


# Current status of biobased and biodegradable food packaging materials: Impact on food quality and effect of innovative processing technologies

Julie Nilsen-Nygaard  | Estefanía Noriega Fernández | Tanja Radusin |  
 Bjørn Tore Rotabakk | Jawad Sarfraz | Nusrat Sharmin | Morten Sivertsvik |  
 Izumi Sone | Marit Kvalvåg Pettersen

Food Division, Norwegian Institute of Food, Fisheries and Aquaculture (Nofima AS), Tromsø, Norway

## Correspondence

Julie Nilsen-Nygaard, Food Division,  
 Norwegian Institute of Food, Fisheries and  
 Aquaculture (Nofima AS), Osloveien 1,  
 1433 Tromsø, Norway.  
 Email: [Julie.nilsen-nygaard@nofima.no](mailto:Julie.nilsen-nygaard@nofima.no)

## Abstract

Fossil-based plastic materials are an integral part of modern life. In food packaging, plastics have a highly important function in preserving food quality and safety, ensuring adequate shelf life, and thereby contributing to limiting food waste. Meanwhile, the global stream of plastics into the oceans is increasing exponentially, triggering worldwide concerns for the environment. There is an urgent need to reduce the environmental impacts of packaging waste, a matter raising increasing consumer awareness. Shifting part of the focus toward packaging materials from renewable resources is one promising strategy. This review provides an overview of the status and future of biobased and biodegradable films used for food packaging applications, highlighting the effects on food shelf life and quality. Potentials, limitations, and promising modifications of selected synthetic biopolymers; polylactic acid, polybutylene succinate, and polyhydroxyalkanoate; and natural biopolymers such as cellulose, starch, chitosan, alginate, gelatine, whey, and soy protein are discussed. Further, this review provides insight into the connection between biobased packaging materials and innovative technologies such as high pressure, cold plasma, microwave, ultrasound, and ultraviolet light. The potential for utilizing such technologies to improve

**Nomenclature:** BOPP, Biaxially oriented polypropylene; CA, Citric acid; CAR, Carvacrol; CFU, Colony forming units; CMC, Carboxymethyl cellulose; CP, Cold plasma; DSM, Defatted soybean meal; EO, Essential oil; EVOH, Ethylene vinyl alcohol;  $F_A$ , Fraction of acetylation; FDA, U.S. Food and Drug Administration; HA, Hydroxyalkanoic acid; HPP, High-pressure processing; LDPE, Low-density polyethylene; MATS, Microwave-assisted thermal sterilization; *mcl*, Medium chain length; MgO, Magnesium oxide; MMT, Montmorillonite; MP, Microencapsulated powder; MW, Microwave; OTR, Oxygen transmission rate; PA, Polyamide; PATP, Pressure-assisted thermal pasteurization; PATS, Pressure-assisted thermal sterilization; PBS, Polybutylene succinate; PBSA, Polybutylene succinate adipate; PCL, Polycaprolactone; PE, Polyethylene; PET, Polyethylene terephthalate; PHA, Polyhydroxyalkanoate; PHB, Polyhydroxybutyrate; PHBV, Poly(3-hydroxybutyrate-co-3-hydroxyvalerate); PLA, Polylactic acid; PNC, Polymer nanocomposites; PP, Polypropylene; PS, Polystyrene; PVA, Polyvinyl alcohol; PVC, Polyvinyl chloride; RONS, Reactive oxygen and nitrogen species; SA, Sodium alginate; *scl*, Small chain length; SiO<sub>2</sub>, Silicon oxide; TiO<sub>2</sub>, Titanium oxide; TPS, Thermoplastic starch; TVC, Total viable count; US, Ultrasound; WPI, Whey protein isolate; WVP, Water vapor permeability; ZnO, Zinc oxide;  $\beta$ -CD,  $\beta$ -Cyclodextrin

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biomaterial barrier and mechanical properties as well as to aid in improving overall shelf life for the packaging system by in-pack processing is elaborated on.

#### KEYWORDS

Biobased and biodegradable materials, Food packaging, Processing technologies, Food quality, Shelf-life

## 1 | INTRODUCTION

Food packaging protects and preserves the quality of the food product, thus contributing to more sustainable food value chains and limiting food waste (Lindh, Williams, Olsson, & Wikström, 2016; Svanes et al., 2010; United Nations, 2015). Polymers, that is, plastic materials, have been widely used as food packaging materials for many decades. Traditional polymers are produced from fossil-based resources and the extensive use of plastics in food packaging today is related to their superior properties, technological innovations, and relatively low cost (Cha & Chinnan, 2004). Conventional plastic materials rely on nonrenewable resources, are nonbiodegradable, and in many cases not fully recyclable. The massive consumption of such materials thereby contributes to environmental challenges such as depleting natural resources, littering, and global warming (Mangaraj, Yadav, Bal, Dash, & Mahanti, 2019; Schmidt Rivera, Leadley, Potter, & Azapagic, 2019; Wohner, Pauer, Heinrich, & Tacker, 2019). Increased public awareness of the environmental challenges related to conventional plastic materials and consumer pressure for improved sustainability has triggered the development of biobased, biodegradable food packaging materials. Further, introduction and implementation of the Single-Use Plastic Directive, expected to be issued by the European Commission in 2020, may accelerate implementation of alternatives to traditional plastic materials.

