced materials	Method of production	Proportions	Mechanical and barrier properties	Physical and morphological properties	Biodegradability	Reference
Cassava starch	Coconut fiber	Melt mixing	5–30 %	The coconut fiber reinforced composite was stronger than the control sample, from 3.24 MPa to 112.68 MPa.	The SEM analysis showed uniform and smooth fractured surface as well as good adhesion of the fiber on the composite cross-sectional view.	–	[49]
Carrageenan	Microcrystalline cellulose	Solution casting	1–3.5 %	The reinforcement of microcrystalline cellulose (MCC) impacts the tensile strength. It causes 20.74 MPa and EAB to have 35.12 %, respectively, for 1 % and 3.5 % MCC, causing a reduction in the tensile strength of 5.16 MPa and EAB of 15.15 %.	–	–	[50]
Cellulose	Cocoa pod husk and sugarcane bagasse	Solvent casting	25–75 %	The addition of fibers improves the moisture barrier for bioplastic development, and 75:25's lowest barrier properties were observed.	–	–	[51]
Corn starch	Sugarcane bagasse cellulose fibers	Extrusion	12 %	Increased Tensile strength and Elongation at break. The injection molding prepared sample showed higher tensile and elongation at break than the sample by compression.	The composite with sugarcane bagasse fibers is arranged in stiff bundles due to the strong bonding of many components, such as cellulose, hemicellulose, lignin, and waxes.	TPAS/SF composite with SSE and TSE processed with extrusion after seven days recorded weight reduction between 26 and 38 %.	[52]
Corn starch	Coffee and rice husk	Melt blending and compression molding	1–10 wt%	Cellulosic fibers from rice and coffee husks improve the tensile properties but not significantly, even though the film's extensibility was deprived due to adding fibers for 5 and 10 wt% than the control starch film.	A rough surface was witnessed in the composites due to the fiber reinforcement. An irregular surface with less gloss was observed than the starch control composite.	–	[16]
Corn starch	Parthenocarpic date palm fiber	Solvent casting	75–100 %	–	The SEM analysis of the bioplastic showed no uniform surface with a cracked appearance and weaker mechanical strength.	Soil microflora was used to assess biodegradability, and it shows that the bioplastic completely decomposed and lost its shape at the end of the test.	[53]
Keratin	Microcrystalline cellulose	Solution casting	2 %	–	Morphology of the keratin with cellulose showed a compatible uniform surface without cavities, edges, and holes.	–	[54]
Polylactic acid	Corn fibers	Injection molding	20 %	UCF/PLA showed a tensile strength of 39.8 MPa and a tensile modulus of 4.97 GPa. The alkali treatment of the bioplastic showed improvement in the mechanical properties up to 49.5 MPa and tensile strength of 6.63 GPa.	SEM observation found no crevices on the fracture surface of ACF/PLA.	–	[55]
Polylactic acid	Snail Shell Nanoparticles and Sugarcane Bagasse Cellulose	Solvent casting	0.5 to 1.0 %	–	The FESEM morphologies showed uniform dispersion in the bioplastic matrix, leading to a barrier to water absorption.	–	[56]
Polyvinyl alcohol	Cellulose nanocrystals	Solution casting	0–20 %	Optimum tensile strength was observed at 10 % of cellulose nanocrystals (CNCs), and the reduction of moisture uptake by 21 % was noticed at 20 % of CNC loading.	Well-isolated and dispersed whisker loading was observed in the composite on the nanometer scale.	–	[57]

(*continued on next page*)

Table 1 (*continued*)

Bioplastic base matrix	Reinforced materials	Method of production	Proportions	Mechanical and barrier properties	Physical and morphological properties	Biodegradability	Reference
Polyvinyl alcohol	Cellulose nanocrystals and snail shell-reinforced	Solvent casting	0–5 % (*w*/w)	The lower concentration of CNC of 2.5 % with 7.5 % of PVA had shown optimum tensile strength of 33 MPa and 35 MPa Young's modulus. About 2.5 wt% of SSN exhibited the superior tensile strength of 33 MPa and 35.4 MPa Young's modulus.	The morphology showed a rod-like appearance with dimensions of 100–200 nm in length and 10–20 nm in width. SSN has a size of 25–65 nm and has semi-sphere morphology.	–	[58]
Polyvinyl alcohol	Pinewood nanocellulose	Solution casting	1.5 %	With the addition of nano cellulose, citric acid, and orange peel extract, the tensile strength and elastic modulus have raised from 317.11 kPa, 31.71 kPa up to 36.12 kPa, 1203.83 kPa, and 1708.54 kPa and 40.86 kPa respectively.	SEM analysis showed a smooth surface with an improper blend of nanocellulose.	The degradation test degraded 86.27 % for neat film and 77.27 % and 78.44 % for citric acid and orange peel extract, respectively.	[59]
Sago starch	Bentonite	Solution casting	1 to 3 % (w/w)	Tensile strength rises from 0.3276 Pa at 1 % BEN to 0.4412 Pa at 3 % BEN. Elongation at break (%) decreased (149.72 % to 73.93 %) with increased bentonite content.	Non-reinforced starch biofilm shows void and vacant spots with homogenous surfaces. With increased BNT, the vacancies were filled, so the voids in SEM were reduced.	The biodegradability rate increased with higher bentonite content in the film. The pristine layer silicate intercalated BEN was well distributed in the TSS composite matrix, leading to readily and rapidly degrading fragments.	[60]
Starch	Silica	Solution casting	0–5 %	The silica-reinforced starch film TS increased from 0.53 to 0.75 MPa, and elongation at break improved to 0.16–0.28 %.	–	The bioplastic degradation was slower after adding the silica as a reinforcing agent.	[61]
Starch/ polyvinyl alcohol (PVA)	ZnO nanoparticles	Solution casting	0–3.75 %	The addition of ZnO nanoparticles to the film caused a reduction in the tensile strength, reducing the elongation by 1.75–16.92. The moisture barrier property Improved.	The inclusion of the ZnO has uniform whisker morphology with an aspect ratio and a thickness of $<$100 nm.	–	[62]
Starch	Cellulose nanofiber/ MMT	Solution casting	1–7 %	The mechanical strength improves by 33 % with 3 % cellulose nanofibers (CNFs) loading and Young's modulus from 2388 to 3173 MPa. It also reduces moisture uptake by 13 %.	A smooth morphology section has been observed with CNF/MMT uniformly distributed throughout the composite.	–	[63]
Taro starch	Bentonite	Solution casting	0.5 to 2.5 % (*w*/w)	The film's tensile strength increased with a higher concentration of bentonite up to 48.48 $\pm$ 8.32 MPa. But beyond 2.5 % of bentonite, the tensile strength decreased for 3 % reinforcement. WVTR decreased with increasing bentonite content for the neat film from 9.22002 $\times$ 10–6 kg/m^2/ day, whereas for 2.5 % of bentonite, 8.39175$\times$ 10–6 kg/m^2/ day.	Surface morphology shows homogeneity and no phase separation, void, or cracks. The surface roughness was decreased as the bentonite increased due to the interaction between negatively charged bentonite and positively charged starch.	The neat film showed a higher rate of biodegradation (4 days, 50 % rapid reduction) than the film with bentonite reinforced (10 days).	[64]
Yam starch	Eggshell	Solution casting	2–3 % (*w*/w)	TS increased from Without filler, the bioplastic shows lower tensile strength of 0.65 $\pm$ 0.15 MPa, whereas it increases with the incorporation of eggshell from 2 to 3 % up to 0.97 $\pm$ 0.06 MPa. WVTR diminished with increased filler in the bioplastic by making a hydrophobic surface due to the eggshell.	The neat film showed a granular oval and spheroidal rounded structure. Agglomeration has been observed with the eggshell due to a high amount of $CaCO_3$.	The neat film was degraded by about 80 % in 21 days. With the presence of the eggshell, the biodegradability decreased up to 56 % in 21 days.	[65]

