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REVIEW

Biobased composites from agro-industrial wastes and by-products

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Abstract

The greater awareness of non-renewable natural resources preservation needs has led to the development of more ecological high-performance polymeric materials with new functionalities. In this regard, biobased composites are considered interesting options, especially those obtained from agro-industrial wastes and by-products. These are low-cost raw materials derived from renewable sources, which are mostly biodegradable and would otherwise typically be discarded. In this review, recent and innovative academic studies on composites obtained from biopolymers, natural fillers and active agents, as well as green-synthesized nanoparticles are presented. An in-depth discussion of biobased composites structures, properties, manufacture, and life-cycle assessment (LCA) is provided along with a wide up-to-date overview of the most recent works in the field with appropriate references. Potential uses of biobased composites from agri-food residues such as active and intelligent food packaging, agricultural inputs, tissue engineering, among others are described, considering that the specific characteristics of these materials should match the proposed application.

Keywords Sustainability · Biomass · Composite materials · By-products · Agri-food waste · Biodegradable

1 Introduction

Climate change due to greenhouse gases emissions (GGEs) is only one of the several problems derived from an unsustainable linear economy: such as hiking raw materials prices, increasing products demand, resources depletion, irreversible environmental damage, and waste accumulation. Thus, a new regenerative economic view based on a balance between economy, environment, and society aims at a circular production/consumption system seeking to maximize resources use and avoid or, at least, minimize environmental impact [1]. A circular approach implies the substitution of fossil fuels as an energy source and petroleum-based products, such as plastics, for sustainable energy systems (i.e., solar, eolic, biofuels, etc.) and renewable feedstock (mainly biobased products); the development of easily recyclable or biodegradable products; optimization of waste management systems; and the design of products from residues.

Most petroleum-based plastics are highly resistant to degradation in nature, being able to persist for hundreds of years in the environment [2]. Even though they are potentially recyclable, only about 20% of all the plastic produced globally is properly recycled or reused [3]. Even with efficient waste management and recollection systems, the recovered polymers are often shipped to countries with low environmental regulations and control [4]. These, along with the continuous increase in total plastic production and consumption have led to severe plastic pollution, especially in the marine environment. Plastic littering in the oceans represent a hazard to marine fauna, directly damaging their habitat, constituting dangerous traps, and tampering with their food chain. Microplastic, plastic broken into small pieces by erosion, is ingested by marine mammals, reptiles, birds, and fish which can be mortal and affects seafood safety, hence human health [5]. Besides, plastic accumulated in coastal regions degrade their natural attraction having a further impact on tourism and local economies based on this activity. Therefore, biobased and biodegradable plastics have gained interest as potential substitutes for conventional polymers with a growing market and a global production accounting for

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about one percent of the over 368 tons of plastic that are nowadays annually produced [6]. Given their biodegradability, these biobased materials offer new end-of-life routes such as organic recycling through aerobic or anaerobic degradation, agricultural mulching, solubilization, or biodegradation in the environment, resulting in fewer wastes accumulation and soil contamination [1].

Given this context, a transition towards sustainable biobased productions is sought. Generally, biomass is recognized as a sustainable alternative to fossil fuels due to its abundant availability, carbon neutrality, and low sulfur content. However, if not based on a sustainable approach biofuels production entails extensive land use, soil acidification, and eutrophication among other environmental impacts related to intensive agro-industry such as competition amid food and energy crop growing and limited net GGE reductions [7]. In this regard, biodegradable and biobased plastics face similar challenges as alternatives to conventional plastics. Therefore, more sustainable biofuels and biobased products, such as food and pharmaceutical ingredients; fine, specialty, and commodity chemicals; polymers; and fibers have been developed from non-food crops, agricultural wastes, and forest residues to be coproduced in biorefineries [8]. Particularly, the use of agri-food waste has enormous potential in sustainable bioeconomy or green economies considering the food manufacturing industry [9]. In traditional agricultural settings, production residues are usually burnt or landfilled, which results in a noticeable CO_2 generation [10]. Other typical agri-food waste management implies its use as animal feed or fertilizer supply, composting, and anaerobic digestion, yet recovery and valorization capacity of these residues remain quite limited [11]. Considering that five percent of global GGEs originate from organic waste decay and that agri-food residues contain macromolecules, such as carbohydrates, proteins, and lipids, as well as active compounds and pigments, strategies to obtain higher value-added materials from agri-food wastes and by-products are both urgent and conceivable [9, 12]. Numerous researches have been done and reviewed in this regard, from agro-industrial waste and by-products as feedstock for biofuels and bioplastic synthesis [8, 13–15], direct biopolymers, and active compounds extraction from agri-food residues [9, 16, 17], are reinforcing materials for composite materials [10, 18–24]. Therefore, the present review focuses on revising and comparing the existing studies on fully biobased composite and nanocomposite materials, considering different processing technologies, and analyzing the potential uses of the developed materials. Current limitations on fully biobased products design and market as well as LCAs availability are discussed, aiming to give a relatively broad outlook on the state of the art and future perspectives on the field of sustainable biocomposite materials.

2 Potential agro-industrial wastes and by-products for biobased composites

2.1 Bioplastics

Plastic material is defined as bioplastic if it is either biobased, biodegradable, or features both properties. The term biobased implies that its components are mainly derived from biomass, while biodegradation is a chemical process by which a material is converted into water, carbon dioxide, and compost by the action of naturally available microorganisms under normal environmental conditions. For a polymer to be categorized as biodegradable bioplastic should as well meet the following criteria [25]:

- *Chemical characteristics:* at least 50% of its final composition should be necessarily organic matter.
- *Biodegradation:* it should degrade by a minimum of 90% of its weight/volume within 6 months under-stimulated composting conditions.
- *Ecotoxicity:* non-degradable residuals after biodegradation for 6 months should not be a potential threat to plant's growth.
- *Disintegration:* components' microscopic fragments should be undetectable (<2 mm) at least within 2 months under controlled composting conditions.

Bioplastics (both biodegradable and nonbiodegradable) can then be classified into 4 main groups: directly extracted from biomass, synthesized from a biobased monomer, synthesized from petrochemicals, and produced by microorganisms (Figure 1). They comprise a whole family of materials with different properties and applications and are nowadays ecological alternatives for many conventional plastics. The latest market data compiled by European Bioplastics in cooperation with the nova-Institute reported that the global bioplastics production capacities are set to increase from 2.11 million tons to approximately 2.87 million tons between 2020 and 2025, of which biodegradable and biobased bioplastics accounts for over 50% of the market [6]. In agreement with Siakeng et al. [26]: the biodegradable character creates a positive impact in society and also attracts researchers and industries. Therefore, this work focuses on those bioplastics that are essentially biodegradable and extracted directly from biomass or obtained by microbial fermentation of biomass.

2.1.1 Biopolymers from biomass

Polysaccharides are the most abundant macromolecules in nature, being many of them suitable raw materials for bioplastics. They are nontoxic and widely available since

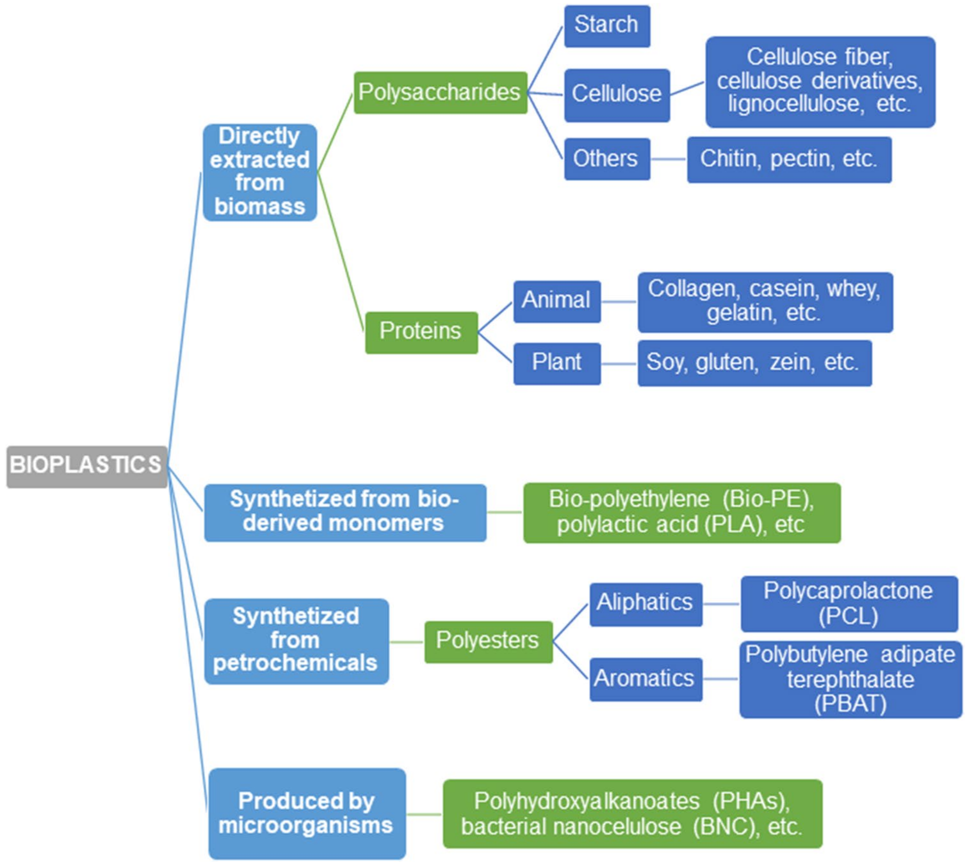

Fig. 1 Bioplastics classification according to production process and origin with some examples

they can be obtained from many different sources such as plants, microorganisms, algae, and animals. Due to their physicochemical properties, many of them are susceptible to physical and chemical modifications leading to enhanced properties with various applications as biomaterials [27].

In particular, cellulose is a widely available polysaccharide derived from renewable resources [28]. It is generally synthesized by plants, but it is also produced by some bacteria. Plant-derived cellulose is usually found mixed with hemicellulose, lignin, pectin, and other substances, while bacterial cellulose is quite pure and has much higher water content and tensile strength owing to its longer polymer chains. The most used cellulose derivatives are methylcellulose (MC), hydroxypropyl methylcellulose (HPMC), and carboxymethyl cellulose (CMC) [29]. Above all, CMC has been reported to have excellent film-forming properties by thermal gelatinization with water-soluble polymers [30]. Moreover, nanocellulose or nanocrystalline cellulose is a versatile material with great mechanical and chemical resistance that is becoming increasingly valued for several applications, from packaging to electronics, yet limited by availability and cost [31–33].

Most plants produce starch as energy storage; thus, this polysaccharide can be extracted from rice, cassava, corn, wheat, and potatoes, among others. Starch is a carbohydrate that contains a great amount of glucose units combined through glycosidic links, however functional and structural dissimilarities are present among different botanical species [34]. Starch differ in type and content of two constitutive D-glucose macromolecules: amylose, linear, and helicoidal polymer; and amylopectin, with a branched structure [35]. The amylose content may vary from 20 to 25% in cereal starches, 15 to 30% in roots and tuber starches, and up to 40% in fruit starches, while the amylopectin content varies from 75 to 80% by weight. Even though starch has proven thermoplastic properties, its efficiency as a raw material for bioplastics depends upon its specific structure and composition [36]. Thermoplastic starch is obtained by the starch granule disruption in the presence of plasticizers. Extrusion processing employing low moisture content, high temperature, and pressure melts the starch granules into a single continuous phase component that can later be mixed with other components to form a film [25].

Pectin is an important natural polymer with diverse industrial applications. It is present in various fruits and vegetables such as berries, apples, and oranges and is extracted mainly from the processing residues of these agricultural products [37, 38]. Pectin has attracted great interest because of its distinctive characteristics: it has the ability to freeze in the presence of acids and sugars, presents high viscosity and aqueous-absorbent gel properties, and it is easily soluble in water but is insoluble in ethanol [39]. Therefore, it shows great potential for the development of biobased membranes, films, and edible coatings in the food packaging field [40, 41]. Pectin films are effective in the protection of low moisture food [42], while pectin coating are used to preserve fresh fruits and vegetables [38]. Moreover, cross-linking of pectin films with polyvalent cations, such as calcium, enhance their mechanical properties [43].

Likewise, being the second-most abundant biopolymer in the world (after cellulose), chitin is a promising raw material for bioplastics. It is the main structural component of the fungi cell wall and can be also found in shells of ocean-dwelling crustaceans such as crab and shrimp. Chitosan is obtained from the chemical modification of chitin, extracted from fungi and shells, after numerous chemical treatments. Soluble in weak acid solutions, chitosan can be dried to a solid plastic film and strengthened by soaking in alkaline solutions with promising uses for packaging and medical applications [44–49]. In addition, near 150,000 tons of chitin-rich waste is annually produced by the seafood industry worldwide [50]. These residues have a high environmental impact, little commercial uses, and high waste-disposal cost, thus extracting a high-added-value bioplastic from this waste creates an excellent cost-effective and more sustainable opportunity for the seafood industry.

Regarding marine origin biopolymers, carrageenan and alginate are worth mentioning as anionic polysaccharides found in the outer cell wall of red and brown algae. Alginates have been used for encapsulation of chemical and biological compounds with a wide range of applications in agriculture, food technologies, pharmaceutical cosmetics, chemical engineering, environmental engineering, paper and textile industry, and many others due to their nontoxicity, biocompatibility, and the ability to cross-link with cations [51–55].

Furthermore, many proteins from vegetable and animal sources can be used as the raw material for developing bioplastics materials. Wheat gluten, for instance, a by-product from the bio-ethanol industry, is relatively inexpensive, abundant, and basically used as animal feed but is thermoplastic in nature and presents interesting film formation capacity, gas barrier, mechanical and biodegradation properties that have risen the interest in its use in the packaging industry [56]. Wheat gluten-based biocomposites and nanocomposites with improved barrier and mechanical properties particularly suitable for packaging have been reported [57, 58].

2.1.2 Polymers from microorganisms

Microorganisms are a source of biopolymers using agricultural wastes as growth media. Although currently more expensive, bacteria have the potential of yielding bioplastics having properties comparable to those of conventional polymers that can be further modified by changing the growing medium and conditions of the bacteria. Bioplastics from the microbial production process have been optimized with a wide range of end products showing diverse properties [25]. Numerous of these microbiologically synthesize biopolymers have gained acceptability in food and other industrial applications among which polyhydroxyalkanoates (PHAs) have attracted particular interest. PHAs are polyesters of hydroxy acids naturally synthesized by bacteria as carbon reserves. These biopolymers are accumulated as cytoplasmic inclusions in certain bacteria during unbalanced growth conditions, usually characterized by an excess in feed supply and the lack of one or more essential nutrients [59]. PHAs are synthesized by different groups of bacteria from cheap renewable resources, yet in order to effectively exploit the commercial production of these biopolymers, it is important to select a bacterial strain having the highest PHAs yields growing on inexpensive carbon sources with efficient fermentation and requiring simple recovery processes.

More than 150 different PHA monomers have been identified, which renders them the largest group of natural polyesters [60]. For instance, poly(3-hydroxybutyrate) (PHB), poly(3-hydroxyvalerate) (PHV), and their copolymer poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV) are typical examples of short-chain-length PHAs. Particularly, PHB is the most popular and promising PHA as an alternative biomaterial, since it has similar properties to conventional polyesters such as PE and polypropylene (PP) [61]. Its application includes packaging materials, bags, containers, sutures, targeted tissue repair, and regeneration devices, cardiovascular stents, polymer-based depots for controlled drug release or implants, and disposable items like single-use cups and diapers [62]. Despite its biobased nature, biodegradability, and versatility, the high production cost of PHB is the main obstacle for its commercialization, being this at least three times higher than the conventional plastics such as PP and PE and similar to biopolymers like PLA [14]. Such high costs are mainly attributed to expensive substrates and processing [63]. Therefore, the use of cheaper feedstocks is a key factor towards reducing PHB production costs. Food wastes [64], wastes from beer breweries [65], cheese whey [66], olive mill wastewater [67], and hydrolyzed corn starch [68] are some resources that have been investigated for sustainable PHB production.

In the last decades, bacterial nanocellulose (BNC) has gained increasing interest because of its remarkable physical and chemical properties, including green technology processing, low production costs, elevated mechanical properties, hydrophilicity, and excellent biocompatibility and biodegradability [69]. Certain gram-negative non-pathogenic bacteria genera were reported to produce nanocellulose extracellularly [70]. It should be noted that despite sharing a common backbone there are marked differences between plant and bacterial cellulose. Plant fibers are composed of lignin, hemicelluloses, pectin, and only 40–70% of cellulose [71]. In contrast, bacterial cellulose is made up of pure cellulose nanofibers, displaying high purity and strength, without requiring subsequent refining treatments. BNC ultrafine structure presents higher crystallinity and polymerization degree, greater liquid absorption capacity, larger specific surface area, and better mechanical properties making it a superior choice to plant-sourced cellulose in many applications, especially in packaging [72] and biotechnological industry [73].