According to the European Bioplastic Organization (European Bioplastics, 2020), a plastic material is defined as a bioplastic if it is either biobased (meaning the material is [partly] derived from biomass as renewable resources), biodegradable, or possesses both properties. Biopolymers for food packaging are materials originating from agricultural and marine sources and can be divided in three categories; polymeric materials produced by chemical synthesis from bioderived monomers; polymeric materials produced by microorganisms; and natural biopolymers extracted directly from raw materials (Cha & Chinnan, 2004). A schematic of the classification of biobased and biodegradable polymers is given in Figure 1 with examples of polymers in the different categories.

Biodegradation is a polymer degradation process assisted by microorganisms in which the material disin-

tegrates to water, carbon dioxide, biomass, and methane (Guzman, Gnutek, & Janik, 2011; Meereboer, Misra, & Mohanty, 2020). The biodegradation process depends on the surrounding environmental factors such as temperature, humidity, and presence of oxygen (aerobic or anaerobic conditions) (European Bioplastics, 2020; Guzman et al., 2011). Specific environment such as industrial composting plant, garden compost and soil, and the material itself (e.g., composition, crystallinity, chemical structure, and hydrophilicity) also have an impact, and thus variation in both the process and outcome can occur (Meereboer et al., 2020). Composting, also called organic recycling, is an enhanced biodegradation under specific conditions such as timeframe and temperature and in the presence of microorganisms. Composting implies that the material, in addition to biodegrading, also becomes a part of usable compost and add nutrients to the soil (European Commission, 2018). To be certified and labeled as compostable, specific standards must be fulfilled (e.g., EN 13432, refer to Section 8). Fully organic packaging materials can be regarded as biodegradable and compostable, whereas use of, for example, co-monomers, additives, and cross linkers altering the material network structure may reduce or enhance the biodegradability of a material (Rasal, Janorkar, & Hirt, 2010).

Biobased and biodegradable materials typically have relatively poor water vapor barrier properties and mechanical properties, heat stability, and processing properties compared to their fossil-based counterparts (Platt, 2006). Thus, the challenges to achieve suitable barrier and mechanical properties without compromising the biodegradability limit their widespread acceptance and use (Nampoothiri, Nair, & John, 2010). Use of commercially available biopolymer films is limited to products with relatively short shelf life or perishable products, for example, fruit and vegetables requiring respiration and humidity (Auras, Harte, & Selke, 2004; Platt, 2006) in addition to long shelf life products such as dry pasta (Jabeen, Majid, & Nayik, 2015; Peelman et al., 2013). Mc Millin (2017) stated conventional plastics are not likely to be replaced by biopolymers due to their less suitable properties in meat packaging. However, Nampoothiri et al. (2010) conclude that techniques such as polymer modification, coating, blending, and use of