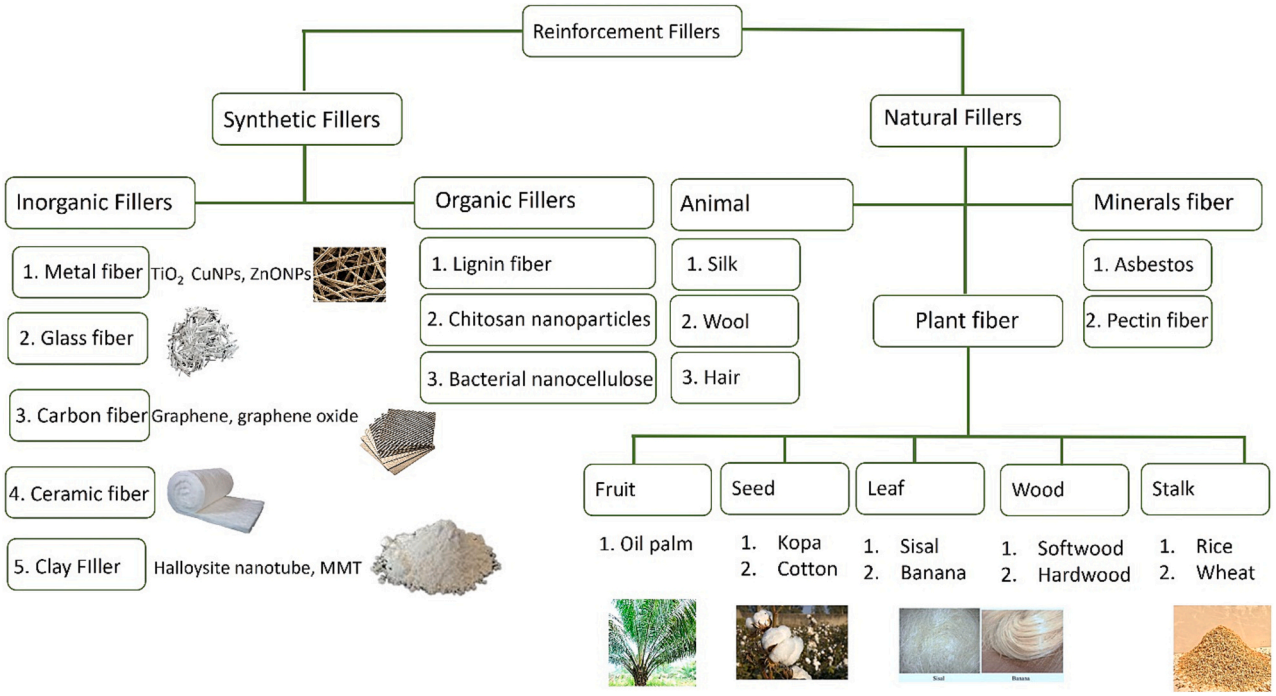

Fig. 5. Classification of reinforcing filler into the bioplastic to improve the overall performance for food packaging application.

features is made possible by these biocomposites, which is significant.

4.1. Organic and natural fillers

Cellulose is an example of an organic nanofiller. At the nanoscale, there are three forms of cellulose: cellulose nanocrystals (CNCs), cellulose nanofibers (CNFs), and bacterial nanocellulose. In recent years, research has been dominated by lignocellulosic fiber reinforcement [168]. Lignocellulosic fiber (LCF) is a natural fiber made from plants. These comprise water-soluble compounds such as cellulose, hemicellulose, lignin, and pectin. The amount and content of these components vary depending on the type of biomass [66,67]. As a result, they exhibit different mechanical behavior. CNCs are acicular cellulose particles with 2 to 20 nm diameters and lengths from 100 nm to several micrometers. CNC particles are made entirely of cellulose and are highly crystallized, with crystalline zones ranging from 54 % to 88 %. The supply of cellulosic materials, preparation circumstances, and experimental procedures influence the degree of crystallinity, dimensional diversity, and morphology. Fibers are abundantly available and have low-density abrasiveness, and high mechanical strength, which encourages the utilization of lightweight composite matrix as reinforcement agents in the packaging matrix [68]. Additionally, it enhances the characteristics of biopolymer composites; LCFs have been demonstrated to increase end-use biodegradation properties of the packaging material [14].

There is a rich supply of lignocellulosic fibers widely available in the market, including kenaf, jute, and sugarcane, as well as high-lignocellulosic biomass such as bagasse, empty oil palm fruit bunches, and rice straw. In addition to strengthening bioplastics, using these materials can reduce the cost of purchasing expensive materials while saving the environment through increased biodegradability and reuse of agricultural waste [69]. Another example of an organic nanofiller is chitosan. It is produced by deacetylating chitin, present in shell fragments and the skeletal components of insects and crustaceans. It is tempting to use it as filler because it is biodegradable and fulfills the goal of reusing waste. It is also known for its antibacterial properties [70]. TPS/chitosan film has been reported to inhibit the growth of *S. aureus* and *E. coli*. The decrease in microbial activity was more dramatic at higher concentrations of chitosan.

On the other hand, chitosan is a naturally occurring hydrophobic and non-toxic polymer found in the exoskeletons of shellfish such as shrimp, crabs, and lobsters. According to Tan et al. (2022), the nature of chitosan is suitable for use as a reinforcing material in solving the shortcomings of bioplastics, especially for improving the mechanical properties while simultaneously increasing the hydrophobicity to increase the resistance of bioplastics to water [71]. The properties of PLA nanocomposite films following extrusion with CNCs produced from CS are being studied. After being introduced at 3 %, CNCs were shown to contribute significantly to the mechanical and protective characteristics. Increased CNC concentrations aided congregation while weakening reinforcing effects. CNC processing improved the crystallinity of the PLA matrix while mechanical flaws were addressed. The addition of waste CS to PLA films increases their industrial usability. PLA/CNC bio-nanocomposite sheets, which have the potential to be innovative food packaging materials, were produced [72]. In a similar study, through solutions casting homogeneous PHB films with a nano cellulose concentration of 2–6 %. The presence of CNC in the bio nanocomposites was confirmed by FTIR and XRD studies. The DSC graphs demonstrated changes in melting conductivity and increased layer thickness due to the CNC incorporation. During cooling, the presence of CNC increased the production of PHB nuclei. Despite a slight decrease in thermal stability due to CNC, all materials remained unaffected until 200 °C. The addition of 6 % CNCs produced significant improvements in mechanical characteristics, including a 50 % increase in Young's modulus and a 35 % increase in tensile strength. We found 6 % CNC concentration as the best equilibrium after investigating the link between UV resistance and other barrier qualities [73].

Clay minerals such as Halloysite nanotube (HNT), zeolite, hectorite, Montmorillonite (MMT), bentonite, and cloisite have the potential to improve various properties and characteristics like higher uniform dispersion and higher aspect ratio, which are required for advantageous application of these mineral clays into polymer matrix development. Clay minerals are increasingly used in food packaging because they are mechanical, chemical, barrier (against oxygen, carbon dioxide, UV, moisture, and volatiles), thermally stable, and biocompatible. Using clay-based food packaging has various advantages over traditional tidy polymer packaging. Nanoscale clays render several advantages,

including enhanced transparency, toughness, gaseous, moisture, odor barrier, puncture resistance, abrasion and flex cracking, heat stability, and neutral to fat, grease, and oil [74,75].

4.2. Inorganic and synthetic filler

Chemical synthesis produces synthetic artificial fibers, categorized as organic or inorganic, based on their chemical makeup. Inorganic synthetic fibers include, but are not limited to, glass, carbon, metal, and ceramic fibers. These fibers are made from a variety of minerals, including silicon boride, silicon carbide, silicon nitride, graphite, alumina, aluminum silicate, zirconia, boron, boron carbide, and boron nitride [76]. Lightweight structural materials with high strength and modulus values that may be customized to certain loading situations have been made using inorganic fiber composites. They have a greater melting point than conventional fibers and are stronger, stiffer, and more heat resistant. Metal oxide nanoparticles such as zinc oxide (ZnO NP), titanium dioxide (TiO_2 NP), silicon dioxide (SiO_2 NP), aluminum oxide (Al_2O_3 NP), cerium oxide (CeO_2 NP), iron oxide (Fe_2O_3) can be used as active nanofillers to the biopolymer films [77]. TiO_2 NPs and ZnO NPs exhibit photocatalytic antibacterial activities due to reactive oxygen species (ROS) production upon UV exposure. Swarup Roy et al. demonstrated that Melanin was used as a reducing and stabilizing agent in the synthesis of AgNP. Carr-based nanocomposite films include AgNP. The optimal synthesis parameters for time, temperature, $AgNO_3$, and Melanin concentration were identified. The size range of AgNP is 10–50 nm, as validated by EDX and XRD. Mel interacts with AgNPs, as shown by FTIR.

Nanocomposite films have enhanced mechanical strength, thermal stability, and UV barrier properties. Antimicrobial activity was shown against Gram-positive and Gram-negative microorganisms. Use in active food packaging for increased shelf life and food safety. Although there was little migration of AgNPs into food, more research is needed [78]. In another study, TiO_2 glycerol-reinforced starch-based yarn film was produced and described in this work. Glycerol and TiO_2 concentrations were investigated for their influence on film characteristics. The concentration of glycerol has minimal influence on tension and elongation. TiO_2 was applied consistently, which increased tensile strength and elongation. Tensile strength (4.68 MPa) and elongation (32.96 %) were improved using the Response Surface technique. The experimental data (4.45 MPa, 37.77 %) agreed with the model predictions. Biodegradability and antibacterial characteristics were evaluated, with 29 wt% breakdown in 15 min and a 3.25 mm *E. coli* inhibitory zone for TiO_2-reinforced films [79]. The biodegradation was evaluated for Cast cassava starch/chitosan bioplastic films with different ZnO nanocrystals. Bioplastics containing ZnO degraded quickly in soil and ocean. Chitosan-containing bioplastics decomposed in saltwater and on land in 21 days, but chitosan-free bioplastics lasted 28 days on land and 14 days in seawater. ZnO crystalline phase boosted tensile strength and worked as an antibacterial agent, improving food shelf life. ZnO can potentially increase the durability of starch and starch/chitosan-based food packaging by acting as an ecologically friendly bioplastic [80]. In another investigation, SiO_2 nanoparticles were introduced into PBAT/TPS films through extrusion-blown film, resulting in high permeability and enhanced strength. The SiO_2 dispersion reinforced the matrix and enhanced tensile strength by up to 40 %. SiO_2 reacted with PBAT and TPS in distinct ways, enhancing surface roughness and inhomogeneity. The micropores allowed the flow of gases and water vapor by up to 39 % and 16 %, respectively, making them appropriate for breathable agricultural product packaging. Linear regressions accurately predicted higher OP and WVP levels. The polarity of the simulant, the microstructure of the matrix, and the nanoparticle-matrix-simulant interaction all influenced the migration of film components. The findings encourage commercializing permeable, robust bioplastic sheets for agricultural applications [81].