2.1.3 Polymers synthesized for monomers derived from biomass

Among bioplastics biobased nonbiodegradable polymers as bio-PP or bio-PE account for 41.8% of the current global bioplastics production [6]. Meanwhile, along with starch-based bioplastics, PLA is one of the most largely produced biodegradable biobased polymers (18.7% of the total annual bioplastic production in 2020). This versatile compostable biopolymer is synthesized from lactic acid, a naturally occurring organic acid easily produced by chemical synthesis or fermentation. Similar to other bioplastics, one of the main obstacles in PLA commercial use is their cost, thus the use of blends with cheaper biodegradable biobased polymers (i.e., starch) and its biocomposites with low-cost natural fillers has been studied and reported [74, 75]. PLA-based packaging can now be purchased almost everywhere, from food containers, disposable cutlery to suture thread and 3D printing filaments. Consequently, research on enhanced PLA biocomposite for such applications is still under study [74, 76–81]. Due to its biocompatibility, biodegradability, nontoxicity, and high strength, it has been studied for innovative biomedical and pharmaceutical applications, as drug delivery systems, wound dressing, and scaffolds for cellular growth [82, 83].

2.2 Reinforcing materials

In spite of their renewable and biodegradable character, the mechanical resistance, permeability and thermal stability of biopolymers tend to be relatively low for some applications [84–86]. Consequently, the best approach to improve their properties and commercial importance is to incorporate reinforcing agents [86–90]. The resulting materials known as environment-friendly polymer composites, biopolymer composites, or biocomposites, have a wide range of next-generation applications in medicine, electronics, construction, packaging, and automotive sectors [91]. Composites can be defined as materials that are formed by two or more constituents which have separate phases and compositions conforming to micro- or nano-structures. The composite properties are strongly dependent on the matrix (continuous phase) and filler (discontinuous phase) interfacial adhesion, as well as the reinforcement composition, size, shape, and content [90]. The smaller the filler particle size, the greater the efficiency for the formation of composites [89]. Biopolymer composites are synthesized using numerous methods, being in-situ reaction, solution casting method, and melt mixing technique the most employed. Diverse types of fillers can be used that, as suggested by Kumar et al. [92], can be divided following several criteria as shown in Figure 2.

In this work, the first classification according to fillers' origin and composition was considered. Distinctively, organic fillers derive from living organisms and are usually carbon-based compounds, while inorganic fillers are salt, metal, and elemental compound obtained from inert things. A further description of nanosized fillers (<100 nm) obtained by sustainable technology was included due to their key importance in high-performance eco-friendly biocomposite applications.

2.2.1 Organic fillers

Most of the organic fillers used in green composites (both biobased and non-biobased) are derived from renewable sources and are generally cellulose-rich materials. Natural cellulosic fibers, such as hemp, sisal, jute, kenaf, flax, and bamboo, among other plant tissue fibers, have been extensively used as filler of polymer composite materials as substitutes for glass-fiber mainly due to their lower density and cost, their renewable character and because they are less abrasive to processing equipment [85, 93]. Green composite materials have been extensively studied and applied in the transport and construction industries, from windows frames and insulation panels to railroad sleepers and automotive parts, and various other low-cost and mild-mechanical-demanding applications such as gardening items, agriculture mulch, and packaging [22, 94–99]. Therefore, interest in renewable and biodegradable fillers has grown as new sustainable materials are sought, specifically since these organic fillers can be obtained from agro-industrial or wood byproducts and residues [23, 88, 98, 100–102]. In this regard, fillers with diverse mechanical and surface properties, chemical composition, size, and form have been studied. Besides, conventional fibers from plant leaves and stems that are cultivated for their fibers, other such as wheat husk [103], rice straw [104], sugarcane bagasse [105], malt bagasse [106], banana leaves, and peel fibers [17, 77, 107] are by-products of agri-food production that, among others, have been studied as biocomposite fillers [18, 108–110]. Furthermore, starch from roots and tubers bagasse and peel have also been reported [19, 98, 101, 111–113], as well

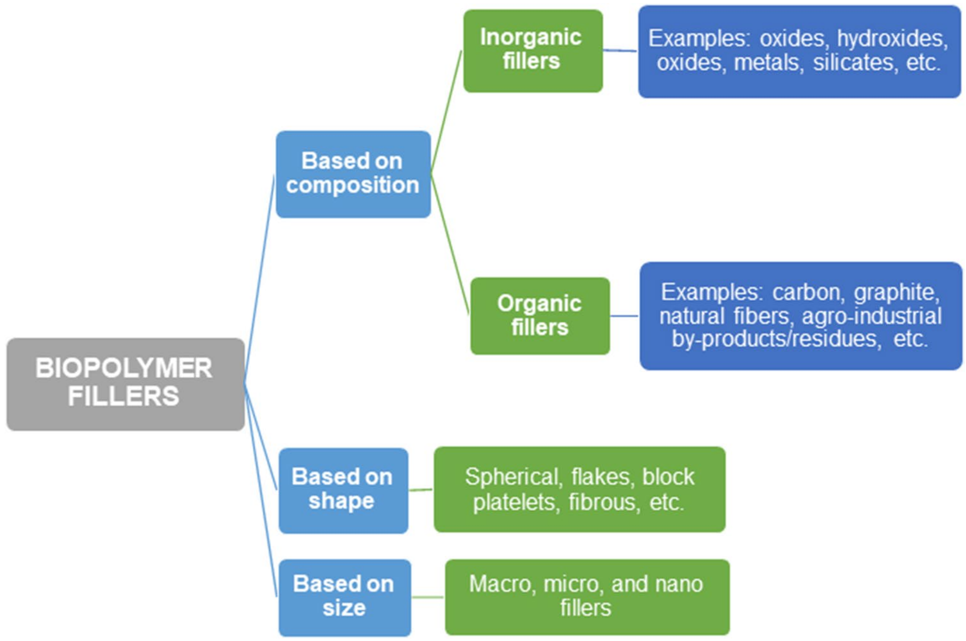

Fig. 2 Biopolymer fillers sorting according to three different criteria with some examples

as algae, microalgae, and their byproducts [83, 114–117]. Recently, grasses such as Sabai grass (*Eulaliopsis binata*), an undervalued abundant grass in Asia [118], Cogon grass (*Imperata cylindrica*), one of the ten most aggressive weeds in the world [119], and Napier (*Pennisetum purpureum schum*) grass [120] have been considered as composite fillers with promising results. Novel green biocomposites have been lately developed from coffee silverskin and starch-rich potato washing slurries [121] or tea leaves from tea brewing wastes [122]. Oil industry by-products, as for instance sesame, rapeseed, peanut, and sunflower oil cakes have also been investigated as fillers for biocomposite films and foams [21, 123–127].

Increasing fiber contents tend to promote stiffer materials with higher impact strength yet diminishing their flexibility. Nevertheless, properties are dependent on filler source and content, surface treatments, particle size distribution, and processing conditions [88, 100, 101, 128–130]. In general, plant tissue fibers are composed of cellulose, hemicellulose, and lignin and their reinforcing efficiency depend on the cellulose nature and crystallinity and its alignment in the cell walls: high cellulose content and low microfibril angle (MFA, defined as the angle microfibrils make with respect to the fiber axis) are desirable [131, 132]. Yet, natural fibers have low thermal stability (approximately up to 200 °C) which limits the processing conditions and the recyclability of biocomposites [85]. Nonetheless, as reported by Ramamoorthy et al. [133] and Chaitanya et al. [74], fiber thermal stability can be enhanced in composite materials as the polymer matrix protects the fiber from degrading. Chemical, physical, and biological pretreatments of the fibers have been firstly proposed to improve filler-polymer interaction in hydrophobic polymer matrices, though such treatments also result in cleaner surfaces, higher moisture content, and thermal stability [134–140]. Notwithstanding, various factors should be assessed in choosing fillers treatments as for enhancing biocomposites properties without compromising their sustainable character: energy and resources consumption and processing cost (especially in the case of complex techniques that result impractical for industrial applications), effluents characteristics and volume generated, reagents toxicity in long term exposures (for work safety) and life cycle environmental impact of the process. Still, one of the major problems in the use of plant-based fillers is their properties fluctuation with botanical source, cultivation region (with different climate conditions and soil composition), and harvest season, which can be somewhat tackled by mixing batches of sources or types of fibers [131].

Other organic fillers are extracted from wastes. Figure 3 illustrates the main sources of organic fillers with some examples for each. Cellulose micro and nanofibers have been isolated by a series of alkali, acid, and mechanical treatments that breakdown the original plant tissue from various sources: soybean hulls [141], sugarcane and cassava bagasse [142–145], and corncob and pinewood [146], among others [120, 147, 148]. Similarly, lignin can be extracted for lignocellulose byproducts and waste [149]. Keratin and chitin, which are extracted from animal feed waste such as chicken wings or shrimp shells are also employed as composites fillers [22, 150–153]. Furthermore, biochar or activated carbon can be obtained from various biomass sources through pyrolytic processes [154, 155]. Both present high adsorption capacity, resulting in special interest in water and air decontamination.

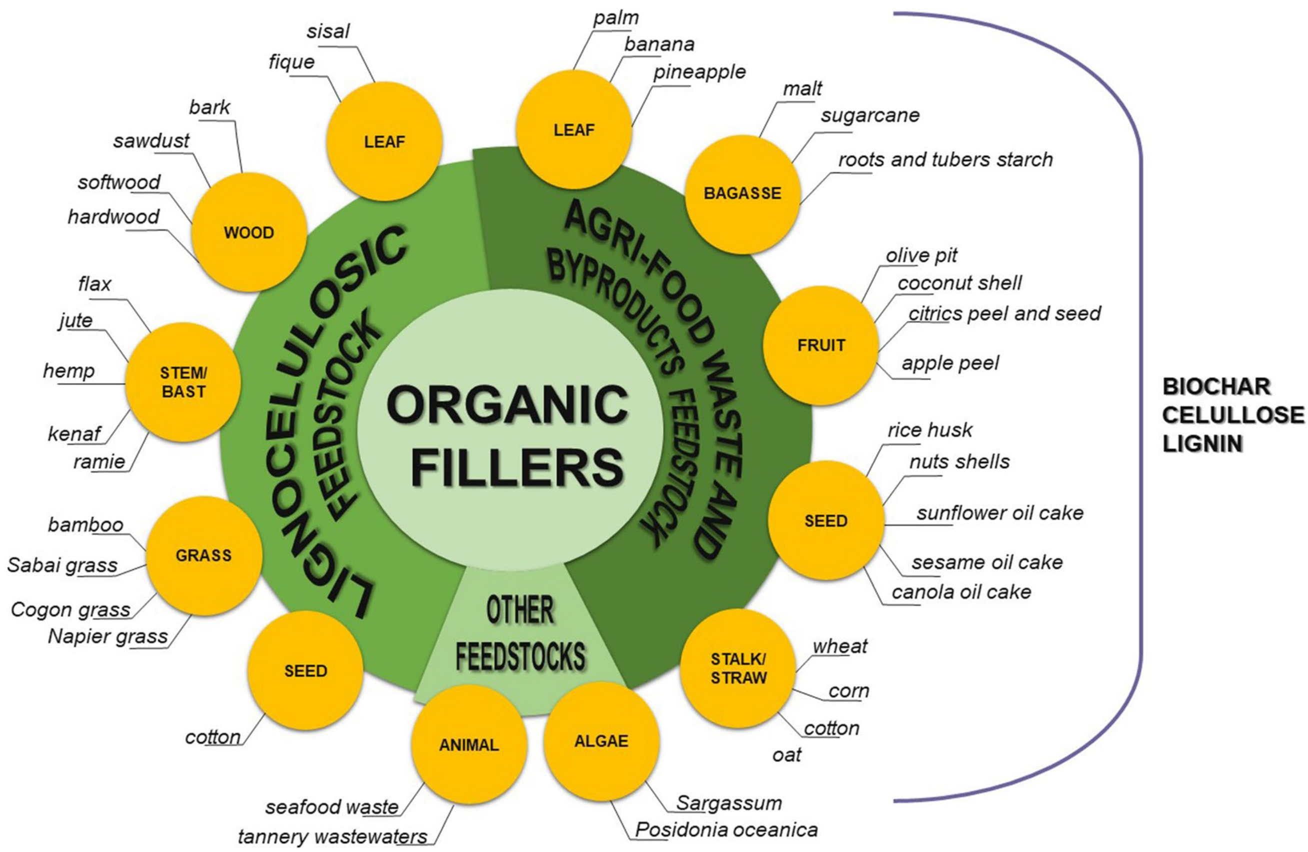

Fig. 3 Organic fillers main sources and some examples for each

2.2.2 Inorganic fillers

Mineral or metallic fillers are considered inorganic fillers: clay and nanoclay, silver nanoparticles (AgNPs), and calcium carbonate ($CaCO_3$) are among the most common inorganic reinforcement agents of biocomposites.

Due to its great natural abundance, clay is the most frequently used inorganic filler in the composite field. These are phyllosilicate minerals usually obtained from the chemical weathering of other silicate minerals on earth [156, 157]. Clay has a good intercalation property and can swell with the absorption of water [158]. Clay can be classified into a variety of groups including kaolinite, montmorillonite, illite, chlorite, and fibrous silicate [90]. Bentonites consist mainly of montmorillonite and can be used as adsorbents, ion exchangers, wine clarification agents, and catalysts. Besides due to their eco-friendly character, availability, and reusability have also been studied as reinforcing agents of polymeric matrices [159, 160]. For their applications, the pillaring process is commonly used to modify the structural, thermal, and surface properties of bentonites. In this regard, Ninago et al. [161] proposed an environmentally friendly method to obtain Al-pillared clays by using microwave irradiation.

Among minerals, calcium carbonate ($CaCO_3$) is a widely inorganic material used as a viscosity modifier in many industries, which is normally obtained from carbonatite-lava, stalactites, stalagmites, skeletons, or shells of some animals. It is an inorganic filler with various potential applications owing to its low-cost, abundance, and safe character [162]. Meanwhile, talc also qualifies as a good reinforcement agent because of its platy nature, presenting micron-sized length and width and nanometric thicknesses, as well as a high aspect ratio (particle diameter/thickness 20:1)[163].

Moreover, granite sand (GS) is an industrial waste derived from the granite polishing industry that can cause health problems and air pollution due to its powder form. Therefore, it is highly desirable to find uses in an effective manner to minimize these damages, reducing as well the need for new dump lands for these wastes [164]. Granite sand is a mixture of different minerals composed of muscovite, orthoclase, quartz, and biotite, among others. Particularly, muscovite is a laminar silicate of the micas-clays family and its structure facilitates the intercalation of organic-inorganic species between mineral slabs, which makes muscovite an excellent filler for polymeric materials [165]. Passaretti et al. [166] employed GS particles as fillers of thermoplastic corn starch films, demonstrating the potentiality of this mineral for this application.

Figure 4 shows a schematic representation of the processing of biocomposite materials with inorganic fillers as well as some examples of SEM micrographs of different inorganic fillers.