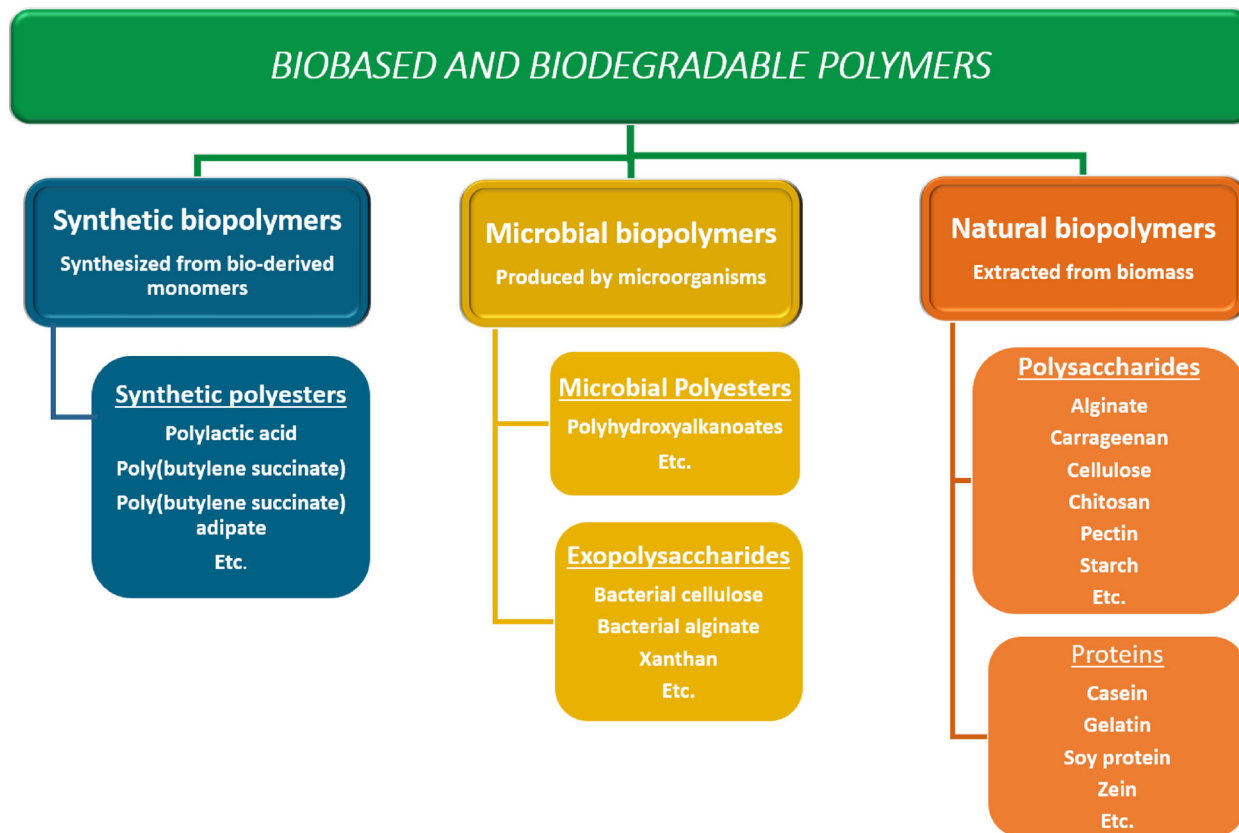


FIGURE 1 Classification of biobased and biodegradable polymers on the basis of their origin (Cha & Chinnan, 2004; Cutter, 2006; Mangaraj et al., 2019)

nanocomposites are likely to contribute to solve these challenges.

Food processing technologies are an indispensable aid in preserving food products by prolonging shelf life and ensuring food safety, besides contributing to better resource utilization and a more stable food supply, which are important factors in reducing food loss and food waste. Nonthermal processing and advanced volumetric heating (e.g., high-pressure processing [HPP], microwave [MW], ultrasound [US], pulsed electric field, UV light [UV], cold plasma [CP], etc.) have received significant attention in the last decade in response to the increasing consumer demand for safe, minimally processed, and value-added products (e.g., fresh-like, healthy, long shelf life) (European Commission, 2013), because traditional thermal processing, extensively used in the food industry, accounts for a relatively high environmental footprint (high energy usage) and undesirable effects on food nutritional (e.g., vitamin loss) and sensory (e.g., texture, color, and taste) attributes (Pardo & Zufia, 2011). Besides nonthermal pasteurization, a palette of commercially sound applications can benefit from such cutting-edge technologies, for example, disinfection of food-contact surfaces; process optimization (e.g., drying and freezing); extraction of intra-

cellular compounds; mitigation of food allergenicity; food waste valorization; food/package functionalization, and so on (Jermann, Koutchma, Margas, Leadley, & Ros-Polski, 2015; Pinela & Ferreira, 2017). The European Commission business innovation observatory has acknowledged their timely and substantial contribution to manufacturing efficiency (enhanced productivity, sustainable resource management, and better self-sufficiency; streamlined supply chain logistics; reduced labor costs), food safety and security (improved public health, reduced prevalence of diet-related diseases, food recalls, and associated costs), and green-shift (reduced food losses/waste and carbon footprint; energy and water savings), while creating market value through new cost-effective niche opportunities (Augustin et al., 2016; European Commission, 2013, 2015; Novel Q, 2011; Pardo & Zufia, 2011).