4.3. Other fillers

Some fillers, like semiconductor quantum dots gaining popularity, are used to improve optical properties and photostability. Because of their strong oxidation and agglomeration potential, increasing the compatibility and stability of this type of nanofiller in biopolymers is desirable. Graphene quantum dots (GQDs) embedded in biopolymer films are a viable candidate for optoelectronic applications due to their availability, biodegradability, cheap cost, and low production cost [82]. Due to their superior mechanical qualities and rapid electron mobility, materials based on graphene have become more and more common. Graphene has a higher surface area than other carbon-based nanomaterials (such as carbon nanotubes), which may facilitate interactions with the polymer matrix. Because it aggregates less than pure graphene, graphene oxide (GO) has the most potential as a nanofiller among the graphene family of materials. The inherent zero-band energy of GO and its limited solubility in organic and aqueous solutions are the most significant limitations to its use [83]. The chemical functionalization of graphene oxide changes it, which expands the possibilities of its possible applications. Using a simple solution casting approach, organically produced NCDs were effectively added to the chitosan-PVP film and orange peel extract as a taste in this work. Using NCD as a reinforcing agent enhanced the mechanical characteristics of the film, resulting in greater tensile strength and elongation rate. The novel combination of NCD and orange peel extract enhances the film's characteristics even more. Biodegradability and plant growth were tested, and the results showed that the qualities were superior to chitosan/PVP. This chitosan-PVP film with NCD and orange peel has the potential to be a greener alternative to synthetic plastic films. The antibacterial activity of the generated films will be studied further in the future [84]. In another study, a solvent was used to cast films made of thermoplastic potato starch and an antioxidant chitosan matrix. A food packaging material with active properties and electrical conductivity was created by altering the polymer mass fraction and adding 25 % wt% Reduced graphene oxide (rGO). The bio nanocomposite with the maximum hydrophobicity (>100°), mechanical performance, water resistance, and electrical conductivity was made of 75 % starch, 25 % chitosan, and integrated rGO. In both water and ethanol, starch-based films' antioxidant activity was enhanced by chitosan and rGO. In food packaging, an electrically conductive nanocomposite might replace non-biodegradable polymers, enabling in-pack food sterilizing and enhancing food safety [85]. The recent previously reported investigations on bioplastic development with the incorporation of bioplastic with a different method, concentration, and their impact on the properties of the packaging have been listed in Table 2.

5. Properties of reinforced bioplastics for food packaging and its factors influencing

Generally, the nature of the raw materials, the components and formulation, and the processing conditions and parameters should be the dominant factors governing the properties of bioplastics [107]. Despite the rapid development of bioplastics, however, the properties of bioplastics as a whole are usually inferior to their synthetic counterparts [108]. In practice, biopolymer blends and composites reinformed by fillers are usually used to enhance the overall properties of bioplastics [109]. The fillers involve various types, including inorganic fillers (e.g., calcium carbonate, nano-clay), organic fibers from plants [52], as well as carbon nanotubes (CNTs) [110]. Adopting biopolymer blends and incorporating fillers can impart bioplastics with better mechanical properties (such as stiffness and tensile strength), barrier properties against gas and vapor, and thermal stability as documented in Tables 1 and 2 [111]. In addition, as with conventional plastics, incorporating plasticizers or compatibilizers can help overcome brittleness and increase the miscibility of blended polymers to stabilize the materials. Especially for bioplastics, their mechanical properties, water resistance,

Table 2
The reinforced bioplastic with different reinforcement filler and their concentration, method of application, active functions, and barrier properties.

Reinforced materials	Base materials	Methods	Concentration	Mechanical strength	Moisture barrier	Active function	Reference
Alfa fiber	Polylactic acid (PLA)	Injection molding	20 wt%	TS- 66.56 MPa, TM- 3030.97 GPa	–	–	[86]
Arrowroot fiber	Arrowroot starch	Solution casting	0–10 %	–	9.45 × 10–10 ×g. $s^{-1}.m^{-1}$. Pa^{-1}	Antibacterial against *B. subtilis B29* and *S. aureus*	[87]
Cellulose nanocrystals	Cellulose	Solvent casting method	CNC (1–8 %)	Tensile strength- 59.38-85.29 MPa Tensile modulus- 3041 MPa	–	–	[88]
Coconut shell powder	Polylactic acid (PLA)	Compression molding	0–60 php	TS- 55 MPa TM-3500 MPa	–	–	[89]
Halloysite nanotube	Chitosan	Solution casting	0.25 %	TS-4.76 to 14.76 MPa EB- 32 %	2.46 10–6 g mm/ mm^2 day KPa	Antioxidant activity	[90]
Halloysite nanotube	Carboxymethyl cellulose	Solution casting	1 % *w*/w	2.91–7.06 MPa EB- 31-60 %	5.57 mg/cm^2/ day	Antioxidant activity	[91]
Montemorillonite (MMT)	Alginate	Casting	3 % *w*/w	–	–	Antimicrobial and antifungal activity	[92]
Polysaccharide Microfibre	Macroalgae	Solution casting	2–8 wt%	TS- 42.12 MPa and TM-0.243 GPa	–	–	[93]
Rice husk	Chitosan		0.4–1.2 g	Tensile strength- 0.005 - 0.266 Kgf/cm^2 EAB- 4.75 %	–	–	[94]
Starch nanocrystals (SNC), cellulose nanocrystals (CNC)	Mango kernel starch	Solution casting	1.5 wt% CNC and 8.5 wt% SNC	Tensile strength 16.68 MPa EB- 12 %	1.265 ± 0.091 g mm kPa − 1 h − 1 m−	–	[95]
Titanium dioxide (TiO_2)/CNC	Cellulose	Casting	CNC- 7.5 % TiO_2–1 %	–	–	Antioxidant and antimicrobial activity	[96]
Titanium dioxide	Pectin-polyvinylpyrrolidone	Solution casting	0.3 % *w*/w	TS- 13.17 to 30.02 MPa	2.938 g mm /mm^2 day kPa	Antimicrobial and antioxidant activity	[97]
Titanium dioxide/ $CaCl_2$	Flaxseed mucilage and pectin	Solution casting	0.25–1.0 % w/w (TiO_2) and 2–5 % w/ w $CaCl_2$	EB- 13.62 %	–	UV-blocking activity	[98]
Zeolitic imidazolate framework (ZIF)-8/ zeolite	Guar gum	Solution casting	0.2–2.0 % w/w	TS- 1.21- 1.49 MPa	0.31 g/m^2/day	Ethylene scavenging and antioxidant activity	[99]
ZnONPs (1 to 3 wt% of CMC)	Carboxymethyl cellulose	Solution casting	1 to 3 wt%	Tensile strength- 43.5 to 81.9 MPa EB- 8-10 %	2.04 to 1.62 10^{-9} g.m/m^2.Pa.s	Antioxidant and Antimicrobial activity	[100]
Zinc oxide	Chitosan	Casting	0–2 % wt.	TS- 26.82-44.74 MPa Tensile modulus- 1.82- 3.30 GPa	1.604 × 10^{-18} g cm^{-1} s^{-1} Pa^{-1}	Antibacterial activity against *B. subtilis* and *E. coli.*	[101]
microcrystalline cellulose	keratin extracted from chicken feathers	Casting method	15 % keratin, 10 % of glycerol, 30 %, and 2 % cellulose	–	–	Better thermal properties.	[102]
reduced graphene oxide	starch and chitosan	solvent casting methodology	1.5 % polysaccharides, 0.375 % graphene	Tensile strength 38.6–27.7 MPa	Water contact angle 100°	Antioxidant activity, Electrical conductivity, Heat sealing capacity	[85]
talc	PLA/PBSA blend	screw extruder	<4 % talc	Tensile strength 27–32 MPa	–	Better light and gas barrier properties, lower glass transition temperature.	[103]
sugarcane bagasse fiber	Cocoa pod husk cellulose	Casting method	3.5 % cellulose, 0–3.5 % bagasse fiber		10–20 % moisture content of the films, 15–45 % water absorption percentage	–	Mohammad-Azmin, Najah Aliah binti Mohd Hayat, & Mohd Shukri Mat Nor, 2020)

(*continued on next page*)

Table 2 (*continued*)