2.2.3 Nanoparticles: green synthesis

Nano-sized particles are characterized by their high surface-to-volume ratio, which confers exceptional features on them. They are synthesized through physical, chemical, or biological methods, that are classified into top-down and bottom-up synthesis regarding whether the reagents are inorganic or are generated from the break-down of a macroscopic material by some external agent (Figure 5). Several physical and chemical methods like hydrothermal, sol-gel synthesis, laser ablation, or lithography, among others, require special equipment and skilled labor [167–169]. In addition, some of these techniques involve the use of toxic reagents that pose health and environmental hazards [168, 170]. For instance, silver nanoparticles (AgNPs) can be obtained by the reduction of a silver salt using strong reducers such as sodium borohydride, which is an extremely irritant and corrosive agent with high flammability risk [171]. Nowadays, green chemistry aims at the total or partial elimination of chemical waste and the implementation of nontoxic reagents, environmentally acceptable solvents and renewable materials, obtaining products with high thermal stability, low volatility, and cost-effective production [172–176]. Not only do these eco-friendly techniques reduce the use of hazardous substances, but also employ natural renewable compounds like polysaccharides, proteins, or those derived from vegetable extracts (mainly leaves, roots, and flowers) and microorganisms like bacteria, fungi, and algae, as reducing or capping agents [177–180]. Therefore, the three main concepts of nanoparticles green synthesis are the choice of the solvent (preferably water); the use of an ecological reducing agent, GRAS (substances generally recognized as safe), or natural reagents; and a nontoxic material for nanoparticles stabilization (i.e., biopolymers). Products of natural origin contain in their structure phenolic compounds, reducing sugar and nitrogen compounds that can reduce metal cations to generate nanoparticles and, in certain cases, can also act as stabilizers (Figure 5). In turn, the implementation of these compounds is also advantageous from the economic point

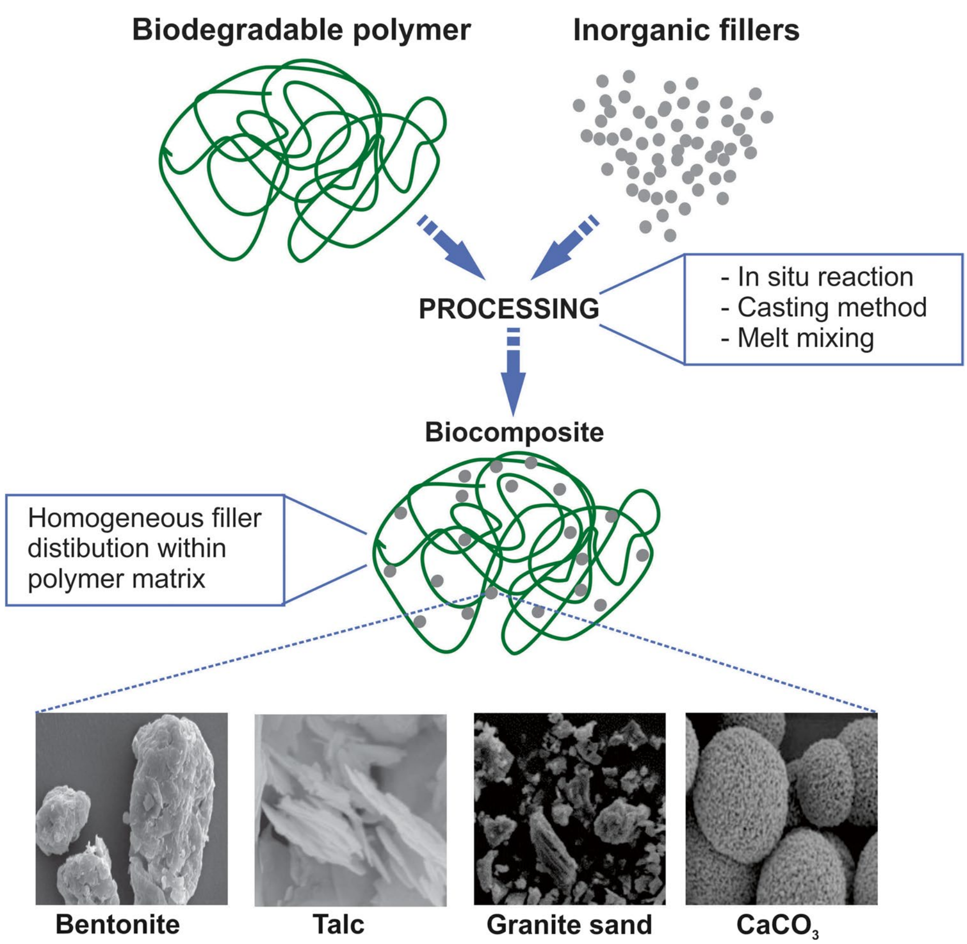

Fig. 4 Schematic representation of the processing of biocomposite materials with inorganic fillers as well as some examples of SEM micrographs of different inorganic fillers

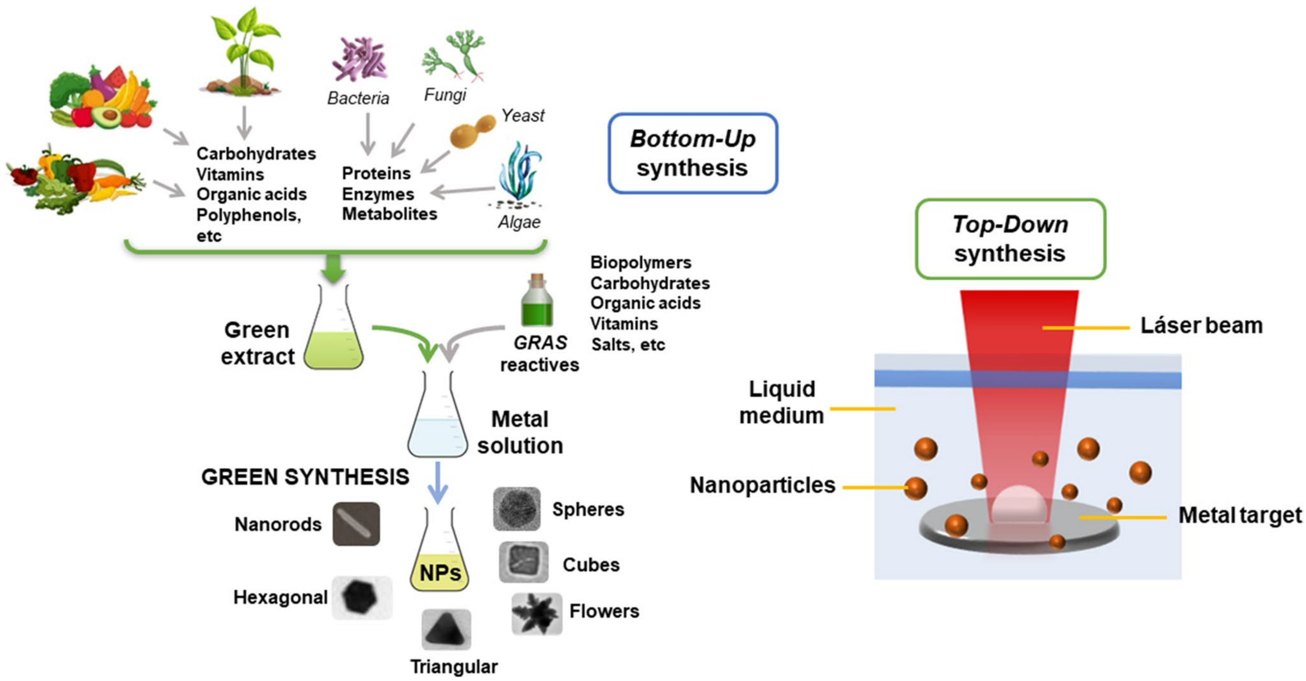

Fig. 5 Nanoparticles top-down and bottom-up synthesis

of view since they do not require high temperatures conditions and reduce energy consumption. Table 1 summarizes common metal and metal oxide nanoparticles obtained by green synthesis techniques.

The electrical conductivity, high stability, and especially the antimicrobial activity of AgNPs have prompted numerous investigations [212, 213]. They can be synthesized by chemical reduction [212, 214, 215], laser ablation [211, 216, 217], electrical and photochemical reduction[218]. Chemical reduction of a silver salt, mostly with organic reagents, is the most widely used and profitable method for large-scale synthesis [219]. Besides, nanoparticles morphology and size can be controlled by chemical synthesis depending on the capping agent and stabilizer. Several authors have reported that spherical and small AgNPs obtained by a completely green chemical process show good antimicrobial properties even when they were used in low concentrations [212, 214, 215, 220]. Ortega et al. [221] have successfully coupled the AgNPs synthesis with cornstarch-based filmogenic suspensions to develop nanocomposite films. Thus, a simple and nontoxic method was proposed to obtain silver nanoparticles where maltose is used a reducing agent, corn starch as a stabilizer, and ultrapure water as a solvent. Proposing the use of corn starch as stabilizers allows the synthesis of AgNPs to be coupled to the filmogenic suspension and thus obtain nanocomposite films in just a few steps, optimizing both processing time and energy, and reagents consumption. Processes coupling requires a prior fine-tuning of the reagent concentrations and reaction times to obtain the AgNPs in the filmogenic suspension [185, 186]. The AgNPs formation is evidenced by the characteristic surface plasmon resonance (SPR) between 420 and 445 nm [222]. Several authors have synthesized AgNPs with lemon juice, which was spherical with diameters around 20 nm as observed by high-resolution transmission electron microscopy (HR-TEM), exhibiting negative charge considering their Z potential measurements [185, 213, 218, 223].

Likewise, ZnO nanoparticles have arisen great interest in sensing applications, since they exhibit high electron mobility, large exciton binding energy, wide bandgap, and high optical transmittance [168]. They can be synthesized with different morphologies, such as spheres, discs, ribbons, flowers, or bars [224]. ZnO nanorods stand out as reinforcement of active packaging materials due to their excellent mechanical performance and their marked antimicrobial activity [225]. They can be prepared by different methods, being the chemical reactions in solution preferred in terms of costs, simplicity, efficiency, and energy consumption [226]. Hydrothermal growth in aqueous solution is a widely used methodology in the literature for obtaining ZnO nanorods. This method uses an aqueous solution of $Zn(NO_3)_2$ containing hexamethylenetetramine (HMTA) which hydrolyzes and produces a basic environment necessary for the formation of $Zn(OH)_2$ and stabilizes Zn^{+2}. In general, seeds of ZnO are incorporated for the hydrothermal growth of ZnO nanorods to improve the morphology and orientation of the bars [227]. The ZnO seeds are synthesized through a simple and low-cost sol-gel process in a nonaqueous solution of zinc

Table 1 Examples of nanoparticles obtained by green synthesis techniques

Nanoparticles	Green synthesis technique	Shape	Size (nm)	Reported applications	References
Ag	Maltose as reducing agent and soluble starch as stabilizer	Spherical with high monodispersity	5	-	[174]
Ag	Microwave-assisted using orange peel extract	Nanospheres	7.36 ± 8.06	-	[181]
Ag	In vivo synthesis of nanomaterials in plants: *Brassica juncea*, *Festuca rubra*, *and Medicago sativa*	Variable	Variable	-	[182]
Ag	Using marine algae *Caulerpa racemosa*	Spherical with few triangular nanoparticles	10 nm mean	-	[183]
Ag	Maltose as reductor and cornstarch as stabilizing agent	Spherical	5–20	Included in nanocomposite films for dairy product shelf-life extension	[184]
Ag	Maltose as reductor and different concentrations of cornstarch as stabilizing agent	Spherical	13.4–19.9	Included in starch films for food packaging area	[185]
Ag	Lemon juice as reducing and stabilizing agent	Spherical	5.5–13.5	Included in starch-nanocomposite films for packaging area	[186]
Au	Using citrus fruits aqueous extract: *Citrus limon*, *Citrus reticulata*, and *Citrus sinensis*	Various shapes	15–80	-	[187]
Au	Using brown algae *Cystoseira baccata*	Spherical	8.4 ± 2.2	Anticancer activity	[188]
Au	Using *Lawsoniainermis* leaf extract	Spherical	~20	Catalytic activity in the degradation of anthropogenic aromatic pollutants and dyes, like bromophenol blue and methyl red	[189]
Au	Using an endophytic strain *Fusarium solani* ATLOY – 8 has been isolated from the plant *Chonemorpha fragrans*	Needle and flower-like structures with spindle shape	40–45	The NPs have shown effective anticancer activities	[190]
CoO	Complexation and surface passivation with starch	Square	15–30	-	[191]
Co_3O_4	Using leaves extract of plant *Calotropis gigantea*	Spherical	50–60	Thermal decomposition of ammonium perchlorate and dye-sensitized solar cells	[192]
CoO	Using *P. granatum* extracts	Spherical	40–80	Photo-catalytic applications	[193]
Co_3O_4	Walnut green skin extract as reducing and stabilizing agent	Spherical	60–80	-	[194]
CoO/Co_3O_4	Rosemary extract	Sheet	~100	-	[195]
Cu/Cu_2O	Synthesized by pulsed laser ablation of copper in water	Spherical and are self-assembled forming a cactus-like	2–7	-	[196]
Cu	Using carboxymethyl cellulose as capping agent	Spherical	5.5 ± 0.5	Inclusion on modified electrode and used as electrochemical nitrite sensor	[197]
CuO	Citric acid mediated synthesis using cinnamon bark extracts	Spherical	50–300	Catalytic degradation of dyes and antimicrobial activity	[198]

Table 1 (continued)

Nanoparticles	Green synthesis technique	Shape	Size (nm)	Reported applications	References
Cu	Microwave-mediated green synthesis using *Citrus sinensis* aqueous fruit extract	Round shape	6.93–20.70	-	[199]
Fe	Using *Eucalyptus Tereticornis* leaves extract	Cubic	40–60	Can be applied as an adsorption – flocculation agent in water purification and contaminated groundwater remediation	[200]
Fe_3O_4	Using *Aloe vera* extract	Cubic Spinel Structure	6–30	-	[201]
Fe_2O_3/Fe_3O_4	By laser ablation method at different values of energy	Spherical and agglomerates	14.9–18.7	-	[202]
MgO	Using bacteria	Cubic	30	Anticancer study	[203]
MgO	Using *Costus pictus D. Don* plant leaf extract as a reducing agent	Hexagonal	~50	Antimicrobial and anticancer activity	[204]
MnO_2	Using fresh lemon juice and peel extract	Agglomerates of fine particles with juice and nanorods with peel extract	< 10 nm and nanorods of 8.8 nm	Possible application as positive electrode for lithium-ion batteries	[205]
TiO_2	Using leaf aqueous extract of *Solanum trilobatum*	Uneven spherical and oval	70	Larvicidal and pediculicidal activities	[206]
TiO_2	Bengal gram beans *(Cicer arietinum L.)*	Spherical	~14	As anode in lithium-ion battery	[207]
TiO_2	Using leaf extract of *Jatropha curcasL*	Spherical	13	Photocatalytic degradation of tannery wastewater	[208]
TiO_2	Using lemon peel extract	Spherical	80–140	-	[209]
ZnO	$Zn(NO_3)_2$ as precursor and *Cayratia pedata* leaves extract as reducing agent	Agglomerated form confirmed by FESEM analysis	52.24	-	[168]
ZnO	Combination of sol–gel with hydrothermal growth, using tetramethylammonium hydroxide as a structure-directing agent	Nanorods	Width 15 nm, and length 236 nm	Incorporated in composite materials	[210]
ZnO	Laser ablation using a starch solution as controller and stabilizer agent	Spherical	6–28	-	[211]

acetate as a precursor. The precursor hydrolyzes by heating and forms acetate and Zn ion that binds to the hydroxyl groups (-OH) present in the solution resulting in ZnO formation [228]. At this point, the concentration of hydroxyl groups in the solution determines the size of the nanoparticles obtained and, in general, is adjusted by incorporating NaOH or KOH [229]. Similarly, the chemical reduction of Zn solution using natural compounds derived from different plant extracts has been reported [168].

Several other nanoparticles with exciting and innovative applications like sensors, biomedical, energy storage, and packaging applications have been studied and synthesized by green technologies as shown in Table 1.

The mechanisms involved in the synthesis of nanoparticles have been extensively reviewed by Polte et al. [230]. Ortega et al. [186] synthesized silver nanoparticles using the active compounds of lemon juice ($AgNP_L$) and proposed a four-step growth mechanism for this process. First, the chemical reduction of the Ag salt occurs by the reducers present in lemon juice (mainly ascorbic acid and citric acid, in addition to other components such as reducing sugars, polyphenols, and flavonoids). Polydisperse particles smaller than 1 nm are then formed by coalescence. The third step, which can last between 5 and 60 min depending on the synthesis temperature, corresponds to a metastable state where the particles reach a mean radius of 1 nm. Finally, the coalescence of the formed particles occurs until colloidal stability is sufficient to prevent aggregation. Ortega et al. [186] reported that the optimized conditions for the synthesis of $AgNP_L$ were 30 min at 90 °C, leading to a 5.5 nm nanoparticle with associated stability ($\zeta = -29.5$ mV) that was maintained for up to 90 days. Yet studies on the stability of nanoparticles during storage are scarce and are strongly recommended for future research in the field [171, 231].

3 Films and foams from biobased composites

3.1 Biocomposite films

Biocomposite films are based on different biopolymer matrices and fillers and can be obtained by diverse processing methods. As an example of the enormous amount of biocomposite films that have been studied to date some of the most relevant developments in the last 10 years from different biopolymers will be mentioned.

Cellulose is the most abundant biopolymer, and it is widely employed for sustainable biocomposite materials because of its renewable character, biodegradability, and other specific properties. It can be converted to different structures with a variety of physical properties, depending on the origin of the cellulose and the method of production [120]. Cellulose microfibers (CMFs) can be obtained by refining dilute cellulose suspensions under high-pressure. Nanofibrillated cellulose (NFC), on the other hand, result from cellulose fibers disintegration using high pressure homogenizers combined with chemical or enzymatic treatments [232]. In addition, if the amorphous parts of the cellulose are removed, leaving single and well-defined crystals in a stable colloidal suspension, microcrystalline cellulose (MCC) can be obtained [233]. Thus, depending on cellulose structure and the desired properties of the final materials, this polysaccharide can be used as a biopolymeric matrix or biocomposite filler. In this respect, Kumar et al. [120] extracted cellulose fibrils (CFs) from Napier (*Pennisetum purpureum schum*) grass and used it as a filler of cellulose matrices. These cellulose-based composites obtained by casting, presented good thermal stability and higher tensile resistance than conventional HDPE and PP, deeming them appropriate for biodegradable packaging, wrapping, and mulching applications. Likewise, Spence et al. [234] worked on microfibrillated cellulose (MFCs) composite films containing kaolin clay and calcium carbonate obtained by casting. Even though the authors demonstrated that the addition of mineral fillers reduced films density and water vapor transmission rate (WVTR) and presented proper mechanical properties for packaging applications, their water barrier properties are low in comparison to petroleum-based plastics. Moreover, Trovatti et al. [235] studied the use of NFC as filler of bionanocomposite films with improved thermal and mechanical properties prepared by casting of water-based suspensions of pullulan: an extracellular homopolysaccharide made up of 1,6-linked maltotriose residues, produced by certain strains of the polymorphic fungus *Aureobasidium pullulans*. Thus, the authors assured that these novel bionanocomposites could be labeled as sustainable materials since they were prepared entirely from renewable resources and through a green approach. In another interesting work reported by Oun and Rhim [236], crystalline cellulose nanofibrils (CNF) were isolated from cotton linter pulp using an acid hydrolysis method and later used as filler of sodium carboxymethyl cellulose (CMC) composite the film's obtained by casting. It was demonstrated that CNF is highly compatible with the CMC and the presence of this filler affected films mechanical and water vapor barrier properties. The CMC/CNF composite films have a high potential to be used as edible coating or packaging films for the shelf-life extension of fresh and minimally processed fruits and vegetables. CNFs have been also used as fillers of starch-based biocomposite films [145, 237, 238]. The addition of CNFs obtained from different biomass sources resulted in increased tensile strength and elastic modulus and led to the reduction of elongation at break, water vapor permeability, and, in some cases, oxygen transmission rate. Furthermore, Farooq et al. [239] utilized a variety of softwood Kraft lignin morphologies to obtain strong and ductile CNF nanocomposite films with potential food packaging, water purification, and biomedical applications. In this work, two techniques were employed to obtain biocomposites: casting and thermocompression. The incorporation of lignin rendered tougher film structure, materials waterproof while exhibiting complementary UV shielding and radical scavenging capability.