Overall, most innovative technologies (e.g., HPP, MW, US, UV, and CP) support the so-called “in-pack” processing, which prevents undesirable downstream recontamination and packaging presterilization (e.g., by irradiation or application of hydrogen peroxide), while enabling new package types. Wherever “in-pack” solutions are not feasible, application of, for example, UV or CP on the package itself can avoid intensive sterilization of the food

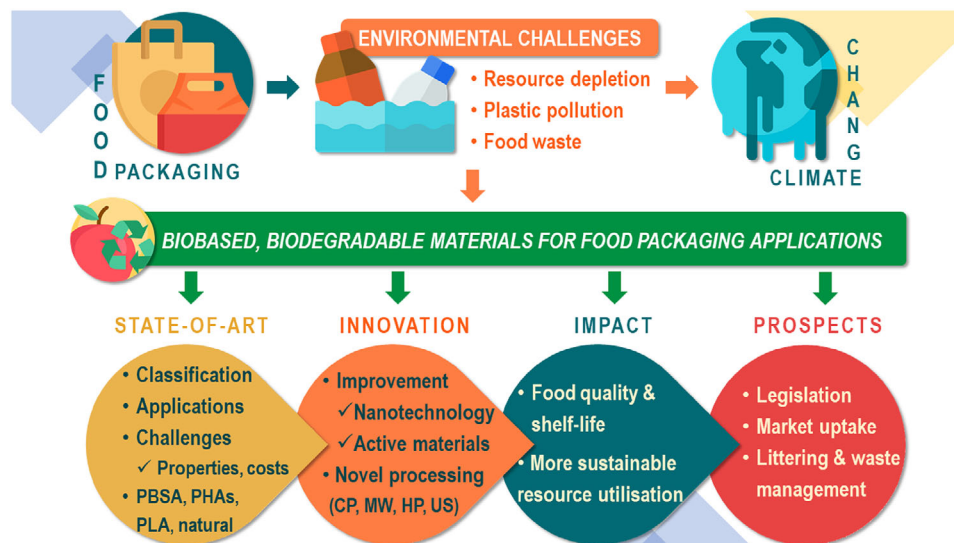


FIGURE 2 Schematic outline of the review paper

or the packaging system (European Commission, 2013, 2015; Pereira & Vicente, 2010). However, in both scenarios, the exposed material may alter its structure and eventually its mechanical and barrier properties. Furthermore, selected technologies are typically used toward improved functionalization, coating and tuning properties of packaging materials, and in the design of nanocomposites, an aspect particularly interesting in the context of biodegradable and biobased materials, where their shortcomings in this respect, as compared to conventional plastics, very often hinder market implementation. Although few available publications address the impact of such enabling technologies when combined with biobased, active, or intelligent packaging systems, particular attention is expected in the near future with regard to safety assessment (e.g., non-intentionally added substances), evolution of mechanical and barrier properties, and food–packaging interactions. The co-utilization of biomaterials and innovative processing technologies for food packaging may aid in overcoming the well-known challenges related to industrial implementation of biomaterials.

The aim of this review is to provide an updated overview of the status of biobased, biodegradable packaging materials intended for food packaging applications (Figure 2). The review is limited to studies focusing on self-supporting films of biobased, biodegradable materials for use as packaging material, that is, edible films and paper-based packaging are omitted. The effect on food quality and shelf life in biomaterial packaging systems is highlighted. Further, potential of innovative technologies toward enhanced manufacture and functionality of biobased packaging materials is assessed, as well as the impact of such technologies on biomaterial performance and subsequent interactions with food counterparts during

in-pack processing. The review mainly encompasses studies published in the last 5 years.

## 2 | SYNTHETIC BIOBASED, BIODEGRADABLE POLYMERS FOR FOOD PACKAGING

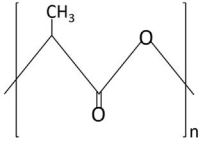
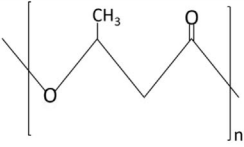
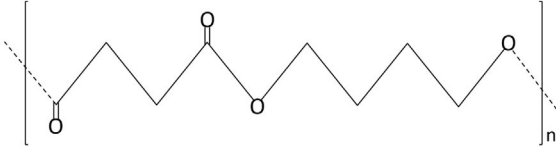
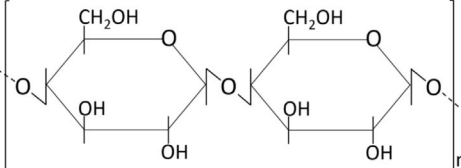
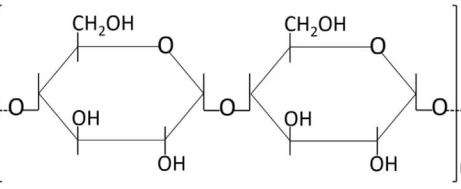
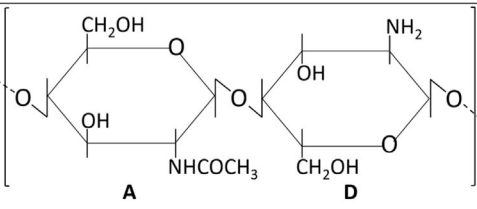
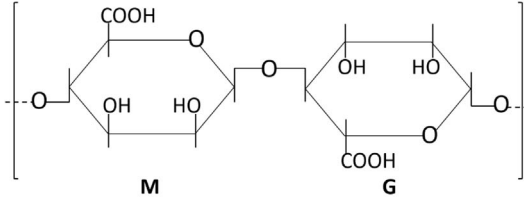
### 2.1 | Polylactic acid