Reinforced materials	Base materials	Methods	Concentration	Mechanical strength	Moisture barrier	Active function	Reference
Cinnamaldehyde	Wheat Gliadins	Casting method	20 % gliadin, 0–1 % cinnamaldehyde	–	Hampering water absorption and weight loss, leading to more water-resistance.	–	[104]
Bentonite	Yam starch	Casting method	3 % starch, 0.5 %–1.5 % bentonite	Tensile strength 2.5–4.0 MPa	Contact angle 45–80°	Good soil degradation properties and salt impermeability.	[31]
Citric acid	Rice and potato starch	Casting method	10 %–50 %, citric acid	–	10–14 % Moisture	–	[105]
Sodium lignosulphonate (LS), sulfite fish oil (FOS), and Fe3+ ions	Gelatin	Casting method	5 % gelatin, 20 mM Fe3+	Tensile strength 20–35 Mpa	Contact angle 90–100°	–	[106]
sugarcane bagasse cellulose fibers	Acetylated corn starch	screw extruder	12 % sugarcane fibers	Tensile strength 19–118 Mpa	Contact angle 80–85°	Biodegradability	[52]
microcrystalline cellulose (MCC)	Carrageenan		1 %–3.5 % of MCC	tensile strength 20.74 MPa	The moisture content, 16.72 %	–	Nor Amira Othman, Fatmawati-Adam, & Nur Hidayah Mat Yasin, 2021)

barrier properties, and biodegradability are internally relevant. From the viewpoint of food packaging application, these properties must be designed and optimized according to the application conditions, which may involve food processing conditions (sterilization), storage conditions (freezing), or possibly cooking conditions (microwave heating). Therefore, the optimum properties are often achieved by trade-offs.

5.1. Mechanical properties

For food packaging films or materials, mechanical properties, including tensile strength, Young's modulus, and the elongation at failure, are important. Tensile strength is defined as the maximum tension (tensile stress) that a material can withstand without breaking, and the value is expressed as the cross-sectional area of the material divided by the maximum load, with unit N/m^2 or MPa. Young's modulus (also known as tensile modulus) is defined as the ratio of stress (force per unit area) along an axis to strain (ratio of deformation over initial length) along the axis, with the unit of MPa. Elongation at failure (%) describes the maximum value of strain caused by elongation, expressed as the ratio of a material's maximum elongation length at the breaking point relative to the initial length of the material. For food packaging materials, adequate mechanical strength is vital to their applicability. The internal structure of bioplastics is considered the dominant affecting factor. For industrial production, mixing biodegradable polymers or introduction fillers is a common way to improve the mechanical properties of bioplastics. Even if the filler content is relatively low, it is still effective at improving the mechanical properties. Moreover, mixing bioplastics with other polymers like chitosan, Starch/polyvinyl alcohol (PVA), and polylactic acid (PLA) can also increase the tensile strength and Young's modulus. Still, the elongation at failure may vary depending on different composites. It has been reported that the incorporation of nano-SiO_2 increased the tensile strength and Young's modulus of the material but decreased the elongation at failure. However, the presence of an excessively high content of plasticizers was reported to decrease the mechanical strength of bioplastics [111]. The reason is ascribed to the interaction between components in bioplastics. It is considered that the strategies to increase the friction between components or increase the orientation of the biopolymers in bioplastics are effective at increasing mechanical strength.

Moreover, the internal structure of bioplastics should also be affected by their production process, such as molding temperature, drying time, and so on. Thus, these factors are also considered to affect the mechanical properties of bioplastics. Currently, the casting method is mostly used to prepare bioplastics at the laboratory scale. Despite its simplicity, this method is difficult to adopt at the industrial scale, requiring a long drying time. Instead, industrial production of bioplastics mostly employs extrusion or injection molding techniques [112]. In this process, undesired water evaporation would cause material instability and air bubbles, which would greatly affect the materials' durability, break strain, and strength. Previously, Delgado et al. used press cake, a byproduct of the rapeseed oil industry, to produce the bioplastics by adopting injection molding techniques at 80, 100, and 120 °C, respectively. It was shown that the viscoelastic properties of the materials increased by 50 % as the molding temperature increased from 80 to 120 °C. The reason was ascribed to the thermally promoted protein cross-linking at higher temperatures [113].

Furthermore, after production, exposure of bioplastics to external environmental factors, such as light, temperature, and storage humidity, would also affect the mechanical properties of bioplastics. A typical example is starches-based bioplastics. During storage, retrogradation of starches can occur where the amylose and amylopectin chains tend to reorganize themselves to a more crystalline structure, which can expel water from the material matrices and result in higher fragility [111].

5.2. Water resistance

Bioplastics should possess high water resistance as food packaging materials to prevent their wetting from contacting with liquid or high water-content food matrices [167]. The evaluation parameters of water resistance include water absorption, contact angle, moisture content, and water vapor permeation [107]. They were incorporating hydrophobic fillers (such as sugar palm fiber, cellulose nanofibers, lignin-containing CNFs, and microcrystalline cellulose (MCC) or polymers is a common way to improve the water-resistance of bioplastics. For instance, when sugar palm fibers were used in starch-based bioplastics, the moisture content and water vapor permeation slightly decreased, attributed to good intermolecular adhesion between sugar palm fibers and starch [52,111]. After CNF or MCC were added, the starch-based bioplastics also possessed a lower moisture content and water vapor permeation, as well as a higher water contact angle, because the hydrophobic lignin surfaces created better adhesion of MCC to starch, leading to lower porosity and higher density of the materials. However,

excessive loadings of CNF and MCC may cause aggregation and higher porosity of the composites, which may reduce their water resistance [114].

Additionally, incorporating inorganic fillers (e.g., montmorillonite) can confer better water resistance to bioplastics [115]. However, one should be careful when using other substances (e.g., plasticizers). As an example, plasticizers are expected to increase the intermolecular spaces in bioplastics and thus weaken intermolecular bonding forces. Therefore, excessive addition of plasticizers may reduce water resistance [109].

Increasing crosslink density in bioplastics is also feasible to improve the water-resistance [116]. Previously, Balaguer et al. (2011). used cinnamaldehyde to crosslink wheat gliadins to prepare bioplastic films [104]. Due to the formation of new covalent bonds by cinnamaldehyde, the films' water absorption and weight loss were significantly reduced, thus imparting the films' higher water resistance. Immersing the films in water for five months did not disintegrate the materials. Similarly, blocking hydrophilic groups like hydroxyl groups in bioplastics can also increase water resistance through cross-linking. For instance, introducing urea-formaldehyde in a PVA-starch blend and chitosan-starch blends as a crosslinker has significantly enhanced the water contact angle and decreased water vapor permeation and water absorption [111]. In addition, starch-based bioplastics cross-linked by citric acid also exhibited a reduced water absorption percentage and water contact angle [105].

5.3. Barrier properties of gases and vapors

Enhancing barrier qualities in bioplastics by reinforcing them with various fillers is a complex project with great potential for environmentally friendly packaging and other uses. When natural fibers like hemp or jute are added, they create a network inside the bioplastic matrix that prevents liquids and gasses from passing through. The surface area of nanofillers, such as nanoparticles or nanoclays, is greatly increased, strengthening the barrier against outside influences. The use of graphene as a reinforcement provides remarkable resistance against the passage of gases and liquids by imparting impermeability. Key problems for all these solutions include achieving an ideal dispersion and making sure that the filler and bioplastic matrix have excellent interfacial adhesion [14,117]. Conventional packaging materials, like metal, glass, and synthetic plastics, exhibit high barrier properties to gases and vapors, which is favorable for retaining good food quality and prolonging food shelf life [118]. However, for bioplastics, water vapor and gas permeability are not as low as in conventional packaging materials [119]. Thus, it is needed to improve the barrier properties of gases and vapors of bioplastics for food packaging applications. The gas and vapor barrier properties depend dominantly on the structures of food packaging systems. Incorporating nanofillers (e.g., clay and metallic nanoparticles) has been suggested to improve the barrier properties of bioplastics due to their large surface area ratio [120]. For example, the incorporation of clay has been reported to increase the tortuous paths in PLA nanocomposites and thus decrease the vapor permeability of the nanocomposites to half of its initial value [121], which was attributed to the remarkable increase in the lengths of the tortuous paths in the nanocomposites.

Moreover, the effect of relative orientation and dispersion (intercalated, exfoliated, or some intermediate) of the fillers on the barrier properties of bioplastics is also important because the orientation of fillers affects the spatial porosity of the materials at nanoscale scales [122]. Previously, Gusev and Lustic (2001) proposed a rational strategy for regulating the barrier properties of nanocomposites [123]. They considered that the high aspect ratio of nanofillers with atomic thickness (nanoplatelets) sometimes can cause transformations of the fillers in the polymer matrix at the molecular level, which changes the local gas permeability coefficients of the nanocomposites.

Moreover, the interaction between the nanofillers and the polymer matrix is also important in improving the vapor barrier properties. It has been reported that the interaction between PLA and silicate layers led to a disordered intercalated system of PLA/saponite due to the formation of phosphonium oxide by the reaction between the hydroxyl edge group of PLA and alkyl phosphonium cation. Therefore, the vapor barrier property of PLA/saponite was unexpectedly higher than that of the other systems [124]. Taken together, if the gas or vapor permeability is in the range of 20–50 $\times 10^{10}$ g/m^2•s•Pa, the films are considered to have acceptable barrier properties.