Starch-based materials offer a very attractive low-cost base for new biodegradable polymers due to their abundance, annual renewability, and ability to be processed with conventional plastic processing equipment [22, 35, 36]. The improvement of mechanical properties of starch-based materials is an ongoing challenge due to their poor mechanical performance, particularly tensile strength [240]. Among the various alternatives to improve these mechanical properties, blends, and composites have been proposed [22, 241–246]. Correspondingly, Ali et al. [241] developed fully biodegradable starch-based films by casting method based on modified (hydroxypropyl) cornstarch and two kinds of commercially available polysaccharide-based macro-crystals (cellulose and starch crystals). They demonstrated that the mechanical properties were modified by crystals addition, increasing the tensile strength and elastic modulus, and decreasing elongation at break. Besides, the biocomposites showed improved protection against UV radiation. Wang et al. [243] studied collagen composites with three different maize starches: waxy maize starch, normal starch, and high amylose starch, showing higher tensile strength and lower solubility in water than collagen film, and increased thermal stability and crystallinity. Noteworthily, Stasi et al. [246] suggested a novel and cost-effective reutilization of carbon waste ashes as a reinforcing agent of biocomposite films based on thermoplastic starch for agricultural applications. Carbon-based ashes produced by pyrolysis of lignocellulosic wastes were added to glycerol and maize native starch in different quantities, which were melt-processed and molded. The authors reported that ash content decreased both moisture sorption and degradation of starch biocomposites. Moreover, Yin et al. [247] proposed to improve the functional properties of starch-based films incorporating chitin obtained from shrimp shell powder into corn starch matrix. Before blending, maleic anhydride was introduced as a cross-linker and composite films were obtained by casting-evaporation. The obtained starch-based nano-biocomposite films presented superior mechanical properties, higher surface hydrophobicity, and enhanced barrier properties, in addition to antibacterial properties against *Escherichia coli* and *Staphylococcus aureus*.

Soy protein isolate (SPI) is another biobased polymer with good film-forming ability that can be produced by casting, extrusion, or injection molding [248–251]. Since SPI films have low strength and absorb a high amount of moisture which limits their applications, reinforcing filler has been proposed [252–255]. Accordingly, Martelli-Tosi et al. [254] investigated the potential use of soybean straw as reinforcing filler in SPI films. Both raw soybean straw and samples treated with alkali (NaOH 5 and 17.5%) and bleached with hydrogen peroxide (H_2O_2) or sodium hypochlorite (NaOCl) was studied. Films added with treated soybean straw presented higher mechanical resistance, lower elongation at break, and lower solubility in water; while the addition of non-treated soybean straw had no significant effect on SPI film properties. Alternatively, Zhao et al. [255] developed a series of epichlorohydrin-crosslinked hydroxypropyl chitosan/SPI films with different soy protein contents. The authors demonstrated that these materials were tunable in terms of their surface structure and mechanical properties by changing the SPI content. Biocomposites exhibited good cytocompatibility and hemocompatibility, improved wound contraction rates, and showed great promotion of granulation tissue regeneration and collagen deposition, which are excellent results for skin tissue engineering.

Among the biodegradable polymers, PHB is the principal and the most widely used type of the PHA, with high potentiality for replacing fossil-based synthetic packaging [256]. Even though this biopolymer displays thermophysical and mechanical characteristics similar to polystyrene and isotactic polypropylene, it presents a narrow processing window which limits its applicability [257]. Therefore, the development of PHB composites has been proposed as a solution. PHB biocomposites employing a wide range of fillers, such as cellulose nanocrystals [258], graphene [259], agave fiber [260], chitosan, and catechin [110] have been developed over the last few years. An interesting work carried out by Araque et al. [261] focused on the development of PHB and hollow glass microspheres and composite films. These materials were obtained through melt intercalation, an innovative technique with low environmental impact because it does not require solvents use. Besides, Seggiani et al. [116] studied PHAs based biocomposites with fibers from *Posidonia oceanica* (PO) to assess their processability by extrusion, mechanical properties, and potential biodegradability in a natural marine environment. These composites provide an interesting valorization route for PO fibrous wastes largely accumulated on coastal beaches and can be suitable to manufacture items usable in marine environments, such as in natural engineering interventions for restoration or protection of coastal habitats.

The processing methods strongly affect the properties of the biocomposite materials [262], thus a description of the most widely used methods to obtain biopolymeric films are given, mentioning the advantages and disadvantages of each of them:

Solution casting: this is the simplest and most frequently used method at a laboratory scale for biocomposite films. The casting technique consists in spreading a film-forming solution or suspension on small plexiglass or plates, in which the film thickness is controlled by the mass of suspension poured onto the plate [263]. The drying process of these films usually takes place at room temperature or in an oven with air circulation [45, 120, 264]. Polymers that are soluble in water are mostly treated with this technique and, as stressed by Bondeson and Oksman [265], the differences in hydrophobicity or the hydrophilicity of the filler and that of the matrix require the use of a suitable solvent. Besides, the solvent concentration can be decided according to the required characteristics and viscosity of the solution. The biopolymers composites obtained through this technique usually show good properties, though constituents distribution throughout the film is strongly dependent on the biopolymer and filler ratio in the solution [266]. Finally, even though this method has been extensively used for research on films based on starch and protein, the difficulty in scaling up production volumes and the long drying times, make this technique impracticable on an industrial scale [267].

Tape-casting: films and coatings can also be prepared by the tape-casting technique at a larger scale than those usually reported by literature using the classical casting technique. In the tape-casting process, a suspension is placed in a reservoir with a blade, which height can be adjusted with micrometric screws [268]. The suspension is later dried on the same support, resulting in a film that can be removed from the surface. Depending on the film's characteristics, it can be rolled, cut, drilled, stamped, or laminated. The spreading of the film-forming solution (or suspension) can be done on larger supports or on a continuous carrier tape. The formed film is dried on the support, by heat conduction, circulation of hot air (heat convection), and infrared heating, resulting in a reduction of its thickness.

Extrusion: melt compounding or extrusion, is a conventional method widely used in the polymer industry and compound composites where the material is shaped through a die. Using extrusion to produce biopolymer-based composites could reduce manufacturing costs and render them more cost-competitive [265]. Extruders can be categorized on the number of screws in single, twin, and multiple screw extruders, further on the rotation mode of the screws can be classified in a single direction or in the opposite direction (corotating or counterrotating) or a mixture of both for a multiple screw extruder [262]. It is important to highlight that biopolymers processing by extrusion requires the use of additives, such as plasticizers and antioxidants to thermo-plasticize the polymer mix and avoid its degradation. However, it is well-known that extrusion processing these kinds of materials is not simple; hence optimization of the operating conditions (screw speed, configuration, and processing temperature) for each composite is necessary.

Blowing: blowing is a process that involves using air or nitrogen to inflate a tube of the melt as it comes out from the die. The blown film usually grows in a vertically upward direction. The die most often has a circular (annular) geometry, which is the simplest and most convenient solution even though the resulting film is less homogeneous. The thin bubble is then drawn by a series of nip rollers, flattened, and wound up in a reel. Both drawing and blowing orient the polymer molecular chains in a preferred manner. Depending on whether drawing or blowing prevailed the final film will be stronger in the longitudinal or transverse direction, respectively [269].

The inherent properties of biocomposite films are relevant for their applications. As it was stressed by Haniffa et al. [270], mechanical and thermal properties are regarded as the most relevant properties of biocomposite films. However, the physical and chemical characteristics of the main components can significantly alter these properties. The thermal, mechanical, barrier, and other relevant properties of diverse biocomposite films are included in Table 2. As it can be observed, the properties of these materials depend on many factors, being the most relevant: the type of polymer matrix, the filler type and concentration, and the selected processing method and conditions. Consequently, when comparing properties and characteristics of different films based on the same biopolymer and filler, it is necessary to consider the filler concentration and size, as well as processing technique and conditions.

3.2 Active biobased composite and nanocomposites

Active containers, usually used in food packaging, are those containing some substance capable of preserving the organoleptic or sensory characteristics of a product to ensure its quality. Of special interest are those active packages that contain natural antioxidants and antimicrobials that not only extend the shelf-life of packaged products by preventing rancidity reactions but also prevent the growth of foodborne pathogens [278]. Biodegradable and biobased polymers are preferred in the development of active materials for single used food packaging due to their low environmental impact

Table 2 Thermal, mechanical, barrier, and other interesting properties of biocomposite films

Biopolymer	Filler and concentration	Processing technology	Thermal properties (°C)	Mechanical properties	Water vapor barrier	Other interesting properties	References
Cellulose	Napier grass cellulose fibrils (NGCFs) (0, 5, and 25%wt)	Casting solution	$T_D{}^a = 358 - 363$	$\sigma_m{}^e = 49.7–107.9$ MPa $\varepsilon_B{}^f = 7.8–12.7\%$	-	Good interaction filler-matrix Random orientation of NGCFs	[120]
Microfibrillated cellulose	Kaolin clay and calcium carbonate (0, 2.5, 5, 7.5, and 10%wt)	Casting solution	-	-	1.04–2.49 10^{-2} gm/(m^2 day)	Low density	[271]
Nanocellulose	Barium titanate nanoparticles (0, 30, 40, 50, and 60%wt)	Casting solution	$T_D{}^a \simeq 300$	-	-	Dielectric/electrical property Piezoelectric performance	[272]
Thermoplastic starch	Carbon ashes (0, 7, 14, and 21%wt)	Melt mixing and compression molding	$T_m{}^b = 167.51–191.95$ $T_g{}^c = 97.70–146.37$	$E^d = 27.87–32.19$ MPa $\sigma_m{}^e = 2.27–2.65$ MPa $\varepsilon_B{}^f = 33–66\%$	-	Degree of crystallinity = 12.7–22.2% Ashes decreased 70% the starch deterioration	[246]
Thermoplastic starch	Talc nanoparticles (0–5%wt)	Melt mixing and thermocompression	$T_D{}^a = 255–275$ $T_m = 144.3–156.4$ $T_{g1}{}^c = -51.8–(-45.4)$ $T_{g2}{}^c = 45.9–46.9$	$E^d = 22.7–38.2$ MPa $\sigma_m{}^e = 1.19–2.34$ MPa $\varepsilon_B{}^f = 59.0–62.2\%$	0.6–1.3 10^{-10} g/(m s Pa)	Degree of crystallinity = 5.4–5.6% Oxygen permeation = 388.7–527.8 cm^3 mil/(m^2 day) Thermosealing capacity	[244, 273, 274]
Thermoplastic starch	Bentonite particles (0–5%wt)	Melt mixing and thermocompression	$T_m{}^b = 98.5–96.9$ $T_g{}^c = -66.3–(-65.0)$	$E^d = 50.9–180.9$ MPa $\sigma_m{}^e = 1.96–3.89$ MPa $\varepsilon_B{}^f = 38.4–96.2\%$	-	Degree of crystallinity = 5.6–9.6% Eventual particles intercalation	[161, 275]
Thermoplastic starch	Granite sand particles (0, 1, and 5%wt)	Melt mixing and thermocompression	$T_D{}^a = 317–321$ $T_m{}^b = 147.4–161.2$	$E^d = 80–405$ MPa $\sigma_m{}^e = 1.8–5.6$ MPa $\varepsilon_B{}^f = 14.7–25.9\%$	-	Translucent UV absorption capacity	[276]
Thermoplastic starch	Agave fibers and henequen fibers (0, 10, 20 and 30%wt)	Extrusion and injection molding	-	$E^d = 128–454$ MPa $\sigma_m{}^e = 11–23$ MPa $\varepsilon_B{}^f = 10–463\%$	-	Fire retardant properties	[277]
Thermoplastic starch	Cassava and ahipa peel and bagasse (0.5 and 1.5%wt)	Melt mixing and thermocompression	$T_D{}^a \simeq 316$ $T_m{}^b = 156.5–229.0$ $T_{g1}{}^c = -43.3–(-35.8)$ $T_{g2}{}^c = 43.0–81.8$	$E^d = 78.2–217.9$ MPa $\sigma_m{}^e = 2.6–5.5$ MPa $\varepsilon_B{}^f = 33.7–76.5\%$	1.24–1.56 10^{-10} g/(m s Pa)	Light and UV blocking capacity	[98]
Potato starch	Coffee silverskin (0–10%wt)	Casting	-	$E^d = 450–1800$ MPa $\sigma_m{}^e = 5–32$ MPa $\varepsilon_B{}^f = 2–22\%$	WVTR 49.4–57.9 g/(m^2 day)	Increases the films surface hydrophobicity and roughness Antioxidant and UV-protection capacities	[121]
Soy protein isolate	Soybean straw treated and nontreated (0 and 5%wt)	Casting	-	$E^d = 298–5527$ MPa $\sigma_m{}^e = 4.6–9.0$ MPa $\varepsilon_B{}^f = 15–35\%$	9.8–12.8 10^{-10} g/(m s Pa)	Moisture = 12.8–15.9% Solubility in water = 36–66%	[254]

Table 2 (continued)

Biopolymer	Filler and concentration	Processing technology	Thermal properties (°C)	Mechanical properties	Water vapor barrier	Other interesting properties	References
Soy protein isolate	Waste from red algae *Gelidium sesquipedale (Rodophyta)* (0–9%wt)	Extrusion and compression molding	$T_D{}^a > 300$	$E^d = 75.0–100.2$ MPa $\sigma_m{}^e = 4.3–5.9$ MPa $\varepsilon_B{}^f = 2.1–65.1\%$	2.29–8.57 10^{-12} g cm/(cm^2 s Pa)	Rough surface with low gloss Greenish and yellowish color of algae waste and soy protein	[250]
Polyhydroxyalkanoates (PHAs)	Fibers of *Posidonia oceanica* (0–30%wt)	Extrusion and compression molding	$T_D{}^a = 295–305$	$E^d = 1.24–3232$ MPa $\sigma_m{}^e = 21.88–24.80$ MPa $\varepsilon_B{}^f = 2.37–6.6.47\%$	-	Total degradation time < 6 months (in marine sediment)	[116]
Polyhydroxybutyrate (PHB) and polyhydroxyvalerate (PHB-HV)	Agave fibers (10, 20, and 30%wt)	Melt mixing and thermocompression	$T_{m1}{}^b = 135.7–151.7$ $T_{m2}{}^b = 148.7–161.8$ $T_g{}^c = 10.8–17$	$E^d = 413–770$ MPa $\sigma_m{}^e = 13.7–15.7$ MPa	-	Good fiber dispersion and compatibility with both matrices Impact strength = 24.5–34.4 J/m	[260]
Poly(β-hydroxybutyrate) / poly(ε-caprolactone)	Cellulosic particles (1%wt)	Melt mixing and injection molding	$T_{g1}{}^c = -53.0$ to -51.5 $T_{g2}{}^c = 17.5–19.7$	$E^d = 1007.7–1575.0$ MPa $\sigma_m{}^e = 24.6–34.9$ MPa	-	Incompatibility cellulosic particles-PHB/PCL	[258]
Poly (lactic acid) (PLA)	Banaba fiber (10, 20, and 30%wt)	Injection molding, extrusion + injection molding or extrusion + thermocompression	$T_D{}^a = 335–337$ $T_g{}^c = 68–69$	$E^d = 1400–1650$ MPa $\sigma_m{}^e = 15–55$ MPa	-	After 20%wt, the tensile strength decreased significantly and the flexural strength of the biocomposite was found to be maximum	[76]
Poly (lactic acid) (PLA)	Seaweed residue after alginate extraction (20, 30, and 40%wt)	Melt mixing and injection molding	$T_D{}^a = 292–310$ $T_{m1}{}^b = 127–149$ $T_{m2}{}^b = 142–157$ $T_g{}^c = 57–61$	$E^d = 3200–3500$ MPa $\sigma_m{}^e = 24.1–47.4$ MPa $\varepsilon_B{}^f = 0.8–2.0\%$	-	Composite show lower glow wire flammability index (GWFI) and no dripping of material upon burning, thus showing potential to be used in applications where high temperature occasionally occur	[52]

[a]Degradation temperature (T_D); [b]melting temperature (T_m); [c]glass transition temperature (T_g); [d]elastic modulus (E); [e]maximum tensile strength (σ_m); [f]elongation at break (ε_B)

[221, 275, 279, 280]. Among the available natural polymers and compounds, some can be obtained from agri-food industrial waste and their use would add value to these residues and keep them in circulation, one of the premises of circular economy. A clear example is the use of chitosan, a biodegradable polymer derived from chitin with antimicrobial activity that is obtained from the fishing industry waste such as crustacean exoskeletons [281]. Likewise, by-products and wastes from fruit and vegetables processing are an important source of compounds with high nutritional and functional value such as vitamins, minerals, antioxidants, and antimicrobial compounds, though are often discarded or derived for animal feed [282]. Essential oils (EOs) that can be obtained from these sources have been widely studied as additives for the development of active food packaging, mainly due to their antioxidant and antimicrobial capacity and their GRAS (Generally Recognized As Safe) character [283, 284]. Therefore, the use of active compounds derived from agricultural by-products not only contributes to the recovery of these compounds with specific activities but also generates added value for them. EOs are mainly phenolic compounds derived from plant secondary metabolites with antimicrobial capacity and several therapeutic and health-promoting attributes. Several authors have reported that the addition of EOs can significantly affect the microstructure, mechanical and barrier properties of the material depending on how they are incorporated into the polymeric matrix [284–287]. The most commonly reported EOs incorporated in biodegradable matrices are rosemary, tea tree, cinnamon, oregano, clove, and thyme [86, 288–292]. For instance, Bof et al. [16] have developed and characterized active biodegradable films based on corn starch and chitosan (CS:CH) with the addition of lemon essential oil (LEO) and grapefruit seed extracts (GSE). The inclusion of these active compounds, which are by-products of residues derived from citrus processing, did not affect the mechanical properties of the material and provided antimicrobial capacity by contact. Similarly, Kanmani and Rhim [293] developed antimicrobial active films with GSE on carrageenan matrix, with additional UV barrier capacity, particularly important for UV-sensitive food packaging. Further interesting biocomposites materials with EOs were reviewed in Table 3.