Poly(lactic acid) (PLA) or poly(lactide) is one of the most promising biobased polymers due to its availability, compostability, biocompatibility, and properties close to conventional fossil-based polymers. PLA is degradable (biodegradable), but due to high melting point and glass transition temperature it requires industrial composting at 55 to 60 °C (Meereboer et al., 2020; Urbanek et al., 2020).

#### 2.1.1 | Structure and properties of PLA

PLA is a partially crystalline thermoplastic polyester (Table 1). It can be obtained by fermentation (from wheat corn, rice, and sugar beets) or by chemical synthesis. The chemical reaction of lactide (cyclic lactic acid diester) formation is an intermediate step in the synthesis of PLA, which in its chain can have two different optically stereoisomeric forms: L (–) - lactide (S, S); D (+) - lactide (R, R) and optically inactive meso-lactide. By chemical synthesis, PLA is obtained by polycondensation or ring-opening polymerization reactions to produce high-molecular-weight polymers (Murariu & Dubois, 2016). The racemic mixture of L- and D-lactide is called D–L lactide, and L- and D–L lactide are used to produce

**TABLE 1** Selected biobased, biodegradable polymers with monomers and chemical structures

	Monomer(s)	Chemical structure
<b>Synthetic biopolymers</b>		
<b>Poly(lactic acid) (PLA)</b>	Lactic acid, lactide	
<b>Poly(hydroxyalkanoate) (PHA)</b>	Hydroxyalkanoic acids	
<b>Poly(butylene succinate) (PBS)</b>	Succinic acid and 1,4-butanediol	
<b>Natural biopolymers</b>		
<b>Cellulose</b>	D-Glucose $\beta$ (1 $\rightarrow$ 4) glycosidic bonds	
<b>Starch</b>	D-Glucose $\alpha$ (1 $\rightarrow$ 4) and $\alpha$ (1 $\rightarrow$ 6) glycosidic bonds	
<b>Chitosan</b>	2-acetamide-2-deoxy-D-glucose and 2-amino-2-deoxy-D-glucose  $\beta$ (1 $\rightarrow$ 4) glycosidic bonds	
<b>Alginate</b>	$\alpha$ -mannuronic acid (M) and $\beta$ -guluronic acid (G)  (1 $\rightarrow$ 4) glycosidic linkages	

Note. The structure of chitosan displays a partially deacetylated chitosan.

polymers. PLA properties vary depending on the relationship and distribution of the two stereoisomers or comonomers (Auras, Lim, Selke, & Tsuji, 2011). The optical purity of PLA influences the ultimate properties of polymers, such as structure, thermal, barrier, and mechanical properties. Poly-L-lactic acid with over 93% of L-lactide is a partially crystalline polymer, whereas a smaller proportion of L-lactide gives amorphous polymers, so by changing this ratio, materials with different properties can be obtained (Auras et al., 2011). Further, crystallinity of PLA can be improved by chemical and physical modifications. Usually chemical modifications include incorporation of small molecules in PLA polymer structure (manipulation on molecular level), whereas physical modification can include addition of, for example, nanoparticles that are going to act as nucleating agents and expand crystalline regions in the polymer matrix (Pilić et al., 2015; Ristić, Radusin, Pilić, Cakić, & Budinski-Simendić, 2013).

In comparison to conventional polymers, PLA cannot meet all the requirements in the field of food packaging. The values of the mechanical properties of PLA are approximate to those of PS (polystyrene) but lower in spinning with PET (polyethylene terephthalate). One of the disadvantages of PLA as a food packaging material is its poorer barrier properties (on water vapor, oxygen, and other gases). Most authors have concluded even small changes in the ratio of L- and D-enantiomers affect the barrier properties of PLA, primarily due to the proportion of crystalline regions in the polymer matrix (Farah, Anderson, & Langer, 2016). In addition to external factors, the barrier properties depend on the degree of crystallinity, that is, in the case of a more crystalline polymer, the path of gas passage through the film is longer and thus the permeability is lower. PET and PLA are hydrophobic polymers, and they absorb minimal amounts of water, which is why they are often compared and have similar barrier properties (Auras et al., 2011). Some of the average physical properties of PLA are listed in Table 2.