5.4. Antioxidant and antimicrobial properties

Antioxidants have been added to food packaging material to inhibit oxidation. Antioxidants in the packaging materials can migrate directly into liquids or indirectly into solid foods and prevent oxidation of food products. Any antioxidant packaging is based on the release of antioxidants.

From the packaging material to the contained food, even though some antioxidants on the package surface may act to scavenge the free radicals in the package headspace. Non-volatile antioxidants, such as tocopherol and ascorbic acid, are considered suited for liquids or semi-solid foods because they can directly contact the package surface and absorb antioxidants. Volatile antioxidants such as natural essential oils

Sesamol is thought to work by inhibiting gas-phase oxidation reactions with headspace free radicals and subsequent autooxidation in the food matrix with indirect migration. Usually, the antioxidant effectiveness of the active packaging film varies depending on the packaged food, packaging material, and storage conditions. Integrating natural antioxidant agents into biodegradable packaging materials is attracting wide attention; one example is PLA films with tocopherol incorporated. Moreover, many natural components (e.g., essential oils) contain ingredients with both antioxidant and antimicrobial activities[166]. Therefore, packaging materials or coatings with natural essential oils may play the dual role of antioxidant and antimicrobial preservation. Such packaging materials meet a need for active packaging that provides antioxidant and antimicrobial functions. Thus, these materials favor foods susceptible to microbial spoilage and oxidative deterioration.

Although many components possess both antioxidant and antibacterial functions, selecting the right antimicrobial agents is essential when an antimicrobial property is a priority. The major functions of food packaging materials are to inhibit the growth of the pathogen and kill the pathogen after growth; thus, antimicrobial properties are important for increasing the food shelf life and retaining food safety [125]. Generally, biopolymer packaging materials reinforced by active substances can exhibit remarkable antibacterial activity against *Escherichia coli, Listeria monocytogenes, Salmonella* sp., and *Staphylococcus aureus* [126,127]. These active compounds usually include essential oils, peptides, proteins, and plant polyphenols [128]. These active components are believed to exert an antimicrobial effect by different mechanisms. The most common mechanism is to generate reactive oxygen species (ROS) by interacting with food-borne pathogens that can lead to cell wall puncturing, cell membrane inactivation, DNA damage, electron transport chain inhibition, mitochondrial perturbation, hindrance of protein synthesis (Fig. 7). The generated ROS during metabolism of aerobic cells include the hydroxyl radical (OH), hydroxyl anion (OH^-), hydrogen peroxide (H_2O_2), and superoxide anion ($O2^-$), whose concentration is considered to be linearly associated with mortality of bacteria. In addition, ROS can also destroy the constituents of the cell membrane and thus cause destruction to the membrane integrity.

Some plant extracts, such as phenolic components, can inactivate cell membranes by disturbing the membrane permeability and hindering the enzymatic activity of microbes [127]. For chitin/chitosan, the positive charge of the polymer chains can interact with the cell wall's negative charge, leading to the wall's destabilization and subsequent cell death. Peptides can bind and insert themselves in the membrane of bacteria, which leads to the formation of pores in the membrane and thus exhibits

significant antimicrobial activity. Lysozyme can efficiently inhibit the growth of gram-positive bacteria by splitting the β-1,4 linkage between *N*-acetylmuramic acid and *N*-acetylglucosamine of the peptidoglycan in their cell walls. Still, this effect is insignificant for gram-negative bacteria because of a thick outer membrane outside the peptidoglycan layer. Moreover, nanocomposites or nanoparticles, including silver nanoparticles, TiO_2, ZnO, and MnO, are also efficient in antibacterial activity owing to their high surface-to-volume ratio and high surface reactivity [128–130]. The major mechanism of metal nanoparticles for killing bacteria is the release of dissociated ions, which interact with the negative charge of the bacterial cell wall and lead to cell death [126,131].

Furthermore, it is believed that the diffusion of antimicrobial compounds through packaging materials is also important to affect the efficacy of the antibacterial function, which depends on physical and chemical factors and sometimes on the interactions with the packaging polymer. Thus, there is no complete preservative protection for all foods, and the efficacy of preservatives in packaging must be studied case by case. Metal nanoparticles, such as AgNPs, ZnO, and TiO2, effectively inhibit microorganisms like *Alicyclobacillus acidoterrestris, Staphylococcus aureus, mesophiles, Escherichia coli, E. coli*, etc. For sodium lactate, potassium sorbate, sodium diacetate, *L. monocytogenes* can be well inhibited. Natural extracts like essential oils cinnamaldehyde, eugenol, and organic acid are shown to inhibit effectively *Enterobacteriaceae, lactic acid bacteria, L. sakei, serratia* spp., *and E. coli.* A more detailed exhibition can be found elsewhere [129].

5.5. Biodegradability

Plastic can be considered biodegradable if a significant change in the chemical structure, i.e., degradation, occurs in the exposed material, resulting in the formation of carbon dioxide, water, inorganic compounds, and biomass (new microbial cell constituents) but no visible or toxic residues under composting conditions. Plastics degradation can occur by five mechanisms: photodegradation by natural (day) light, oxidation by chemical additives, thermal degradation by heat, mechanical degradation by mechanical effects,

And biodegradation by microorganisms (Fig. 6). Good biodegradability is extremely important for eliminating environmental concerns toward bioplastics [118]. In addition to their properties, extrinsic conditions, including temperature, availability of oxygen and water, and the presence of microorganisms, can also affect the biodegradability of bioplastics as documented in the Table 1. For example, the degradation of PLA in the natural environment is significantly better than polyethylene (PE), a widely used raw material to manufacture most plastic food and beverage packaging materials [132]. In a wild environment, slow hydrolysis of PLA may occur when exposed to an aerobic environment with suitable moisture and temperatures around 30 °C. Recently, Ghasemlou et al. studied the environmental fate of non-isocyanate polyhydroxyurethanes (PHUs) under soil biodegradation. They found that soil microbiomes effectively and quickly digested PHUs and starch bioplastics-PHU hybrids. All starch bioplastics-PHU hybrids were rapidly biodegraded with mass losses of up to ~88 % when buried in soil for 120 days, suggesting good biodegradability [133]. Recently, Meng et al. (2023) evaluated the degradation process of three commercial bioplastics made from starch, PLA, and polybutylene adipate terephthalate (PBAT) in different soil environments. The degradation of all bioplastics was found to follow two distinct stages. In the initial period (days 0–30), the bioplastics experienced a major weight loss (35.8–41.9 %), accompanied by a steep increase in the soil's dissoluble organic carbon and the change of distinct bacterial communities. The weight loss of the bioplastics occurring in this stage was attributed to starch degradation. In the second step (days 30–360), the weight loss was maintained at a relatively slow rate, and the bacterial communities in the soil started to recover gradually, which is attributed to the degradation of the remaining PLA and PBAT [132].

Despite these synthetic polymers being considered biodegradable, in most cases, their degradation is a slow process. Natural polymer-based bioplastics, such as starch, zein, cellulose, chitosan, and lignin, have a biodegradation kinetic quicker than synthetic polymers [134]. A major reason is that natural biopolymers can be easily used as substrates for the biochemical processes of bacteria and thus be degraded by most