On the other hand, several investigations in recent years have focused on studying the effects of the incorporation of metal or metal oxides nanoparticles in biodegradable matrices [304, 305]. Starch-based and nanoclay biocomposites have been the most studied [212, 220, 306–308], although other biopolymer matrices have been used, such as chitosan [309], agar [310, 311], proteins [312, 313], or their combination [298, 314]. Usually, this strategy manages to improve the mechanical properties and susceptibility to the water of the system and, if the nanoparticles have antimicrobial activity the composite material also acquires this property. For instance, Abreu et al. [220] incorporated silver nanoparticles into wheat starch films obtained by solvent evaporation (casting). The materials obtained presented lower hydrophilicity and bacteriostatic activity against *Staphylococcus aureus* and *Escherichia coli*. Likewise, Ortega et al. [221] showed that the incorporation of AgNPs in cornstarch-based films decreased the solubility of the material and improved the mechanical properties by increasing Young's modulus and tensile stress values without decreasing deformation at the break. Similar effects have been reported by Malathi and Singh [315] by TiO_2 nanoparticles addition into rice starch films. In this case, the authors showed that the nanocomposite material presented a better water vapor barrier, enhanced mechanical properties, and bacteriostatic activity against *Escherichia coli*. The incorporation of ZnO nanoparticles has also proven to be an interesting strategy for improving the properties and adding bactericidal activity to starch films. In this regard, Mirjalili et al. [316] showed that starch-based films containing ZnO nanoparticles have better mechanical properties and antimicrobial activity against *Escherichia coli* and *Staphylococcus aureus*. Likewise, Nafchi et al. [225] reported that the incorporation of ZnO nanobars reduces hydrophilicity, improves mechanical properties, and provides antimicrobial activity against *Escherichia coli* to sago starch films. Yet, Guz et al. [210] demonstrated that the properties of starch films containing ZnO nanobars depend strongly on their size.

Nanoparticles and EOs biocomposites have also been studied with other innovative applications, such as wound dressing [45, 317]. Table 3 summarizes different types of nanoparticles included in nanocomposite formulations, being AgNPs the most commonly used for food packaging applications since they effectively reinforce biopolymer matrices and provide antimicrobial capacity over a broad spectrum of bacteria, virus, and fungi [185, 186, 221, 318]. Although numerous nanocomposites or active systems have been studied, the available literature refers mainly to materials manufactured with the solvent evaporation methodology, which is difficult to scale up. The development of biodegradable nanocomposite materials through scalable technologies is essential for the industrial implementation of such systems. Even though extrusion is a continuous and scalable processing technology, the study of the effect of extrusion on nanoparticles or bioactive molecules is still required. In this regard, it is necessary to analyze the mixing and distribution of the components within the matrix, the resulting interactions, and their effect on the material properties, as well as the possible degradation of the additives during the manufacturing process. Recent studies have successfully extruded biocomposites of thermoplastic starch with nanoparticles and EOs [294, 319].

Table 3 Active biocomposite and nanocomposite films relevant properties

Biopolymer	Active compound	Processing technology	Mechanical properties	Water vapor permeability	Solubility (%)	Antimicrobial activity	Antioxidant capacity	References
Cassava starch	ZnO nanorods (1%wt)	Casting solution	$\sigma_m{}^a = 2.8–3.2$ MPa $\varepsilon_B{}^b = 50–58\%$	$4.7–5.3\ 10^{-10}$ g/m s Pa	8–16	Inhibition of *E. coli* growth	-	[210]
Cornstarch	Green synthesized AgNPs in situ (0–143 ppm)	Casting solution	$E^c = 36.9–166.8$ MPa $\sigma_m{}^a = 161.6–368.1$ MPa $\varepsilon_B{}^b = 38.1–39.7\%$	$1.6–2.2\ 10^{-10}$ g/m s Pa	29 – 34.4 (25 °C) and 34.8–52.4	Concentration up to 28.6 ppm inhibition by contact and higher than 71.5 ppm halo of *E. coli*, *Salmonella spp*, *Penicillium spp*, and *S. aureus*	-	[184]
Cornstarch	AgNPs synthesized with lemon juice (0–143 ppm)	Casting solution	$\sigma_m{}^a = 2.1–4$ MPa $\varepsilon_B{}^b = 41–55\%$	$0.63–1.62\ 10^{-10}$ g/m s Pa	34.6 – 46	Concentration up to 28.6 ppm inhibition by contact and higher than 71.5 ppm halo of *E. coli*, *Salmonella spp*, *Pseudomona spp*, and *S. aureus*	-	[186]
Thermoplastic maize starch	Chitin nanoparticles (0–9%wt)	Casting solution	$\sigma_m{}^a = 3.89–9.32$ MPa $\varepsilon_B{}^b = 30–87\%$	$7–12\ 10^{-11}$ g/mhPa	-	Against *S. aureus* and *E. coli*	-	[247]
Cassava starch TPS	Rosemary extracts (2.5–10%wt of starch)	Extrusion + thermo-compression	$E^c = 9.3–23.3$ MPa $\sigma_m{}^a = 1.75–2.28$ MPa $\varepsilon_B{}^b = 50–70\%$	$2.62–4.94\ 10^{-10}$ g/m s Pa	20–25	-	29.5–140.9 (µmolTE/g DPPH method) in hydrophilic, lipophilic, and acid simulants	[294]
Rice starch and fish protein	Oregano EO (4–8%wt)	Casting solution	$\sigma_m{}^a = 3.9–6.8$ MPa $\varepsilon_B{}^b = 83.5–108.8\%$	0.18–0.21 g mm m^{-2} kPa^{-1} h^{-1}	8–22.6	-	4.2% DPPH inhibition and 63.1% peroxidase inhibition	[290]
Jackfruit starch and PVA	Jamun extract and/or nutmeg oil (10%wt) and ZnO nanoparticles	Casting solution	$\sigma_m{}^a = 14.11–27.22$ MPa $\varepsilon_B{}^b = 2.8–26.6\%$	-	5–28	Inhibition of *S. typhimurium* growth	-	[295]
Chitosan	Rosemary EO (0.5–1.5%v) + montmorillonite nanoclay (1–5%wt)	Casting solution	$\sigma_m{}^a = 63.7–83.0$ MPa $\varepsilon_B{}^b = 3.0–4.3\%$	$0.31–0.47\ 10^{-10}$ g/m s Pa	9.5–15.0	With 1.5% Rosemery EO inhibition of *S. agalactiae*, *E. coli*, and *L. monocytogenes*	1.4–2.2 mg gallic acid/g sample	[288]

Table 3 (continued)

Biopolymer	Active compound	Processing technology	Mechanical properties	Water vapor permeability	Solubility (%)	Antimicrobial activity	Antioxidant capacity	References
Chitosan	Protocatechuic acid (0.5–2%w/v)	Casting method	$\sigma_m{}^a$ = 40.8–26.7 MPa $\varepsilon_B{}^b$ = 62.8–7.9%	12.69–9.90 10^{-11} g/m s Pa	19.2–31.4	-	Antioxidant activity in aqueous and fatty food simulants	[296]
Chitosan	AgNPs with Tea polyphenol (0.1%)	Casting method	$\sigma_m{}^a$ = 32.8–53.7 MPa $\varepsilon_B{}^b$ = 25.9–12.6%	39.0–16.7 10^{-9} g m^{-1} min^{-1} Pa^{-1}	-	Films showed antibacterial activity to both *E. coli* and *S. aureus*	Films with TP-AgNPs has improved antioxidant capacity	[48]
Chitosan	Oregano and or cinnamon leaf EOs (0.25%wt)	Casting method	E^c = 1078–1471 MPa $\sigma_m{}^a$ = 40–43 MPa $\varepsilon_B{}^b$ = 5.9–6.2%	5.9–6.9 g mm m^{-2} kPa^{-1} h^{-1}	-	Reduce the growth of total aerobic and coliform microorganisms on sliced pork meat	-	[297]
Chitosan and rice starch	AgNPs (0 – 38 ppm)	Casting solution	$\sigma_m{}^a$ = 66.8–74.5 MPa $\varepsilon_B{}^b$ = 4.6–7.6%	WVTR 47.60–59.21 g m^{-2} day^{-1}	-	Inhibition of *E. coli*, *S. aureus*, and *B. cereus*	-	[298]
Chitosan and corn-starch	Lemon EO (1–3%wt)	Casting solution	E^c = 84–30 MPa $\sigma_m{}^a$ = 6.7–11.8 MPa $\varepsilon_B{}^b$ = 62–66%	7.5–7.4 g mm m^{-2} kPa^{-1} h^{-1}	20–23.7	Inhibition by contact of *Listeria innocua, E. coli*, *Alternaria alternata*, *Rhizopus stolonifer*, *Penicillium expansum*	No antioxidant activity was detected (TEAC method)	[16]
Chitosan and corn-starch	Grapefruit seed extract (1–3%wt)	Casting solution	E^c = 120–63 MPa $\sigma_m{}^a$ = 14.4–14.6 MPa $\varepsilon_B{}^b$ = 58.3–67%	7.5–7.8 g mm m^{-2} kPa^{-1} h^{-1}	24.7 – 26.1	*Inhibition by contact of Listeria innocua*, *E. coli*, *Rhizopus stolonifer*, and *Penicillium expansum*	No antioxidant activity was detected (TEAC method)	[299]
Chitosan-cassava TPS	Oregano and or cinnamon leaf EOs (0.25%wt)	Casting + melt blending + thermocompression molding (bilayer films)	E^c = 910–918 MPa $\sigma_m{}^a$ = 17–18 MPa $\varepsilon_B{}^b$ = 1.9–2%	7.87–8.03 g mm m^{-2} kPa^{-1} h^{-1}	-	Reduce the growth of total aerobic and coliform microorganisms on sliced pork meat	-	[297]
Curdlan + PVA	Thyme EO (1–2%)	Casting method	$\sigma_m{}^a$ = 10.4–11.5 MPa $\varepsilon_B{}^b$ = 190–225.9%	1.2–1.6 10^{-10} g m^{-1} min^{-1} Pa^{-1}	-	Inhibition of *E. coli*	-	[292]

Table 3 (continued)

Biopolymer	Active compound	Processing technology	Mechanical properties	Water vapor permeability	Solubility (%)	Antimicrobial activity	Antioxidant capacity	References
Whey protein	Oregano or garlic EO (2%wt)	Casting solution	$\sigma_m{}^a = 0.3–1.4$ MPa $\varepsilon_B{}^b = 5–15\%$	-	-	Against *S. Enteriditis*, *E. coli O157:H7*, *L. monocytogenes*, *S. aureus*, and *Penicillium spp.*		[299]
Gelatin	Red propolis ethanolic extract + clove or basil EO (0.15–0.8%wt)	Spreading technique	$E^c = 3.4–7.6$ MPa $\sigma_m{}^a = 19.3–42.7$ MPa $\varepsilon_B{}^b = 47.2–75.4\%$	0.27–0.40 g mm m^{-2} kPa^{-1} h^{-1}	84.6 – 92.4	Against *S. aureus*, *L. monocytogenes*, *S. enteriditis*, and *E. coli*	4.6–41.8% (ABTS method); 5.5–76.6% (DPPH method)	[300]
Gelatin-chitosan	3-phenylacetic acid (1–4%)	Electrospinning	-	5.74–11.22 10^{-15} kg/m s Pa	15.3–28.9	Against *S. aureus* and *S. enterica*	-	[301]
Gelatin-PLA	Encapsulated eugenol (0–4 mg/g)	Electrospray-electrospinning	$E^c = 19.3–21.6$ MPa $\sigma_m{}^a = 55.9–64.4$ MPa $\varepsilon_B{}^b = 6.01–6.75\%$	-	-	Against *S. aureus* and *E. coli*	12.15–33 mg DPPH/$g_{DB}{}^{(*)}$ 17.8–43.8% radical scavenging	[302]
PLA	Copper salt ion-exchange zeolite nanoparticles (1–3%wt) + nanocellulose (1.5–2%wt)	Dual electrospinning	$E^c = 602–807.9$ MPa $\sigma_m{}^a = 2.3–3.3$ MPa $\varepsilon_B{}^b = 4.1–13.1\%$	5.73–5.87 10^{-15} g/m s Pa	-	Against *S. aureus* and *S. typhi*	-	[303]

[a]Maximum tensile strength (σ_m); [b]elongation at break (ε_B); [c]elastic modulus (E); [(*)]dry basis

Particularly, since many biopolymers are hydrophilic, the naturally hydrophobic EOs must be compatibilized with the polymeric matrix generally by the addition of surfactant agents. The most widely used technique for this purpose includes the formation of an emulsion or a nanoemulsion to ensure the homogeneous distribution of the active compound in the matrix. Moreover, given the volatile nature of EOs, the processing conditions must be optimized to minimize activity loss. Consequently, when extrusion processing is desired EOs need to be protected from the drastic treatment conditions for which purpose encapsulation is a useful tool [320]. Alternatively, Li et al. [302] have efficiently encapsulated eugenol (83.4 to 92.7 g/100g) in PLA and gelatin nanofibers obtained by electrospinning. Electrospinning has also been used by Scaffaro et al. [321] to modulate EO migration from solvent casting films in a multilayer composite material. Active laminates were formulated with a PLA film containing carvacrol (14%wt) obtained by solvent casting and one or two fibrous layers of PLA applied by electrospinning. These fibrous layers modulated the carvacrol release kinetics, which proved to progressively reduce the burst delivery at an early stage of immersion, therefore increasing the delivery device lifespan from 288 to 795 h.

Inactive and nanocomposite materials, the study of the microstructure helps to understand the interrelation of the structure with the properties that determine the performance of the material, in addition to being able to infer the effectiveness of the filler inclusion within the matrix. In general, when homogeneous surfaces without pores or cracks as well as compact cross-sections are visualized by SEM the included active compounds or nanoparticles are compatible with the polymeric matrix, which also leads to its reinforcement. These observations agree with the reported mechanical behavior of the nanoreinforced materials listed in Table 3. Correspondingly, Bof et al. [16] stressed that the presence of discontinuities in the matrix of cornstarch/chitosan composite films with lemon EO (visualized as oil microdroplets by SEM) was indicative of the lack of miscibility of the active compound with the polymer and lead to poor gas barrier properties. A similar trend was found by other authors working on biodegradable films containing different EOs such as orange peel, tea-tree, and ginger oil, among others [286, 287, 322]. Consequently, the compatibility of the active agent with the matrix determines how efficiently the former is incorporated into the polymer network and therefore the film microstructure characteristics. In this regard, ATR-FTIR spectroscopy is a useful technique that has been widely employed to study the interaction among the composite constituents. In the same work, Bof et al. [16] found by FTIR analyses that hydrogen bonding occurs in the corn starch and chitosan blend films containing grapefruit seed extract as filler, leading to a more compact and denser film structure. Similarly, Sharifi and Pirsa [320] working on black mulberry fruit pulp pectin films studied the effect of both chlorophyll encapsulated with carboxymethylcellulose and silica nanoparticles addition. SEM analysis indicated that both active components act as fillers of the pectin structure. Moreover, their high compatibility was demonstrated through FTIR studies since spectra confirmed electrostatic interactions between pectin chains with encapsulated chlorophylls and silica nanoparticles. Besides, thermogravimetric analysis (TGA) results revealed that the simultaneous addition of these active compounds increases the thermal stability of the film [16, 46, 247].