### 2.1.2 | PLA film preparation

PLA has superior processing properties compared to other biopolymers and can be processed by almost all known processing techniques— injection molding, film extrusion, blow molding, thermoforming, and so on (Castro-Aguirre, Iniguez-Franco, Samsudin, Fang, & Auras, 2016; Murariu & Dubois, 2016). However, PLA processing is still a great challenge as during thermal processing in polymer processing processes, small variations in temperature lead to a very rapid loss of thermal stability of the polymer itself. In addition, the PLA processed by extrusion and injection molding reduces the molecular weight, which certainly

affects the properties of the offset product (Auras et al., 2011; Castro-Aguirre et al., 2016). The addition of plasticizers can improve the properties of PLA during processing, and typically uses low-molecular-weight plasticizers based on lactic and glycolic acids (Rasal et al., 2010). The PLA polymer begins to decompose at 300 °C and completely decomposes at 400 °C. Compared to conventional polymers, PLA has similar thermal stability as polyvinyl chloride (PVC), but significantly lower compared to PS, PP (polypropylene), PE (polyethylene), and PET (Lim, Auras, & Rubino, 2008).

### 2.1.3 | Applications and commercial availability of PLA

The PLA market has witnessed increased demand over past years for various packaging applications (dry products and perishable products such as fruits and vegetables), resulting in increased production in Europe, the United States, and Japan. The global PLA market was valued at 700 million USD in 2019 with estimated increase by 2025 up to 2,500 million USD (Clark & Singh, 2013; Grand View Research, 2019; Markets and Research, 2020). Key PLA resin companies in the industry are Futerro (Belgium), NatureWorks LLC (Minnesota, USA), BASF SE (Germany), Total Corbion (Netherlands), Hitachi Ltd. (Japan), Sulzer Ltd. (Switzerland), Zhejiang Hisun Biomaterials Co., Ltd. (China), and Thyssenkrupp AG (Germany) (Grand View Research, 2019).

Benefits of using PLA for food packaging application are reflected in the fact that it is compostable under industrial conditions, produced from renewable sources, biocompatible, recyclable, and has potential to substitute conventional plastic materials (Farah et al., 2016; Mangaraj et al., 2019). It has also been approved as safe by the U.S. Food and Agriculture Agency (U.S. Food and Drug Administration [FDA]). However, due to inferior barrier and mechanical properties, the application of PLA for food packaging is currently limited. It is possible to design and balance the properties of PLA by changing its chemical composition and varying its molecular characteristics. Further, combining PLA with other compounds allows for fine-tuning properties to meet the requirements of different food products. Please refer to Section 4 on biomaterial modifications and the review by Rasal et al. (2010) for detail on potential modifications of PLA. The prospects of PLA are that this polymer becomes economical in production over time and more extended use of PLA for different food packaging applications is expected. For detailed information on food packaging applications of PLA in recent literature, focusing on the effect on product shelf life, we refer the reader to Section 5.

TABLE 2 Comparison of selected thermal, mechanical, and barrier properties for some conventional and biodegradable polymers/films