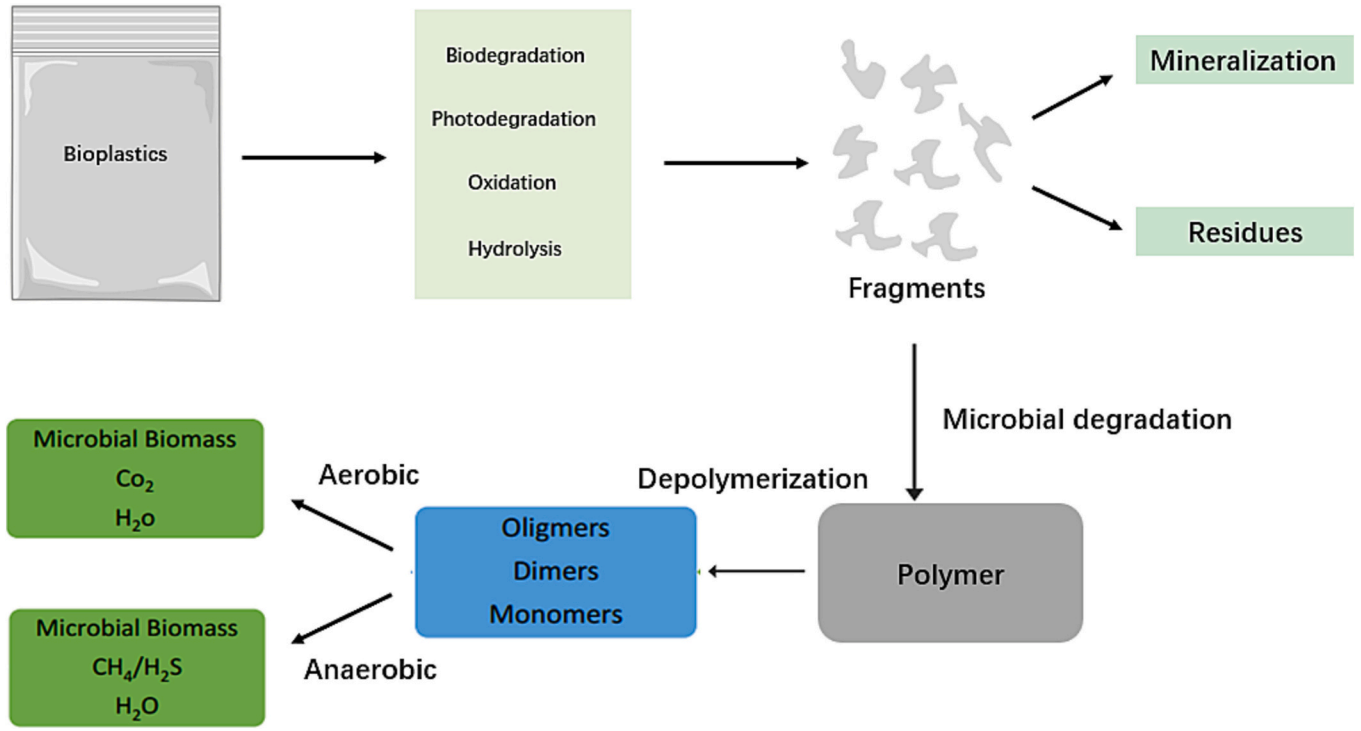

Fig. 6. Degradation path of bioplastics.

microbial enzymes [135]. Overall, the degradability of bioplastics depends on intrinsic factors and extrinsic factors. The former includes chemical properties (polymeric composition, additive chemicals) and physical properties (molecular weight, hydrophobicity, morphology, crystallinity) [163]. The extrinsic factors refer to environmental factors, including extracellular enzymes, temperature, salinity, pH, oxygen, pressure, sunlight and UV exposure, moisture, fragmentation, adsorption or other substances, etc.

5.6. Other considerations

Bioplastics' mechanical, barrier, and antioxidant properties and biodegradability are considered internally relevant. When extrinsic antioxidant or antimicrobial components or fillers are incorporated, the mechanical and barrier properties of the plastics will be altered. For example, some antimicrobials can act as plasticizers; thus, their incorporation will improve the tensile properties of the materials. Moreover, adding essential oils will change the film's barrier properties by altering the systems' interfacial compatibility and hydrophobicity. If the interfacial compatibilities are low, the film will show low adherence and bigger voids in the material, increasing gas transmission rates.

6. Nano-approach to improve the reinforced bioplastics

Currently, numerous nanofillers, such as clay, montmorillonite, cellulose nanowhiskers, starch nanocrystals, layered titanate, and carbon nanotubes, are used to improve the properties of bioplastics [121,122,136]. These nanofillers can be three-dimensional spherical and polyhedral particles such as colloidal silica, two-dimensional nanofibers like nanotubes, or one-dimensional discs like clay platelets. They should possess a geometric shape with at least one dimension in the nanometer range [137]. Mixing nanofillers with biodegradable polymers can confer the composite materials better mechanical and oxidation stability, decreased solvent uptake, self-extinguishing behavior, and even tunable biodegradability [121]. The most used nanofillers are layered inorganic solids such as clay minerals, graphite, metal phosphates, bentonite, and graphene oxide [31,85]. Incorporating clay minerals, montmorillonite (MMT), hectorite, saponite, and laponite, has been proved to be very effective at enhancing the mechanical and barrier properties of the bioplastics while maintaining their biodegradability [115,138]. Previously, Alves et al. designed starch-chitosan bioplastic films reinforced by reduced graphene oxide and found that blending starch chitosan with reduced graphene oxide conferred the films a hydrophobic surface (water contact angle>100°), lower water solubility, and improved antioxidant activity. The optimum film composition consisted of 75 % starch, 25 % chitosan, and 25 % graphene oxide, which can achieve the maximum electrical conductivity (6.51 × 10–3 S/m) while maintaining the heat-sealing properties of starch [85].

Moreover, cellulose nanofibers or whiskers are also good choices for mixing with bio-based polymers due to their abundance in plants and natural origin [114,139,140]. Earlier, Mohan and Panneerselvam reviewed the mechanical and barrier properties of PLA-based films as reinforced by various fillers. They found that the incorporation of nanofillers, such as bacterial cellulose nanocrystals, cellulose nanowhiskers, and cellulose nanofibers, contributed to the increased tensile strength, thermal stability, and crystallinity, thereby improving the barrier properties of the PLA-based composite films [119]. Compared with layered silicates, cellulose nanofibers or nanowhiskers are more advantageous because of their better renewability, higher specific strength and aspect ratio (100−1000), lower cost, and material density. However, cellulose nanofibers and nanowhiskers usually exhibit low thermal stability and low production yield, which remain the major drawbacks when processing cellulose nanofibers or whiskers-reinforced bioplastics. Major cellulose types for reinforcing polymer composites are MCC [102], CNC, and fibrillated cellulose, depending on the extraction methods. Chemical pretreatments, ultrasonication, and ball milling can further modify these cellulose types to produce smaller sizes and improved properties.

In the food industry, many organic, inorganic, and biologically active substances have been employed for food packaging applications as antimicrobial agents to prevent the growth of pathogenic microorganisms [127,138]. For inorganic fillers, metal nanoparticles (NPs) such as Ag and ZnO have been reported to possess considerable antibacterial activities. The antibacterial mechanisms are considered to result from their huge surface-to-volume that offers more direct interaction with bacterial surfaces and the possible electrostatic interactions between some cationic NPs and negatively charged external layers of the membrane of some bacteria [127]. The attachment is believed to damage the bacterial cell wall, releasing the bacteria's DNA and causing cell death [125]. Incorporating other nanoparticles, such as silver nanoparticles, in packaging materials can also confer good antibacterial properties on the materials, further delaying or inhibiting the spoilage of food products [125]. Antibacterial activities of these metal nanoparticles have been effective against gram-positive and gram-negative microorganisms. Notably, different types of nanoparticles may possess different sensitivities against bacteria. For example, ZnO nanoparticles have been reported to suppress the growth of *Staphylococcus aureus*. In contrast, silver nanoparticles exhibited antibacterial activity against *Escherichia coli*, *Aeromonas hydrophila*, and *Klebsiella pneumoniae* in a concentration-dependent manner [141].

Furthermore, nanocomposites also promise to improve bioplastic's barrier and mechanical properties, as discussed earlier. Overall, a significant advantage of using nanofillers is that a very low content of addition (< 5 %) can exhibit remarkable enhancement in tensile modulus, heat resistance, and barrier properties [115]. In addition, nanofillers offer other benefits to bioplastics, such as low density, good transparency, better surface properties, and recyclability [120]. Also, it has been reported that incorporating nanofillers (e.g., graphene layers) in a polymer matrix can create a tortuous path that increases the difficulty of gases to pass and thus functions as a barrier (Fig. 7).

7. Environmental risk of the reinforced bioplastics

The major types of commercial biodegradable plastic include PLA, PHA, biodegradable aliphatic and aliphatic/aromatic copolyesters, blends of starches and derivatized starches, blends of celluloses and cellulose derivatives, PVOH and blends [142–144]. In most cases, these materials are used as blends rather than single materials, for example, PLA/PHB blends, PLA/copolyester blends, PLA/starch blends, starch/copolyester blends, as well as starch/PVOH blends [110,145]. Moreover, many other biobased materials are also being explored, including protein-based compositions [146] and bacterial-, algal-, and fungal-based materials [147]. Compared with traditional synthetic polymers, these materials can be degraded much more easily by biological path (biodegradation), leading to bioplastics' stepwise fragmentation and further degradation into CO_2 H_2O [144]. In this process, methane, a greenhouse gas twenty times more potent than carbon dioxide, may be produced [148]. In addition, the degradation of bioplastics require suitable extrinsic conditions, and inadequate degradation will cause a potential pollution risk to the environment. Compared with conventional plastics, however, it is basically accepted that the environmental burden from bioplastics was much less. At present, determining the environmental fate of bioplastics seems to be difficult because the sources and entry routes of bioplastics into the environment are so different, which requires different timescales to determine their degradation pathways. A quick degradation of wood-derived bioplastic was reported by Chen et al., who constructed this material by treating natural balsa and natural rubber latex via delignification, in situ infiltration, densification, and vulcanization. In addition to the excellent mechanical strength and oxygen barrier property, the bioplastic was also found to display good biodegradability, with an almost complete degradation after five weeks in soil [149].