Biodegradable films formulated with polymers derived from biomass are, in general, hydrophilic in nature due to the presence of a large number of hydroxyl groups in their structures. These biomaterials are susceptible to humidity: water causes a weakening of the intra- and intermolecular bonds (plasticizing effect) which leads to an increase in the WVP of this type of polymers [323]. Given the hydrophobic character of EOs, it is expected that their inclusion in a polymeric matrix formulated with biopolymers improves their water susceptibility, measured by WVP, water absorption capacity, swelling, contact angle, solubility, and moisture content. This trend has been widely reported in the literature and is summarized in Table 3. However, as already mentioned, the microstructure of the biocomposite is decisive, especially regarding barrier properties.

In general, the addition of nanoparticles exerts a reinforcement effect on biopolymeric matrices, which has been evidenced in the improved mechanical properties (enhancement of mainly tensile strength and elastic modulus) as well as barrier properties (Table 3). Ortega et al. [221], for example, stressed that AgNPs incorporation in cornstarch-based films maintained the material UV-barrier capacity while decreasing WVP with higher AgNPs concentration.

EOs incorporation usually confers both antioxidant and antimicrobial properties. In this regard, Varghese et al. [282] have reviewed the effect of EOs on the physical properties of biopolymer films highlighting the migration release of the active compounds to different food surfaces or simulant media as well as their action mechanisms. Their antioxidant capacity can be evaluated by diverse complementary techniques such as DPPH, ABTS, FRAP, ORAC, and total phenolic compounds content. The addition of some nanoparticles also provides antimicrobial properties to the materials, such as in the case of silver, zinc, copper, and chitin, among others. The antimicrobial capacity is evaluated over common foodborne microorganisms frequently by the agar disc diffusion method. It has been observed that, even though nanocomposite films generally exhibit inhibition by contact, the observation of an inhibition halo depends on the nanoparticle concentration (Table 3). For instance, nanocomposite corn starch films containing AgNPs concentrations greater than 71.5 ppm inhibit the growth of *E. coli*

ATCC and *Salmonella spp*. and exhibit the corresponding halo [221]. Several examples of different biopolymer matrices containing different active compounds and processed by casting, thermocompression, extrusion, or spread-coating are included in Table 3, considering also the effect of different processing methodologies as well as the protection of these active compounds by encapsulation or electrospinning.

Several studies have been carried out on active composites biodegradation kinetic though the comparison of results depends on the conditions of assay. Rech et al. [324] evaluated the biodegradation of eugenol-loaded PHB films obtained by casting and buried for 60 days in agricultural, sandy, and landfill soil. The soil acidity, phosphorus availability, moisture level, and polymer crystallinity were key factors in explaining the differences in microbial growth and biodegradation rates. PHB films buried in agricultural soil presented a higher rate of biodegradation, which may be associated with the high fungi load and higher soil-phosphorus availability. Likewise, Castro-Aguirre et al. [325] evaluated the effect of bioaugmentation on the biotic and abiotic degradation of PLA and PLA bionanocomposites (BNCs) in simulated composting conditions. Bioaugmentation with *Geobacillus* increased the evolution of CO_2 and accelerated the biodegradation phase of PLA and BNCs in compost environments. Finally, it is important to remark that the evaluation of the ecotoxicological impact on the composting soil is necessary. In this regard, Salehpour et al. [326] analyzed the effect of PVA films containing cellulose nanofibers on municipal solid waste composting quality after film biodegradation. An ecotoxicological test revealed that nanocomposite films did not generate any negative effects on germination or development of the studied vegetal species (cress and spinach).

3.3 Sustainable composite foams

Foam-like polymer materials, such as expanded polystyrene (EPS) and polyurethanes (PUs), are greatly desired for their lightweight and insulation properties. Due to their non-renewable resource basis and high resistance to abiotic and biotic degradation, biobased alternatives are under study. In this regard, innovative more sustainable PU foams have been developed using biobased polyols and recycled polymers [327, 328], tannins extracted from lignocellulosic wood by-products [329–335], and other biopolyols obtained from agri-food waste and by-products such as crops straws [336] and citric peels [337]. Among fully biobased composite foams studied as an alternative to PU insulation materials, the alginate and orange peel biocomposite foams developed by Vincent et al. [338] also present fire-retardant properties, an important safety feature for building materials. Owing to their mechanical performance, easy confirmation and biocompatibility PUs are also attractive for tissue regenerating scaffolds. Yet, because of their heterogeneous structure and the nature of their building blocks PUs, are quite resistant to biodegradation even in the absence of stabilizing additives [339], therefore new fully biodegradable scaffolds have been developed for biomedical applications [340–343]. Different natural fillers are used as reinforcement of biobased PUs, some of which can be obtained from agri-food waste [344–346].

Expanded polystyrene (EPS), on the other hand, has been broadly used in packaging such as disposable food containers and protective packagings for products susceptible to mechanical damage, due to its low-density, moisture resistance, thermal insulation, and low-cost. In addition, other characteristics such as high durability, acoustic insulation, and strength have proven EPS useful for building and construction [347, 348]. In addition, compared to other conventional food packaging materials, EPS shows 7–28% lower environmental impact than aluminum containers and 25% less than disposable polypropylene (PP) ones, even when reusable PP containers are considered [349]. Given its versatility and performance, EPS demand has had a marked increase with the consequent increase in the amount of waste generated of this nonbiodegradable synthetic polymer [347]. Besides, EPS is difficult to collect inadequate conditions for recycling, since large plastic waste volumes are generated and washing water is needed. Moreover, even though closed-loop recycling systems show better performance in greenhouse gases emission (GGE) and energy consumption, as well as lower landfilling, optimized recycling technologies, and waste management systems are needed to reduce the large water volume consumption and scrap generation or sorted materials that cannot be reused [350, 351]. To reduce EPS waste accumulation, its use as composite filler or raw material for building and construction materials, such as bricks [352, 353], thermal or acoustic insulation materials [347, 354–356], and as an absorbent substrate for gaseous pollutants removal [357] has been studied.

Furthermore, biodegradable biobased foams are being investigated as substitutes for EPS, especially for disposable packaging applications. In this regard, a study by Razza et al. [358] indicated that a 50% reduction in non-renewable energy resources consumption and 60% lower GGE could be achieved with starch-based prototype packaging materials obtained by microwave technology in comparison to benchmark EPS cushioning packing. Various bioplastics-based foams from starch to PLA and their mixture have been reported [21, 79, 106, 126, 359–371]. These materials should be cheap, lightweight to minimize transportation environmental impact as well as compressible, and strong enough to prevent physical damage to the product. In general, foams based on biopolymers such as polysaccharides or proteins are susceptible to moisture sorption and its consequent effect on permeability and mechanical properties.

Thus, biocomposites and nanocomposites are proposed to enhance their water and mechanical resistance [21, 79, 106, 361, 362, 372, 373]. Most of the studied fillers are derived from agri-food waste and by-products, aiming to reduce materials cost and reinforce their sustainable character without compromising their biodegradability. Bergel et al. [371] found that contents up to 20%wt. of the modified starches reduced water absorption and increased impact resistance. Similarly, Guan et al. developed composite acetylated starch-based foams with corncob fiber and cellulose [374] or with PLA [375]. Crosslinked starch foams presented promising results, with higher thermal stability, and lower water sorption and improved flexion properties [368, 376]. A composite blend of starch with plant proteins, kraft fibers, palm oil, or chitosan studied by Kaisangsri et al. showed enhanced mechanical performances [366, 377]. Furthermore, biodegradable hydrophobic coatings have been also studied as a promising alternative to improve biocomposite foam's water resistance [364, 369, 370, 378].

Several technologies have been studied to obtain biobased foams, from extrusion-cooking (a method commonly used in the food industry) for loose-fill cushioning materials [367] to thermoforming for containers production [126, 360, 363]. Soykeabkaew et al. [379] summarized a wide range of processing techniques for starch-based foams, among which the most common are extrusion, baking (thermoforming), microwave, freeze-drying/solvent exchange, and supercritical fluid extrusion. The same process technologies are used for other biobased foams [373, 380–383].

During thermoforming, the porous structure of the foam is formed by insufflation of gas in the molten polymer blend, which expands as pressure is reduced, or by gas formation within the batter due to the use of chemical blowing agents that produce gas by thermal decomposition or chemical reaction. Carbon dioxide is currently the most widely used gas for physical blowing of polymer foams as an eco-friendlier alternative to hydrochlorofluorocarbons (HCFCs), due to its low toxicity, high stability, and low-cost [384]. Some foams are obtained by air diffusion into the battery by whipping before curing [385]. Supercritical inert gases, like carbon dioxide and nitrogen, are used as more environmentally friendly alternatives to blowing foams [373, 381, 386–388]. Yet these blowing agents require specific and expensive equipment to work under high-pressure conditions, thus chemical blowing agents that are easily incorporated during mixing in the polymer matrix are sometimes preferred [384]. The latter leads to highly diffusing gas molecules (CO_2, N_2, and H_2) resulting in open-cell structures that limit the foam's fields of application. Sodium bicarbonate is another well-known low-cost chemical blowing agents, commonly used with citric acid being both safe for food contact usage [389]. Urea was also studied as a blowing agent in biocomposite starch foams considering both its plasticizing and cross-linking properties in starch matrices [126]. A vast variety of chemical blowing agents have been reviewed by Coste et al. [384], yet the authors highlighted that unreacted blowing agent and by-products can compromise the material's properties and toxicity due to migration throughout their life cycle. Consequently, various parameters and reaction conditions should be determined for each blowing agent/polymer system to meet the final material requirements with no side effects. As regards, biopolymeric composite foams water vapor is mainly used as a blowing agent [21, 24, 390]. Water-based polymer and filler batters are prepared and baked at 140–220 °C where water vaporizes inflating the batter as it dehydrates and solidifies forming the foam's cells.

In thermoforming processing, either thermoplastic polymer pellets are used [365, 383, 391] or biopolymers water-based batters are prepared and poured onto the preheated mold [21, 126, 359, 390, 392–394]. In this regard, molding time, temperature, and pressure must be optimized according to the batter formulation [126]. Additives such as magnesium stearate as a release agent, guar gum as solids suspension stabilizer and glycerol as plasticizer have been reported [106].

A key problem for natural biopolymers foams, mainly starch-based materials is their water susceptibility; therefore several strategies have been studied to overcome this limitation. Biocomposite foams with several fillers have been studied to reduce water uptake [21, 124, 140, 280, 359, 361, 379, 395]. Moreover, despite of the increase in process complexity and cost, the use of coating with other biodegradable bioplastics, such as PLA, or natural waxes has also been reported as a promising alternative to enhance the water-resistance of starch-based biocomposite foams [78, 364, 370, 393].

Biobased foams obtained by microwave from extruded biobased thermoplastic were studied by various authors [358, 383, 396–399]. Besides, Razza et al. [358] indicated that biodegradable and biobased foams expanded by microwave technology resulted in more sustainable materials than EPS benchmark material. Alternatively, noteworthy starch and alginate with micro-fibrous clay composite foams with fire retardant properties were developed via lyophilization by Darder et al. [400]. Nonetheless, the industry-scale design of these processing technologies is needed for future research, with a particular focus on process energy consumption optimization.

Unlike biocomposite films, foam's properties depend not only on its components (polymer, filler, plasticizer, and other additives) intrinsic characteristics and interaction within the composite structure but unequivocally due to the type, number, and size distribution of pores of the foam. Foam structures are cataloged as open or closed-cell types depending on whether pores are interconnected or isolated keeping the gas or air trapped inside the foam, respectively. Therefore, open-cell systems are more permeable to gases and usually less rigid, resulting in less attractiveness for specific building and insulation and sound-canceling requirements. Generally, more homogenous and smaller cell size structures derive in higher density materials with higher compressive strength [362, 364, 383, 401].

Comparatively, mechanical resistance to compression and flexion is usually increased with filler content in biocomposite foams as shown in Table 4, though optimal filler content depends on its nature and size. Water uptake, on the other hand, is strongly dependent on filler nature. For instance, Machado et al. [359] observed a marked increase in foams contact angle with peanut skin in starch-based foams, indicating an increase in the material's hydrophobicity. Similar results were shown by sesame oil cake residue on starch foams [21], yet others such as sunflower oil cake residual particles presented the opposite effect [126]. Similarly, the biodegradability of foam biocomposites tends to be favored by natural fillers content, some evidencing higher biodegradation rates than others according to assay conditions [21, 126, 395].

A wide range of biocomposite foams have been developed and studied over the last decades with diverse potential applications. Nevertheless, to reinforce their sustainable character further investigations on the use of renewable waste products as raw materials, cost-effective and low-energy-consuming processing technologies and innocuous foaming agents are needed [85].

4 Applications of biobased composites

4.1 Active and intelligent food packaging

Active and intelligent packaging materials protect and preserve food ensuring its microbiological, organoleptic, and nutritional quality until it reaches the final consumer. Unlike traditional materials, active and intelligent packaging is polymeric matrices that serve as vehicles for a wide variety of additives such as antimicrobials, antioxidants, gas absorbers, and pH indicators among others, depending on the primary mode of deterioration of the food that limits its shelf life (Figure 6). Intelligent food packaging systems are small, simple, and inexpensive real-time indicators of food quality or storage conditions. In this regard, Firouz et al. [407] have critically reviewed intelligent and active packaging in the food industry; meanwhile, Yang et al. [408] have summarized the advanced applications of chitosan-based hydrogels as both, biosensors, and intelligent food packaging systems. Table 5 summarizes different types of intelligent systems and active packaging used in the food industry.

In the last 10 years, there has been an overwhelming advance in the development of nanocomposites, especially those with antimicrobial activity. These materials have been studied as active packaging for application in the food industry in order to eliminate or at least reduce the growth of pathogens responsible for foodborne diseases [304, 305, 310]. The most common developments include formulations containing AgNPs and ZnO based on starch, chitosan, gelatin, PVA, and starch among others [48, 295, 318, 430].

Ortega et al. [221] studied the effectiveness of nanocomposite starch-based films containing 143 ppm of AgNPs synthesized within the filmogenic suspension as active film packaging for a dairy product. These active films were able to extend the shelf-life of fresh cheese samples for 21 days. Regarding nanocomposite materials in contact with food, studies are necessary to evaluate not only the cytotoxicity of the nanoparticles themselves but also their migration to the food matrix. In this regard, tests are performed using different food simulants. Abreu et al. [220] carried out a contact test to determine if the nanostructured starch films comply with current European regulations without detecting a significant migration of Ag. Furthermore, Metak et al. [431], working on polyethylene containers with 1% AgNPs inclusion, did not detect Ag migration to the matrix or organoleptic changes in the packaged products. As regards the cytotoxicity of the silver nanoparticles, Bacchetta et al. [432] studied the effects of waterborne AgNPs on juvenile fish *Piaractus mesopotamicus* and analyzed toxicological endpoints such as metal burdens, oxidative stress, and genotoxicity. DNA damage in fish erythrocytes was observed after 24 h exposition at 25 µg/l AgNP. Bidian et al. [433] demonstrated that if AgNPs (0.8–1.5 mg/kg) were administered to offspring rats during pregnancy, they could cross the placental and testicular barriers and induced oxidative stress, DNA damage, and autophagy as mechanisms of cell toxicity. However, more research is still needed on this topic.

On the other hand, no migration of montmorillonite (MMT) nanoparticles from soybean protein matrices towards tuna fish muscle was detected through atomic absorption spectroscopy but MMT presence enhance the antimicrobial capacity of clove EO included in the formulation [291].