Property (units)	Conventional, fossil-based				Biodegradable, synthetic				Biodegradable, natural		
	LDPE	PP	PET	EVOH 44% ethylene	PLA	PHA	PBS	PBSA	Starch (Cas-sava)17% glycerol (w/w)	Gelatin (Tilapia)25% glycerol (w/w)	Cellulose acetate (DS 1.48) 20% glycerol (w/w)
Glass transition temperature, $T_g$ (°C)	-120 <sup>a</sup>	-10 <sup>a</sup>	73 to 80 <sup>a</sup>	55 <sup>a</sup>	45-60 <sup>c</sup>	-50 to 4 <sup>e</sup>	-18 <sup>h</sup>	-32 <sup>h</sup>	36 <sup>k</sup>	-	-
Melting temperature, $T_m$ (°C)	105 to 115 <sup>a</sup>	160 to 175 <sup>a</sup>	245 to 265 <sup>a</sup>	164 <sup>a</sup>	150 to 162 <sup>c</sup>	160 to 175 <sup>f</sup>	112 <sup>h</sup>	96 <sup>h</sup>	-	-	-
Young's modulus, $E$ (MPa)	200 to 500 <sup>a</sup>	1,100 to 1,500 <sup>a</sup>	-	2,100 <sup>a</sup>	1,280 <sup>c</sup>	1,200 <sup>f</sup>	48 <sup>m</sup>	110 <sup>i</sup>	-	22 <sup>l</sup>	7 <sup>j</sup>
Tensile strength, $\sigma$ (MPa)	8 to 31 <sup>a</sup>	31 to 43 <sup>a</sup>	220 to 270 <sup>a</sup>	59 <sup>a</sup>	44 to 59 <sup>c</sup>	15 to 40 <sup>f</sup>	34 <sup>h</sup>	19 <sup>h</sup>	2.4 <sup>k</sup>	9 <sup>l</sup>	57 <sup>j</sup>
Elongation at break, $\epsilon$ (%)	100 to 965 <sup>a</sup>	500 to 650 <sup>a</sup>	70 to 110 <sup>a</sup>	380 <sup>a</sup>	4 to 7 <sup>c</sup>	1 to 15 <sup>f</sup>	560 <sup>h</sup>	807 <sup>h</sup>	213 <sup>k</sup>	45 <sup>l</sup>	8 <sup>j</sup>
Water vapor transmission rate (g-mm/m <sup>2</sup> -day)	0.375 to 0.500 <sup>a</sup>	0.1 to 0.3 <sup>a</sup>	0.390 to 0.510 <sup>a</sup>	0.724 <sup>a</sup>	2.05 to 4.30 <sup>b,*</sup>	2.36 <sup>f</sup>	1.69 <sup>g,*</sup>	-	405 <sup>k</sup>	-	257.8 <sup>l</sup>
Oxygen permeability (cm <sup>3</sup> -mm/m <sup>2</sup> -day-atm) 23 to 30 °C, 1 atm	160 to 210 <sup>a</sup>	50 to 94 <sup>a</sup>	1.2 to 2.4 <sup>a</sup>	0.0013 <sup>a</sup>	216 <sup>d</sup>	55.12 <sup>f</sup>	41.2 <sup>k,*</sup>	45.3 <sup>k,*</sup>	-	-	-

Note. Test conditions for WVTR were reported to be 37.8 °C, 90% RH, and 1 atm for the conventional, fossil-based polymers and 23 to 25 °C, 50% to 75% RH, and 1 atm for the biobased polymers.

\*The values of water vapor transmission rate (WVTR) and oxygen permeability (OP) have been converted to a standardized material thickness of 1 mm. Certain values have been converted to OP from oxygen transmission rate (OTR) based on information about material thickness and assuming a linear correlation between thickness and OTR. Test conditions for WVTR were reported to be 37.8 °C, 90% RH, and 1 atm for the conventional, fossil-based polymers and 23 to 25 °C, 50% to 75% RH, and 1 atm for the biobased polymers.

References: (a) Mangaraj, Goswami, & Mahajan, 2009; (b) Jamshidian, Tehrani, Imran, Jacquot, & Desobry, 2010; (c) Farah et al., 2016; (d) Hamad, Kaseem, Yang, Deri, & Ko, 2015; (e) Raza et al., 2018; (f) Bugnicourt et al., 2014; (g) Vytejková et al., 2017; (h) Xu & Guo, 2010; (i) Zhang & Zhang, 2015; (j) Gonçalves et al., 2019; (k) Souza et al., 2012; (l) Syahida et al., 2020.

## 2.2 | Polyhydroxyalkanoates

Polyhydroxyalkanoates (PHAs) are biobased polyesters obtained through bacterial fermentation (Samui & Kanai, 2019). These biogenic polyesters can be obtained with pure microbial cultures grown on different renewable sources such as glucose under sterile conditions. However, PHAs can also be produced from substances contained in waste water (e.g., organic acids and sugars) from different industrial processes (Colombo et al., 2019; Mannina, Presti, Montiel-Jarillo, & Suárez-Ojeda, 2019). PHAs are gaining much attention as a potential replacement for fossil-based plastics not only due to the similarity between their physicochemical properties with conventional plastics but also due to the biodegradability of PHAs in different environments (Chan et al., 2019; Koller, 2014). For in-depth information on the biodegradability of PHA, we refer the reader to the recent review paper by Meereboer et al. (2020).

### 2.2.1 | Structure and properties of PHA

PHAs are linear thermoplastic polymers that can be produced by many microorganisms as intracellular carbon and energy stocks. Structurally PHAs are thermoplastic polyesters of hydroxyalkanoic acids (HA) connected by an ester bond (Table 1) (Akaraonye, Keshavarz, & Roy, 2010). The arrangement of the monomers within the polymer and the polymer chain length depends on the microorganism, carbon source, and growth conditions used. PHAs are generally classified in two main categories: small chain length (*scl*)-PHAs with monomer units containing three to five carbon atoms and medium chain length (*mcl*)-PHAs with monomer units containing six to 14 carbon atoms (Zheng, Chen, Ma, & Chen, 2020).