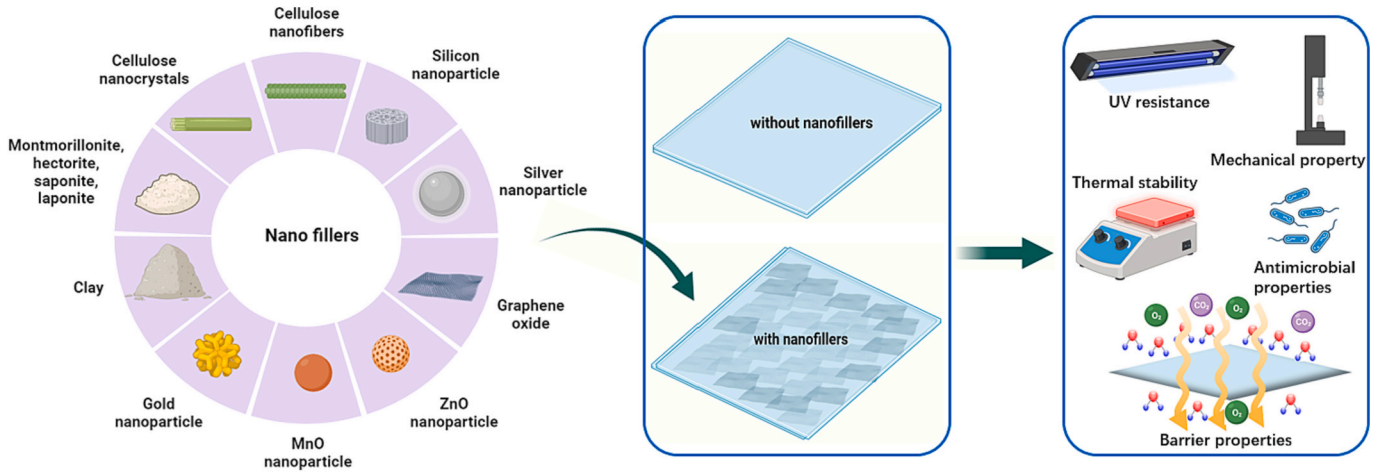

Fig. 7. The methods of using nanofillers to improve the properties of reinforced bioplastics.

Like conventional plastics, bioplastics can also undergo fragmentation and degradation via various physicochemical or biological processes in the environment, during which much smaller particles or residues, widely known as microplastics or nanoplastics, can be produced. Primary pieces of bioplastics are more likely to degrade to microplastics under mechanical fragmentation than photooxidation. In contrast, PBAT/PLA and PE bags are more likely to degrade to microplastics through photooxidation than mechanical forces. Although bioplastics are considered to completely degrade in the environment, a complete degradation may require a sufficiently long time and specific media and conditions (e.g., water, soil, and compost) [150]. Therefore, the production of microplastics or nanoplastics is also ubiquitous for bioplastic degradation. Tong et al. (2022) assessed the formation of secondary microplastics and nanoplastics of various biodegradable plastics (PBAT, PBS, and PLA) and conventional plastics (PE, PS, and PVC) under the effects of aging factors (UV radiation, mechanical forces). It was found that secondary microplastics were increasingly produced over the prolonged degradation time, with the particle size falling within 1–50 μm for all plastics. Polymer thickness was believed to dominate in determining degradation patterns and rates [151]. It was considered that most problems associated with the environmental rate of conventional plastics are also largely associated with biodegradable plastics.

Moreover, due to superior biodegradability, bioplastics may break into microplastics or nanoplastics faster than conventional plastics, causing an additional threat to the soil environment or the ecosystem [152–154]. These produced micro- or nanoplastics are believed to possess stronger toxicity due to their smaller particle size and higher specific surface area. It is noted that, during biodegradation, bioplastics can also release potentially toxic substances (e.g., plasticizers and dyes) into the soil, which may cause the enrichment of other pollutants. Therefore, biodegradable plastics are not completely harmless, as declared by producers. Unfortunately, the present data for the harms posed by bioplastic degradation are still lacking, which entails more experimental data and in-depth explanations to demonstrate the ecological effects of bioplastic degradation.

Additionally, the present ecotoxicity studies of biodegradable microplastics are usually focused on certain particles, and chemicals released during the natural weathering of bioplastics should also be considered [152]. Akoueson et al. previously tested the chemical and toxicological profiles of two types of food packaging materials (PP and PLA). It was shown that both PP and PLA polymers released organic plastic additives into seawater under the tested conditions (dark conditions, 20 °C). Still, the chemical content and the leachate composition differed depending on the polymer types and suppliers. Therefore, the authors considered it impossible to attribute a chemical pattern to a specific polymer type [155]. Despite that, most of the nanofillers applied have been approved; however, a detailed investigation of their possible migration from packaging to food products and an estimation of the potential toxicity is still urgent for safe delivery [125].

To alleviate the current environmental pollution, it is suggested that bioplastics should be degraded by composting after recycling [153]. In addition, to minimize the influence of bioplastic degradation on the environment, a new composting strategy, such as anaerobic digestion, is recently proposed as the end-of-life of biodegradable plastics. However, different materials exhibit varying sensitivities to anaerobic digestion. For example, thermoplastic starch and PHA can reach a high degree of degradation in a relatively short period under mesophilic and thermophilic conditions. In contrast, other polymers, such as PLA, have a very low biodegradation rate under mesophilic conditions. Moreover, some polymers that are considered biodegradable under industrial composting conditions (e.g., PBS and PBAT) are not well biodegraded under anaerobic digestion, even if the degradation time is very long [134] (Fig. 8).

8. Future trend of reinforced bioplastics and their application for food packaging

Over the past decade, the development of bioplastics has been greatly prompted by the rising oil prices and the increasing public's awareness of environmental protection. However, the commercialization of most bioplastics requires stable product quality and successive large-scale production with low cost, which remains the major issue to be overcome. To this end, bio-based feedstocks are widely used for the production of bioplastics [107], including 1) natural polysaccharides, such as starches, pectin, chitosan, etc. [156]; 2) cellulosic, lignocellulosic, and cellulose derivatives, such as wood, stover, straw, plant biomass; 3) fats and oils, like animal fats (fatty acid glycerides), vegetable oils (e.g., soy, castor, palm, waxes and triglycerides, and algal oils); 4) some proteins, such as keratin, casein, whey, gelatin, collagen, soy, zein, gluten [156]; 5) other biobased packaging materials, include polycarbonates, aliphatic polycarbonates (PCs), with the latter being under development based on carbon dioxide feedstock. Especially each raw material has unique intrinsic properties, and thus, attention should be paid when considering the target application. Recently, ongoing efforts have endeavored to identify new applications for these new raw materials, such as creating edible and active films, coating or encapsulating materials, etc.

Regarding the application of bioplastics, some large companies have developed commercial materials for food packaging. There are two types of PlantBottle® in the market, one made from bio-based High-Density Polyethylene (HDPE) and the other from partially biobased

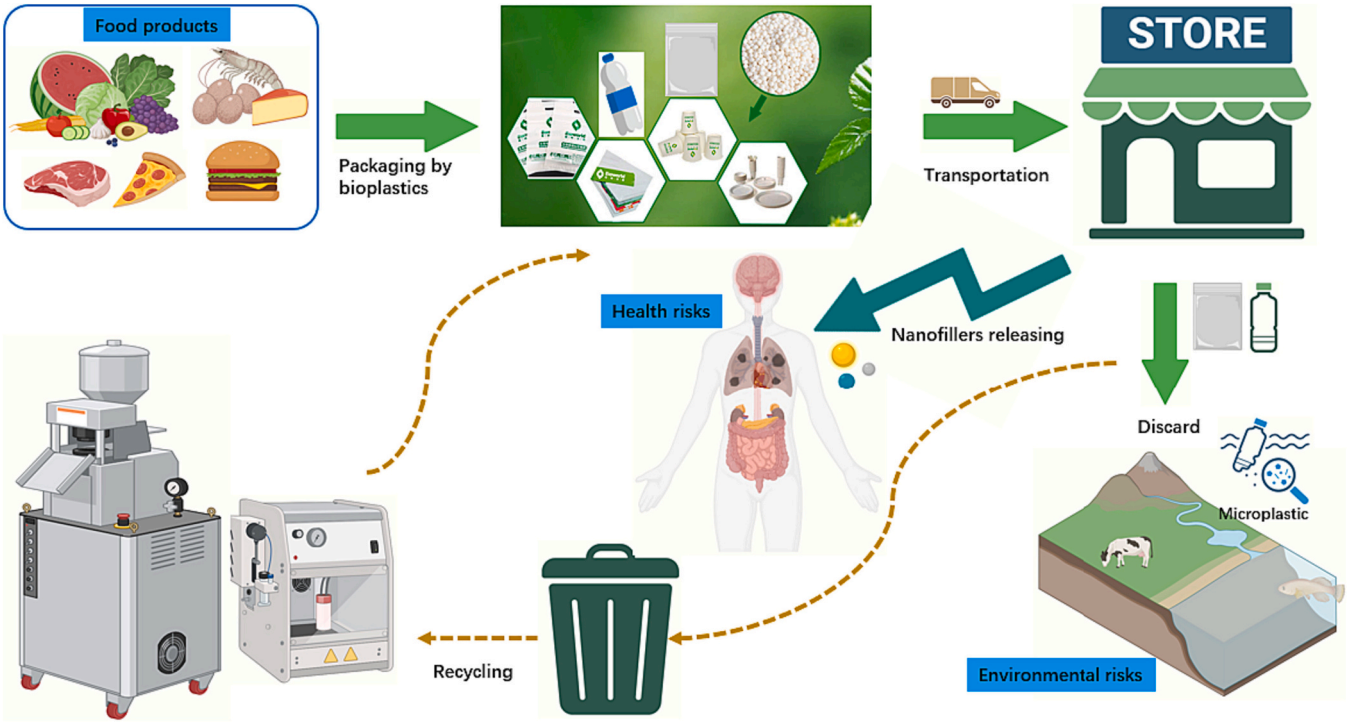

Fig. 8. The potential risks of bioplastics in food chains.