4.2 Biobased composites as fertilizer delivery and their applications as agricultural inputs

Biodegradable plastic mulch films have been developed and studied to reduce plastic waste generated from plastic mulch films disposal after crop harvest. Since the recovered films are usually greatly contaminated with soil and organic residues, their recyclability is difficult and not cost-efficient [434, 435]. The main reason is that mulch, especially thin mulch, cannot be picked up completely and that recollection is highly time and manpower-consuming [435]. Moreover,

Table 4 Morphology, density, mechanical performance, water related, and other interesting properties of some recent developments on bio-based biocomposite foams

Bioplastic	Filler and concentration	Treatments and coatings	Processing technology	Thickness (mm)	Density (g cm^{-3})	Cell size and density	Mechanical properties	Moisture content and water sorption	Other interesting properties	References
Glyoxal cross-linked cornstarch	Corn husk fiber and kaolin (0 and 10%wt dry starch basis)	Beeswax coating	Gelatinization and thermoforming	3.38–3.87	0.27–0.31	300–600 µm for only starch foam and 750–900 µm for beeswax foams	Flexion test $\sigma_m{}^a$ = 3.2–4.49 MPa $\varepsilon_B{}^b$ = 4.79–5.30% E^c = 134.2–109.6 MPa	9.64–10.31% 0.1–0.45 g water/gDB (both decrease with filler and coating)	The addition of kaolin, fiber, or beeswax appears to enhance the foaming ability of the trays	[402]
Cassava starch	Kaolin (3, 6, 9, 12, and15%wt)	-	Compression thermoforming	~ 4	0.21–0.29	370–810 µm (increase with filler content)	Impact test Maximum strength = 246.1–1151.6 J m^{-2}	Water sorption 9.79% for 3% kaolin foams at RH of 55%	Kaolin increase foams impact strength and density with kaolin content	[403]
Cassava starch	Malt bagasse (0 to 20%wt of solids)	-	Compression thermoforming	2.16 – 2.24	0.415 – 0.450	-	Tensile test $\sigma_m{}^a$ = 10.37–12.92 MPa $\varepsilon_B{}^b$ = 1.1–1.8% (at 58% RH)	0.18–0.21 g water/gDB (decreases with filler)	-	[106]
Cassava starch	Sesame cake (SC) (0 to 40%wt of solids)	-	Compression thermoforming	3.3–4.6	0.23–0.30 (filler addition decrease density)	-	Tensile test $\sigma_m{}^a$ = 0.43–1.16 MPa $\varepsilon_B{}^b$ = 2.0–3.3% E^c = 17.8–40.4 MPa Flexion test $\sigma_m{}^a$ = 1.9–7.4 MPa $\varepsilon_B{}^b$ = 1.1–1.4% E^c = 174–522 MPa	Water sorption capacity 141–154% (decreases with residue content)	> 20% SC biocomposite foams resulted in most mechanical properties comparable to commercial EPS	[21]
Cassava starch	Shrimp shell (SH) and egg shell (EG) (0 to 20%wt)	-	Compression thermoforming	~ 4	0.206–0.286 (increased by SH and decreased by EG)	Not reported, SH foams show denser structure and EG foams a more expanded one	Impact test Maximum strength = 144.18 J/m^2 (5% SH) and 171.18 J/m^2 (15% EG)	-	The izod impact strength of EG starch based reinforced foams increased in 300%	[362]
Cassava starch	Grape stalk (0–7%wt)	-	Compression thermoforming	~ 3	0.18–0.21 (filler decrease foam density)	Composite foams present a sandwich-type structure	Flexion test $\sigma_m{}^a$ = 2.5–2.9 MPa $\varepsilon_B{}^b$ = 1.3–1.9% E^c = 150–202 MPa (no significant differences between 0 and 7% filler content)	2.8–8.5% (increases with filler addition)	The more expanded structure of biocomposite foams favored biodegradation kinetics	[390]
Cassava starch	Peanut skin (0–24%wt of solids)	-	Compression thermoforming	~ 3	0.21–0.27	-	Tensile test $\sigma_m{}^a$ = 1.0–1.1 MPa $\varepsilon_B{}^b$ = 3.9–4.9% E^c = 25–26 MPa	9.0–9.7% (lower with filler addition)	Biocomposite show rougher surface with some holes, but with reduced hydrophilicity	[359]
Cassava starch	Macroalgae (*H. macrolaba*) (0 to 20%wt dry starch basis)	-	Compression thermoforming	~ 4	0.16–0.27	Biocomposite foams present smaller cell-size, higher cell density and thicker cell wall	Impact test Maximum strength = 33–45 J/m^2	-	Thermal stability of the foam was improved with filler addition	[404]
Cassava starch	Water hyacinth (WH) (0, 3, 5, 7, or 10%wt dry starch basis)	Beeswax coating	Compression thermoforming	~ 4	0.320–0.433	440–560 µm (filler decreases cell size and increases cell density)	Flexion test $\sigma_m{}^a$ = 2.95–3.42 MPa $\varepsilon_B{}^b$ = 3.09–3.52% E^c = 93–232 MPa	6.77–10% (lower value for 5%wt of WH)	Beeswax coating reduced water solubility over a 50%	[364]
Cassava starch	Sunflower oil cake (SOC) (0 to 40%wt of solids)	-	Compression thermoforming	~ 3	0.20–0.40 (increases with filler content)	169–278 µm (decreases with filler content)	Flexion test $\sigma_m{}^a$ = 3.18–6.26 MPa Compression test H^d = 3.91–21.69 kgf En^e = 31.2–118.2 mJ	12.3–12.8% 40.4–46.9 g water/gDB	Biocomposite foams show greater biodegradability	[126]
Cassava starch	Sunflower oil cake (SOC) (0 to 20%wt of solids)	Urea as blowing agent	Compression thermoforming	~ 3	0.15–0.23 (increases with filler content)	113–139 µm (decreases with filler content)	Flexion test $\sigma_m{}^a$ = 1.88–2.61 MPa Compression test H^d = 2.39–6.9 kgf En^e = 18.4–50.8 mJ	12–14.3% 41.9–50.4 g water/gDB	Biocomposite foams expanded with urea show greater biodegradability	[126]

Table 4 (continued)

Bioplastic	Filler and concentration	Treatments and coatings	Processing technology	Thickness (mm)	Density (g cm^{-3})	Cell size and density	Mechanical properties	Moisture content and water sorption	Other interesting properties	References
Cassava starch	Cassava inner bark (0–50%wt of solids)	Starch gelatinizing	Compression thermoforming	2.9–3.2	0.021–0.032	-	Flexion test $\sigma_m{}^a = 1.8$–4.5 MPa $\varepsilon_B{}^b = 0.80$–1.30% (filler decrease flexibility)	2.78–5.29 g water/gDB	Biocomposites with the incorporation of cassava inner bark presented lower water absorption	[405]
Cassava starch + Chitosan (0–6%wt)	Kraft fiber (0–40%wt of starch)	-	Compression thermoforming	-	0.11–0.15	-	Tensile test $\sigma_m{}^a = 0.30$–1.17 MPa $\varepsilon_B{}^b = 1.68$–2.07%	8.07–15.2 g water/gDB (decreases with fiber content)	Solubility decreased with chitosan and fiber addition	[377]
Oca starch	Sugarcane bagasse (0 to 40%wt of solids)	-	Compression thermoforming	2.57–2.60	0.157–0.272	-	Flexion test $\sigma_m{}^a = 0.5$–0.79 MPa $\varepsilon_B{}^b = 0.60$–1.10%	64.9–94.7 g water/gDB	SB addition above 20% decreases water absorption capacity and mechanical resistance	[372]
Oca starch	Asparagus peel fiber (0 to 40%wt of solids)	-	Compression thermoforming	2.49–2.61	0.144–0.291	-	Flexion test $\sigma_m{}^a = 0.51$–0.62 MPa $\varepsilon_B{}^b = 0.90$–1.60%	79.2–93.8 g water/gDB	Biocomposite density decrease at low filler content	[372]
Wheat starch	Barley straw, grape and cardoon filler (0 and 5%wt of solids)	-	Extrusion, thermo-compression and microwave foaming	~ 2.1	292–347	200–600 μm (average with a wide size range)	Compression test $\sigma_m{}^a = 1.24$–2.18 MPa $E^c = 43.7$–52.8 MPa $W^f = 1.91$–4.54 mJ m^{-3}	1 – 2.5%	Barley straw fibers use result in tougher biocomposites foams	[383]
Citric acid cross-linked potato starch	Microcrystalline cellulose powder (25%wt of solids)	Carnauba wax as mold release and hydrophobic agent	Compression thermoforming	~ 2.2	-	Cross-linked starch biocomposites show higher cell-density structures	Tensile test $\sigma_m{}^a = 1.76 - 2.25$ MPa $\varepsilon_B{}^b = 1.20 - 1.97$% Flexion test $\sigma_m{}^a = 3.76 - 7.61$ MPa $E^c = 445 - 600$ MPa	-	Foams crosslinked with 5% citric acid presented the better mechanical properties	[376]
PLA	Sepiolite (0 – 10%wt of total solids)	Commercial chemical foaming agent (CFA)	Injection molding	~ 4	0.82 – 1.30 (increases with filler content, decrease with CFA)	Biocomposite foams show smaller cell size and a higher cell density because nanoclay acts as cell nucleating sites	Flexion test $\sigma_m{}^a = 36.6 - 121.8$ MPa $\varepsilon_B{}^b = 1.79 - 8.89$% $E^c = 1883$–3464 MPa	-	The incorporation of nanoclays down to a 5%wt load improves the toughness of the nanocomposite but greater amounts act in detriment of it	[382]
PLA	Kenaf fiber powder (20%wt of solids)	Azodicarbonamide (ADC) as foaming agent	Extrusion and thermopressing	~ 3	0.87–1.13	61–78 μm (lower extrusion temperature)	Tensile test $\sigma_m{}^a = 2.77 - 18.66$ MPa	2.7 – 3.0% 0.05–0.24 g water/gDB (increases with blowing agent content)	-	[79]
Alginate	Orange peel (10%wt) + $CaCO_3$ (10%wt)	-	Oven dried (35 °C for 4 days)	1–3	40–42	Pore-size is difficult to quantify because of shape irregularities but were estimated to range between 100–500 μm	Mechanical properties comparable to those of commercial fire-retardant PU	14.67 ± 0.65%	Fire retardant properties and less hazardous combustion gases production than PU $k^g = 0.03$ W m^{-1} K^{-1} Cp$^f = 1600$ J kg^{-1} K^{-1}	[338]
PBS	Cellulose Nanocrystals (CNC) (0–0.5%)	CNC acetylation	Melt mixing and supercritical CO_2	~ 1	0.033–0.144 (decreases with filler)	27.7–33.2 μm 0.72–1.06 10^9 cells cm^{-3}	-	-	Thermal insulation increase with filler k^g decrease from 0.063 to 0.21–0.27 W m^{-1} K^{-1}	[406]

[a]Maximum strength (σ_m); (T_D); [b]elongation at break (ε_B); [c]elastic modulus (E); [d]hardness (H); [e]compression energy absorption (En); [f]compression energy absorption per unit volume (W); [g]thermal conductivity (k); [h]specific heat (Cp)

the different aging degrees of the recycled polymer films conversely affect the regenerated product properties [436]. Therefore, most collected films end up in landfills or are used for energy recovery, few countries, such as Germany, France, and Canada, have established recycling systems for plastic mulch waste [437]. In addition, large amounts of residual plastic waste may remain on the field leading to polluted soils with diminished porosity, moisture content, and increasing bulk density which affects quality, health, and fertility, as has been evidenced in Xinjiang Autonomous Region (China) where residual plastic films can reach 200 kg/ha, known as “white pollution” [1, 435, 438]. Moreover, these plastic residues enter the soil ecosystem by different routes depending on water flow, insects’ ingestion and egestion, root plant growth, and weathering in the soil leading to microplastic that can enter deep aquifers and hence the aquatic food chain [439]. Even though its long-term sustainability is arguable, soil covering is an established agronomical technique intended to increase yield and quality of the production by conservation of soil temperature and moisture, as well as weed growth control [434, 440]. Consequently, as food demand increases, plastic films consumption grows annually, with an estimated 1.4 million ton global market used to cover around 80,000 km^2 (equivalent to 0.6%) of the global arable land [1].

The latter has derived considerable research, development and commercialization of novel biodegradable, and biobased mulch films, that at the end of their lifetime degrade and mineralize in soil avoiding recollection and acting as in situ soil fertilizer [1, 246, 441, 442]. Various oil-based and biobased biodegradable polymer films and their blends and copolymers have been studied as mulch films [120, 435, 443–445]. Field studies have also been performed on commercial biodegradable bioplastics [434, 436], yet biodegradation rate is clearly dependent not only on bioplastic nature but also on climate conditions and soil characteristics. The EN 17033:2018, a new European Norm concerning “Plastics - Biodegradable mulch films for use in agriculture and horticulture - Requirements and test methods”, sets the standard methods necessary to determine biodegradability, performance, and environmental impact of biobased mulch films [1]. Additionally, additives liberation control and ecotoxicity testing after full biodegradation are also needed to ensure no further environmental impact [1, 446].

Biodegradable mulches can be obtained by thermo-plasticizing, casting, or spraying using renewable and biodegradable biopolymers such as starch, cellulose, chitosan, alginate, and glucomannan [442]. A wide range of studies on this type of mulching system is listed in Table 5. Natural fillers from renewable sources were incorporated into composite mulching films and coating to improve their mechanical performance, decrease water sensitivity, and/or confer better UV-light barrier properties for soil temperature

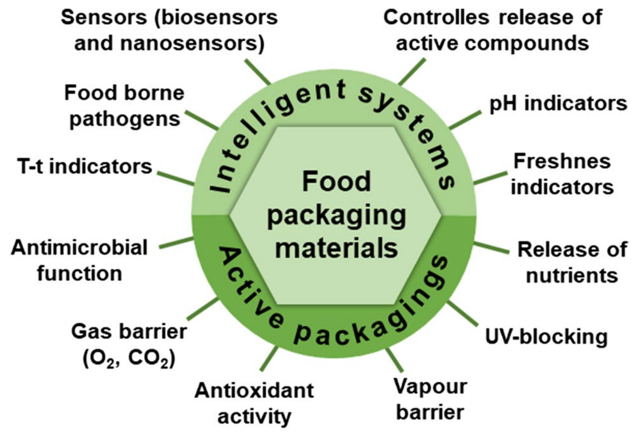

Fig. 6 Active and intelligent food packaging materials properties

conservation [420, 442, 447, 448]. The most recent works focus on spraying water solutions onto soil forming a biodegradable mulching coating directly in the field, using mainly biopolymers obtained from marine and agricultural waste and byproducts [419, 420, 442, 447, 449–453].

Likewise, excessive fertilization and run-off are known to pollute surface and groundwaters leading to eutrophication of lakes and rivers that, in turn, result in the deterioration of aquatic ecosystems due to algae bloom, oxygen loss, aquatic wildlife mortality, and the consequent biodiversity loss [454]. These, among other serious environmental issues derived from current agriculture practices, generated interest in new methods using innovative technologies to ensure high yields and quality of agricultural products while minimizing agrochemicals use. On the one hand, the study, extraction, and use of natural pesticides and herbicides derived from renewable sources have been promoted [455, 456]. On the other hand, in view of cleaner and more sustainable agriculture practices, the use of biodegradable composite fertilizers controlled-release systems has been considered to achieve more efficient use of nutrients, for increased productivity yields with lower cost and environmental impact. Many works have focused on urea dosage by its inclusion in biodegradable films and composites or encapsulation (see Table 5). Similarly, other elemental nutrients for crop production could be encapsulated with bioplastics and introduced as filler of biocomposite materials for fertilizers-controlled release, though further investigations are needed in this respect.

4.3 Biomedical applications

Nowadays, biopolymer composites are widely employed in biomedical applications such as tissue engineering and wound healing. According to Al-Enizi et al. [457], polymer-based nanofibrous materials are used in tissue engineering

Table 5 Potential applications of biocomposites regarding their main components and processing method

Biopolymers/bioplastics	Filler/active compound	Processing technology	Suggested application	References
Packaging				
Soy protein isolate	Montmorillonite (MMT, 0.5%wt) + clove essential oil (0.5%v)	Casting solution	Bluefin tuna (Thunnus Thynnus) fillets	[409]
Chitosan and cornstarch	Lemon essential oil (1–3%wt) and grapefruit seed extract (1–3%wt)	Casting solution and heat-sealing	Blueberries conservation simulating transport and commercial conditions	[49]
Cornstarch	Green synthesized AgNPs in situ (143 ppm)	Casting solution and heat-sealing	Cheese preservation	[184]
Tapioca Starch	Grape pomace extracts (8%v/v) and cellulose nanocrystals (10%v/v)	Casting solution	Ready to eat chicken meats and evaluate their antimicrobial properties	[410]
PLA	Commercial nanoparticles: TiO_2 (3%wt); (2%wt) nano-TiO_2 + (1%wt) nano-Ag	Casting and dried in a vacuum oven at ambient temperature	Cottage cheese preservation	[411]
Curdlan + PVA	Thyme EO (1–2%wt)	Casting solution	Chilled pork meat preservation	[292]
Whey protein	Oregano and garlic EOs (2%wt)	Casting solution	Kasar cheese	[299]
Chitosan-cassava TPS bilayer films	Oregano and or cinnamon leaf EOs (0.25%wt)	Compression molding	Sliced pork meat	[297]
Gelatin/gellan gum	Red radish anthocyanins 5, 10, 15, and 20 mg/100 mL	Casting	Milk and fish quality	[412]
Zein	Laurel or rosemary leaves extracts (1–10%)	Electrospinning	Cheese slices	[413]
Starch (15–30%wt)	Stearic acid (0–0.15 M)	Temperature-assisted electrospinning	Waterproof food packaging with high hydrophobicity and self-cleaning ability	[414]
Cassava starch	Grape stalk (7%wt)	Thermoforming foaming	Low moisture food packaging	[390]
Chitosan	Dextran produced by a *Leuconostoc mesenteroides* strain (0 to 4% w/v dispersion)	Casting	Preservation of fresh mushrooms	[415]
Chitosan-carboxymethyl cellulose-oleic acid	ZnO nanoparticles (0.5, 1, and 2%)	Casting	Increase the shelf life of bread	[416]
Carboxymethyl cellulose-Guar gum	AgNPs	Casting	Improved shelf life of strawberries	[417]
Agroindustry application				
Cassava starch	Cassava bagasse + urea (25–37.5%)	Casting	Fertilizer controlled-release system	[418]
Protein hydrolysate and Poly(ethylene glycols) (PEGs)	Carbon black and wood-cellulose microfibers (18%wt)	Spraying	Opaque spray coated mulch	[419]
Protein hydrolysate and Poly(ethylene glycols) (PEGs)	Beech wood flour and saw dust (18%wt)	Melt mixing and hot pressing	Biodegradable plant nursery containers	[420]
Biomedical				
Chitosan	PHB	Casting	In-vitro drug release	Evangeline and Sridharan 2019
Soy protein isolate-agar	Ciprofloxacin hydrochloride (Cip)	Casting	As dressings with drug release and antibacterial activity	[421]