Polyethylene terephthalate (PET) [118]. The PET PlantBottle was first introduced in Denmark in 2009, then in Western Canada and the US, followed by Japan. Now, the use of PET has been spread to over 20 countries. Volvic in Europe has also begun to use biobased PET as water bottles. In 2011, Coca-Cola introduced a 100 % biobased HDPE bottle plant for packaging Odwalla juice products. The attempt to develop other commercial bioplastics-based packaging material is still in progress. Recently, Aversa et al. evaluated the suitability of bioplastic-based bottles for wine packaging, which is developed based on PLA/PBSA and PLA/PBS blends. By adopting the extrusion technique, the polymer blends can form high-quality bottles, and the production cost was approximately 3.30 Eur/kg at raw material prices [103].

For bioplastic packaging materials, it's important to address moisture-induced deterioration and improve printability, sealability, dye uptake, resistance to glazing, and so on, which involves the surface properties of these materials. Since most bioplastics are prepared from natural polysaccharides and proteins, they are by nature more hydrophilic and have low water barrier ability, which is a major limitation to their application as

Packaging materials. To address this problem, lipids or other hydrophobic substances such as resins, waxes, fatty acids, or even some insoluble proteins are recommended to be added to retard moisture transfer. Moreover, the development of heat-resistant proteins or modified sealing techniques for liquid or high-water content foods is also needed for protein-based bioplastics. Food packaging polymers' surface tension (viewed as surface energy) should also be improved to improve printability. For example, good adhesion (where cohesion is defined as the internal strength in any substrate due to various cohesive interactions among similar molecules) in the seal areas is highly desirable at polymer-polymer or polymer-metal interfaces, particularly those composed of laminated materials. Food may become contaminated with foodborne organisms or extraneous materials without adequate adhesion. In recent years, several technologies have been developed to improve the surface energy of food packaging polymers, including flame, corona, and plasma treatments, the latter being considered the most effective. However, the effect of plasma treatment on the surface of biopolymer films needs to be further tested. A greater interest in understanding the surface properties of biopolymer films is expected within the next few years. This may be the key to resolving the fundamental issue of excessive hydrophilicity, allowing full-scale commercial utilization of biodegradable films as food packaging materials.

The risk assessment of reinforced bioplastics as food packaging applications is also urgent, which is currently limited by the shortage of detection and quantification methods for microplastics, especially for biodegradable microplastics. A solution lies in developing new biodegradable polymers with excellent performances, improving bioplastic recycling processes, and tightening regulations on plastic waste disposal [152]. To this end, improving the public's attitudes is also very important [138], which is unfortunately inadequate. Dilkes-Hoffman et al. collected 2518 online surveys from Australia and summarized the public's knowledge and perceptions regarding bioplastics in a recently published review article. It was shown that the Australian public's knowledge of bioplastics is low, but perception, particularly of biodegradable plastics, is positive. Fifty-eight percent of respondents were unsure whether biodegradable plastics can have negative environmental impacts. Sixty-eight percent of people held positive attitude toward using biodegradable plastic items. Also, 62 % of people would like to dispose of bioplastic items in the recycling bin [157]. Fortunately, to promote the use of bioplastics, many countries, including the European Union, United States, China, South Korea, and Japan, have launched some directives and strategies to ban the use of single-use plastics and provide a wide range of programs to support increases in the research, development, and customer awareness of bioplastics [148].

Paper and paperboard packaging materials represent another trend with distinct environmental sustainability advantages. On the one hand, paper and paperboard packaging materials are renewable, highly recyclable, and biodegradable, which can be sourced from sustainably managed forests. On the other hand, sustainable forests can also promote biodiversity and provide numerous benefits to the ecosystem [109]. Furthermore, direction transformation from natural plant wastes into bioplastics may also be a promising approach. Previously, Bayer et al. prepared bioplastics with a wide range of mechanical properties from industrially processed edible vegetable and cereal wastes (including parsley wastes, spinach stems, rice hulls, and cocoa pod

husks) by simply aging them in trifluoroacetic acid (TFA) solutions regardless of their bio-origin. The mechanical properties depended on the plant species, ranging from brittle and rigid to soft and stretchable [158].

Finally, it is also considered promising to develop bioplastics from very abundant biomass on Earth, such as marine algae [159,160]. Algae and microalgae can degrade plastic materials through toxins or enzymes synthesized by themselves, provided that the plastic polymers are used as carbon sources. It has been reported that algae-derived bioplastics exhibited identical properties and some characteristics comparable with petroleum-based conventional plastics but with remarkably better biodegradability in nature [161]. Using algae or microalgae as raw materials to produce bioplastics also has other advantages: including 1) algae can be cultivated on non-arable lands and have short harvesting time, without any effect on the food production for human consumption; 2) algae are tolerant to harsh environmental conditions and can remediate wastewater and utilize carbon dioxide as a nutrient source; 3) during the manufacturing of algae-based plastic, the encapsulation of nonbiodegradable polymer (e.g., polyolefin) in the thermoplastic algal blends can capture and store carbon dioxide in biomass form, blocking the emitting of carbon dioxide back into the atmosphere and thus alleviating the greenhouse effect; 4) bioplastics produced from microalgae are inexpensive due to the huge availability of the biomass [162] (Fig. 9).

9. Conclusions

Developing bioplastics-based materials for food packaging applications is expected to grow rapidly in the next decade, owing to the urgent need to reduce the environmental impact caused by the extensive use of conventional synthetic plastics. However, to realize this target, bioplastic's mechanical, water-resistance, and thermal properties should be improved to compete with the properties of conventional plastic packaging materials. To this end, various nanofillers, including clay, montmorillonite, layered titanate, carbon nanotubes, metal nanoparticles, cellulose nanowhiskers, and nanocrystals, have been increasingly used to be incorporated into bioplastic matrices. It is believed that incorporating nanofillers can improve bioplastic's mechanical, barrier, water resistance, and antimicrobial properties by actively interacting with the bioplastic matrix, increasing the crosslink density, and creating a tortuous path that functions as a gas barrier. Additionally, due to the huge surface-to-volume, most nanofillers can either directly interact with bacterial surfaces or strongly attach to the membrane of bacteria via electrostatic interactions, which damages the bacterial cell wall and causes cell death. A remarkable advantage of using nanofillers is that a low addition content (< 5 %) can exhibit a significant enhancement in the properties of bioplastics. Besides, a careful trade-off is required to obtain the optimum properties of bioplastics, including the rational selection of biomass, filler types, concentration, and suitable processing conditions.

Although bioplastics are considered compostable and degradable in natural ecosystems, their degradation needs suitable external conditions, including temperature, oxygen water availability, and microorganisms. Therefore, the degradation rate of bioplastics can vary drastically under different conditions. Moreover, due to excellent biodegradability, bioplastics can break down into microplastics or nanoplastics faster than conventional plastics, which can cause a threat to the ecosystem. On the other hand, the safety of bioplastics should also be considered for food packaging applications, which is especially important for nanofillers-reinforced bioplastics where the release of nanocomponents is possible during food transportation, storage, and consumption. In this regard, the relevant data and risk assessment is still inadequate.

Many countries, including Europe, the US, Japan, Italy, Mexico, and South Korea, have developed commercial bioplastics for food packaging. Also, to further promote the development of bioplastics, these countries have launched programs to limit the use of single-use conventional plastics. However, commercializing most bioplastics requires stable product quality and successive large-scale production with low cost, which remains a major issue to be addressed. To reduce the production cost and negative environmental impact, two strategies seem promising: 1) abundant biomass such as marine algae or microalgae can be considered as the major feedstock to produce bioplastics industrially; 2) active governmental policies should promote bioplastic recycling and

Fig. 9. A global map with the companies and countries already developing the reinforced bioplastics commercially.

composting. Moreover, developing new functionalities of bioplastics-based packaging materials is also promising.

CRediT authorship contribution statement

Shahida Anusha Siddiqui: Writing – review & editing, Writing – original draft, Visualization, Validation, Resources, Project administration, Methodology, Investigation, Formal analysis, Data curation, Supervision, Funding acquisition. **Xi Yang:** Writing – review & editing, Writing – original draft. **Ram Kumar Deshmukh:** Writing – original draft, Data curation. **Kirtiraj K. Gaikwad:** Formal analysis. **Nur Alim Bahmid:** Writing – review & editing, Conceptualization. **Roberto Castro-Muñoz:** Validation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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