Table 5 (continued)

Biopolymers/bioplastics	Filler/active compound	Processing technology	Suggested application	References
Hyaluronan/PVA	AgNPs	Electrospinning	Wound dressing nanofibrous mats	[422]
Collagen protein	Silica and calcium phosphate bioceramics	Electrospinning	Bone tissue engineering	[423]
PVA-chitosan	Cabreuva essential oil	Electrospinning	Wound dressing	[424]
PLA	Lignin	3D printing	Tissue culture or regenerative medicine	[80]
Collagen and gelatin	Hyaluronic acid	Casting + freezing + lyophilization	Sponge bio-porous scaffolds	[425]
Chitosan-PVA	Doxazocin	Casting + freezing + lyophilization	Scaffolds	[426]
Other				
PLA	Sepiolite/Cu or Ag-sepiolite (1%wt)	Electrospinning	Antifouling membranes	[427]
Wheat gluten	Sugarcane bagasse	Compression thermomoulding	Gypsum based ceiling tiles substitution	[105]
Starch-chitosan	Carbon nanotube – pristine graphene-organic conductive poly(3,4-ethylenedioxythiophene) ((SCNT)-(PG)-(PEDOT))	Spray coating and casting	Biodegradable transparent electrode	[428]
Alginate	Cobalt ferrite nanoparticles	Disperse cross-linking with calcium chloride	Dye removal	[54]
Chitosan	Activated carbon and iron nanoparticles	Beads by alkaline solution precipitation	Cadmium removal	[429]

(bones, blood vessels, and oral tissues) and wound dressing. Synthetic biodegradable and natural polymers offer advantages over the conventional materials employed for medical devices because of their biocompatibility, biodegradability, lower antigenicity, and renewability [458]. The development of biomaterials based on biodegradable polymers has driven a new generation of materials for tissue regeneration and wound healing, which is in line with nanotechnology-based engineering strategies. In this sense, Hamdan et al. [459] stressed that numerous engineered nanotechnologies have been proposed demonstrating unique properties and multiple functions that address specific problems associated with wound repair mechanisms. The versatility of biopolymers enables to develop of diverse biomedical devices such as scaffold and wound dressings with high performance, biomimetic properties, and several other tailored characteristics which offer multiple applications. Correspondingly, Sahana and Rekha [458] assure that the technological advances in material science, regenerative medicine, nanotechnology, and bioengineering aid to improve the functional and structural characteristics of biodegradable polymers to suit the current wound care demands such as tissue repair, restoration of lost tissue integrity, and scarless healing.

Wound dressing protects the wound from microorganism deposition and dehydration and must improve the healing process by interacting with the wound through the release of bioactive molecules while maintaining the necessary favorable conditions for the re-establishment of the skin integrity and homeostasis [460]. In addition, wound dressing must ensure complete skin recovery with the best functional and cosmetic results [461]. Biopolymer-based wound healing materials can absorb tissue exudates, prevent wound dehydration and allow oxygen to permeate the wound, and can also be loaded with bioactive substances to be delivered into the wound [462]. As reported by Kalashnikova et al. [463], there are two main categories of biomaterials used in wound healing: materials that exhibit intrinsic properties beneficial for wound treatment, and materials employed as delivery vehicles for therapeutic agents. Various innovative biocomposites films and hydrogels with controlled drug or active compound release have been reported for wound dressing [47, 255, 424, 464, 465]. Collagen, cellulose, chitosan, alginate, hyaluronan, fucoidan, and carrageen are the most widely used biopolymers to develop wound dressing materials. These present either antimicrobial, anti-inflammatory, water retention, proliferative, angiogenic, or other targeted actions on specific cells, hence playing a key role in the healing process [458]. A very interesting and comprehensive review of collagen and collagen-based wound dressings published by Brett [466], remarks that wound dressings based on this biopolymer are cost-effective and present high water holding capacity, mechanical resistance, and flexibility. Collagen-based wound dressings can be obtained for diverse wound types and degrees, such as bedsores, minor burns, foot ulcers, large open cuts, chronic wounds, low to heavy exudation wounds, and surgical wounds. Likewise, Keshk [467] reviewed several industrial applications of bacterial cellulose, including wound healing. Wound dressings for burns, chronic wounds, plastic/reconstructive surgeries can be developed from this biopolymer due to its antibacterial capacity [458]. Alginate has been also widely studied in this regard, either alone or in combination with other biomaterials or biomolecules. In the review by Varaprasad et al. [462] about alginate-based composite materials for wound dressing application, the importance of alginates and the roles of their derivatives in wound dressing biomaterials, besides numerous studies on recent alginate-based wound dressing materials. Alginates and their derivatives present homeostatic character, which is relevant to treat draining wounds, pressure ulcers, dermal wounds, surgical incisions, or dehisced wounds, as well as infected and postoperative wounds. In the case of hyaluronate-based wound dressings, these are characterized for their flexibility, high biocompatibility, and bacteriostatic character [468]. This kind of biomedical devices is very useful to treat chronic wounds as well as partial and full-thickness wounds. Regarding chitosan, its antimicrobial capacity is the more relevant property to develop wound dressing for acute wounds and pressure ulcers as was reported by Dai et al. [469]. In addition to these widely studied and reported biopolymers, there are several similar biomolecules of therapeutic interest such as; fucoidan, carrageenan, and glucans that have been less explored [458].

The concept of tissue engineering aims to the self-regeneration of damaged tissues with the support of a scaffold that acts as a guide and support for new cell growth [470]. Biocompatible and biocomposite scaffolds aid the fast integration of tissues, their biocompatibility allows human cells to organize and grow around the polymer. Initially, the scaffold was merely designed to give support to the cell, but nowadays scaffolds may be loaded with biological factors to facilitate cells growth [471]. Tissue engineering also contemplates the development of artificial tissues that are physiologically functional; hence technological advances in this area continuously occur. In this regard, Gauvin et al. [472] stressed that studies about cell-seeded scaffolds have accomplished novelty materials and processing methods leading to well-engineered biocompatible systems. Another technological advance in tissue engineering is the use of nanostructured biomaterials such as nanoparticles and nanocomposites, as well as organic–inorganic hybrid polymers to develop scaffolds for organs regeneration. According to Iqbal et al. [473] and Khan et al. [474], diverse synthetic and natural polymers and their composite materials have been used to fabricate scaffolds for bone tissue engineering, nerve regeneration, controlled drug release, tooth structure regeneration, guided tissue regeneration (GTR), reinforcement of dental composite, bone and cartilage regeneration. Electrospinning, foaming, and 3D printing have been studied for biocomposites scaffolds, being PLA the most used bioplastic for these applications [80, 423, 475]. Regarding micro-fabrication technologies, Iqbal et al. [473] mentioned that lithography, bioprinting, micromolding, or photolithography are now becoming more routine and are emerging as powerful tools for the manufacture of biomaterials and tissue-engineered constructs. These authors also stressed that the use of these micro and nanotechnologies not only replicates cell-scale complexities by providing the cells with a microenvironment that mimics the native structure but also allows obtaining 3D architectures.

A few further examples of biomedical applications of biocomposite with raw materials from agri-food by-products are shown in Table 5.

4.4 Other innovative biobased composites applications

Numerous novel and groundbreaking applications have been reported for biobased composites and nanocomposites using by-products from the agri-food industry and green synthesized nanoparticles. Various applications have been proposed for copper nanoparticles from the field of electronics, catalysis, and industrial wastewater treatment [476]. Its use has also been proposed for cloth treatment since these nanoparticles have a high disinfectant capacity. In the context of the COVID-19 pandemic, this property is more than relevant and would improve the safety of both health personnel and patients, considering personal and medical hygiene clothing. In this regard, an Argentine company jointly with CONICET has developed AtomProtect®, a chinstrap made with a cloth treated with a polymeric solution containing Cu and Ag nanoparticles, which retains its sanitizing properties even after 15 washes [477].

Moreover, several recent works have been reported on the use of biobased biocomposites and nanocomposites for water treatment and pollutants removal [54, 55, 239, 427, 429, 478]. Such research is of primal importance to ensure clean water sustainability. For instance, Jayalakshmi and Jeyanthi [54] studied cobalt ferrite-alginate nanocomposite synthesized for highly polluting dye removal from watercourses. In addition, Dasari et al. [427] developed a biocide PLA-based nanocomposite membrane for drinking water purification. With a different approach, Goldhahn et al. [479] developed wood and gelatin tunable biocomposite membranes for water decontamination from various pollutants. Furthermore, the use of nanomaterials in the fabrication of superhydrophobic membranes for water desalination via membrane distillation has been deeply revised by Gontarek-Castro et al. [480] and Castro-Muñoz [481].

Over the last few decades, different biopolymers have been employed in membrane preparation for pervaporation (PV) applications that are currently used in the preparation of mixed matrix membranes (MMMs), a new-generation membrane for purification applications of great interest to chemical engineering processes. These MMS have been proved effective for ethanol recovery and dehydration [482, 483], EOs and aroma compound recovery [484, 485], azeotropic separation of organic mixtures [486], among others. In this respect, membranes based on chitosan, cellulose acetate, sodium alginate, PLA, and PVA have served as support materials for membranes for pervaporation, alone or including different types of nanoparticles such as Au, Cu, and Ag as well as zeolites and carbon nanotubes [482, 483, 487, 488]. In addition, Castro-Muñoz and coworkers have extensively reviewed the use of MMMs for PV, emphasizing that specific components transport and selectivity enhancement through the incorporation of inorganic materials into different polymeric membranes, mentioning key principles that conditioned the filler selection for a synergistic effect [482].

Furthermore, biobased foam with natural fillers have proven thermal and acoustic insulation properties which are attractive for construction and building [331, 333, 338, 391, 406, 489]. Several studies on biocomposites using by-products and wastes were revised and detailed in Section 3.2. Besides, biomass by-products and waste have been studied for construction panels and materials. For example, sustainable wood panels from wood industry by-products and ecofriendly adhesives from cassava starch were developed by Monroy et al. [490, 491] as an alternative to synthetic adhesives in medium-density fiberboard (MDF) manufacture. Likewise, Guna et al. [105] studied sugarcane bagasse-gluten composites as a potential substitute for gypsum based ceiling tiles.

Finally, biobased composites and nanocomposite materials show promising use of inflexible and biodegradable electronics and energy storage systems [492–495]. In this regard, Thiangtham et al. [496] designed biocomposite membranes based on MCC extracted from sugarcane bagasse added into PLA/PBS matrices.

5 Life cycle assessment of single use biobased composites

Within the context of circular economy, aiming to preserve the value of products, materials, and resources for as long as possible minimizing or eliminating, if possible, their environmental impact, biobased materials have become key players, especially owing to their renewable character. Yet, despite their renewability, biobased composite materials sustainability is not granted and depends on various factors, from raw materials source to produced materials end of life. In general, life cycle assessments (LCAs) of biobased plastic materials have indicated significantly lower greenhouse gases GHG emissions than the fossil oil counterparts [497]. Differences are mostly attributed to plants CO_2 absorption in photosynthesis before harvest or felling, considering that biobased materials are either composted or burned at the end of their life cycle resulting in CO_2 emission with a net-zero carbon balance [498]. Besides, biobased materials use intrinsically implies a reduction in non-renewable energy resources due to the raw material source shift. Razza et al. [358] demonstrated that despite their greater density, starch-based expanded packaging could have a 50% cut in fossil fuels use, a 60% decrease in greenhouse gas emission, a 90% reduction in volatile organic compounds (VOCs) emissions, and a 15% of landfilling, considering a current standard of 40% of organic recycling rate compared to EPS packaging.

The major LCA-reported impact of biobased plastics is the source and production of the raw materials. Food crop feedstocks are land extensive, therefore large new land areas shall be destined to crop production exclusively for bioplastics purposes. Such land-use changes imply deforestation for agricultural production, as have the conversion of rainforests to sugarcane plantations in Brazil or the Great Chaco (Argentina) deforestation for soy expansion, which comprise several direct and indirect environmental and social impacts within the raw materials production regions [499, 500]. Furthermore, land-use change is one of the primary global causes of increasing greenhouse gases emissions and soil degradation, biodiversity loss, and fresh-water scarcity [501]. In this regard, the use of agri-food waste and by-products for bioplastic and biocomposite production would decrease land-use requirements safeguarding its negative impacts. For instance, seed oil cakes, are byproducts of vegetable oil industries, are known for their high fiber, polysaccharides, and proteins content that can be extracted and that may represent a renewable source to produce innovative biobased materials [15]. In addition, current agricultural practices have a great impact on the LCA of biobased products, being sustainable practices needed to reduce eutrophication, prevent soil erosion, and protect biodiversity. Incidentally, an LCA study carried out by Vigil et al. [502] on active packaging for the fresh-cut vegetable industry, indicated that agricultural production of fresh lettuces is responsible for most impact factors, followed by packaging manufacturing. An alternative to tackle the latter, the use of fully renewably sourced electricity for packaging molding technology could reduce in over 50% the potential impact [358].

To close the loop in the plastic products industry, reusable product design and recycling systems have been established. Nonetheless, products that can be reuse is somehow limited and recycling is rarely total: mechanical recycling seldomly transforms products back into their original product system;

usually, the virgin polymer is needed for reprocessing; and a substantial volume of material is rejected and redirected to other waste management types, of which a percentage may even end up as ocean debris [503]. Biodegradable biobased plastics introduce another form of waste transformation intending to reduce plastic waste generation and increase the product's circularity by aerobic and anaerobic degradation. However, effective waste management systems are needed to prevent the incorrect disposal of bioplastic waste and its consequent accumulation in the environment as have been observed with conventional plastics use [1, 2]. Among biodegradable bioplastics, PHAs are both compostable and biodegradable in marine environments which may comparably reduce their environmental impact [4].

Lastly, the sustainability of biobased nanocomposites is also questioned in terms of the unknown hazards and toxicity [504]. Therefore, migration and cytotoxicity studies of nanofillers are currently being conducted [180, 432, 505, 506]. Furthermore, the use of hazardous chemicals in some biopolymers and biobased filler treatments must also be considered and reevaluated aiming to find greener alternatives [507].

6 Final remarks on current limitations and future trends

Over the last decade, a shift in the design dynamics of new materials has been observed, having a more specific approach born from the application requirements up. In this regard, composite materials offer a major advantage since their final properties can be tailored by selecting polymer matrix and filler type, their content ratio, and filler size distribution and morphology. Besides, the limitation of fossil resources and their consequent environmental impact has driven the search for alternative biomass-based polymeric materials and composites. Over 7000 research articles on biocomposites have been reported during the last 10 years with a growing tendency, which highlights the novelty and potential of these materials.

In addition, bio-sourced and biodegradable materials are key players towards a circular or sustainable economy. In recent years, the focus has been set on those materials with optimized end-of-life cycles considering reusability, recyclability, and biodegradability. However, structural changes and improvements are needed both in waste management systems as in production and retail allocation and distribution to achieve fully sustainable products and processes. Therefore, a tendency towards fully integrated production systems (i.e., biorefineries) in which different products from raw materials, by-products, and residues are entangled and nearby so as to reduce waste generation, transportation costs, and emissions. Nonetheless, to fully understand the role that bioplastics and biocomposites could play in a global economy, further reliable information on their sustainability is needed. Providing a basis to guide future technological developments. Research towards developing more ecologically responsible biocomposites not only needs to focus on properties optimization according to the application of the materials but also aim to minimize resource use through the selection of the process technologies and to avoid (if possible) environmental and health hazardous chemical reagents. In this regard, biomass production is crucial to the sustainability of these materials, thus the need for agricultural systems that are respectful for farmers, the environment, and the communities.

The major drawback of bioplastics and their products is that they are not economically competitive with commodity plastics. Therefore, as it was once for conventional plastic, the development of composite materials provides a lower-cost alternative to pure components as well as providing enhanced and unique properties. Furthermore, fully renewable biobased composites materials can be produced from currently devalued raw materials, regarded as low-cost by-products or wastes. This implies not only a potential drop in production cost of biobased material, but also a reduction in energy, water, and soil used in raw materials production and a further reduction in waste generation.

The numerous and innovative possible applications for biobased composites and nanocomposites that have been reviewed and reported demonstrate the potentiality of these materials. The nontoxic nature and biodegradability of these materials are key features for their use in active and intelligent food packaging that can be composted and functionalized biodegradable mulching plastic and fertilizers-controlled release aiming for more environmentally friendly agricultural systems. Their water affinity and retention capacity are relevant for absorption systems for pollutants retention and membrane development for water purification treatments. Furthermore, these properties along with the biocompatibility have triggered the development of wound dressing, drug-delivery systems, and scaffolds for tissue engineering. Several other investigations have been done in insulation construction materials and flexible electronics and energy storage systems. The results shown are very promising, thus future research should examine strategically the scaled-up production and process optimization of these ecological materials to meet the market demands.

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Data availability Data sharing is not applicable – no new data generated.

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Declarations

Conflict of interest The authors declare no competing interests.

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