PHAs are among the most versatile groups of biopolyesters with over 150 different types of monomer available (Koller, 2014). However, only few PHAs are produced on an industrial scale and available commercially such as polyhydroxybutyrate (PHB), poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV), and poly(3-hydroxybutyrate-co-3-hydroxyhexanoate) (Zheng et al., 2020). PHBV is suitable for heat shaping and producing flexible plastic bag used in the food packaging sector (Zhao, Ji, Kurt, Cornish, & Vodovotz, 2019). PHAs are mostly used as flexible food packaging with high oil content such as marinated olives, cheese, and nuts (Innocentini-Mei, Bartoli, & Baltieri, 2003).

PHA materials can range from highly brittle and crystalline thermoplastic to more elastomeric (Koller, 2014). The *scl*-PHA co-polyesters possesses high plasticity and accessibility to melt extrusion, injection molding, thermoforming etc. and as such can be considered as a suitable

candidate for food packaging applications. On the other hand, as compared to *scl*-PHA, *mcl*-PHA shows latex- to resin-like properties with extremely low glass transition temperature, making them an interesting option to be used as packaging material for storing food at freezing condition (Koller, 2014).

PHA polymers could have different properties depending on their chemical composition such as homo or copolyester, presence of contained hydroxyl fatty acids, and so on. In general, PHAs are water insoluble, relatively resistant to hydrolytic degradation, show good ultraviolet resistance, but poor resistance to acids and bases, and are soluble in chloroform and other chlorinated and nontoxic hydrocarbons (Raza, Abid, & Banat, 2018). The hydrophobic character and board flexibility in mechanical features PHAs make them promising materials to compete with fossil-based plastics in the food packaging sector. Average properties of PHAs are listed in Table 2.

PHAs could be produced with properties close to traditional food packaging materials such as PE, PP, or PET. Moreover, due to the hydrophobicity of these water-insoluble polyesters, PHA films display very high water vapor barrier properties, close to low-density polyethylene (LDPE). In addition, PHAs show high barrier properties for oxygen, water, and CO<sub>2</sub>, making them suitable for producing bottles for liquid food and CO<sub>2</sub> containing liquids.

### 2.2.2 | PHA film preparation

PHAs can be processed using different techniques depending on the molecular weight of the polymer and the co-monomer content, including injection molding and blow molding (Bugnicourt, Cinelli, Lazzeri, & Alvarez, 2014). Moreover, PHAs can be processed in different ways to be used for different applications such as packaging, molded products, paper coatings, adhesive, films, and so on (Vahabi, Rohani Rad, Parpaite, Langlois, & Saeb, 2019). Therefore, the processability, biodegradability, and naturalness of PHAs have made these biogenic polyesters a desirable candidate for different food packaging applications.

### 2.2.3 | Applications and commercial availability of PHA

One of the commercially available PHA products focused on food packaging applications are Biopol™ produced by Metabolix Inc. (USA) (Bajpai, 2019). Biopol™ possesses excellent film-forming and coating properties and is mainly used to produce disposable food containers and utensils. Telles, a joint venture between Metabolix, Inc.



and Archer Daniels Midland Co., has produced Mirel™ bioplastics, which is a type of PHA (Color, 2009). Mirel™ can be processed into high-melt-strength grade bioplastics suitable for low heat sag in thermoforming. They are suitable for a wide range packaging applications, including hot and cold cups, cup lids, yogurt containers, tubs, trays, and single-serve food packaging.

## 2.3 | Polybutylene succinate and polybutylene succinate adipate

Polybutylene succinate (PBS) and its copolymers are a family of commercially available aliphatic polyesters with good biodegradability and compostability, wide window for thermoplastic processing, balanced mechanical properties, and good thermal and chemical resistance (Chen & Yan, 2019).

### 2.3.1 | Structure and properties of PBS and PBSA

The structure of PBS is composed of succinic acid and 1,4-butanediol monomers and is normally produced via polycondensation of these two (Table 1). Traditionally, the monomers used in PBS production are extracted from fossil feedstock. However, the recent advances have also enabled the production of these monomers from renewable biomass such as starch, glucose, xylose, and so forth on an industrial scale (Chen & Yan, 2019).

The physical properties of PBS could be varied over a very wide range via copolymerization with different types and contents of monomers (Xu & Guo, 2010). The copolymerization of these aliphatic polyesters is a powerful and efficient approach to tailor the material's physicochemical, mechanical, and gas barrier properties to fit specific applications such as in food packaging (Siracusa, Lotti, Munari, & Dalla Rosa, 2015). Some of the physical properties of PBS and PBSA are presented in Table 2.

PBS is a semicrystalline polymer with high crystallization ability ( $\chi_c = 35\%$  to  $45\%$ ) and a melting temperature that is one of the highest among poly(alkylene dicarboxylate)s (Gigli et al., 2016). It has been reported that during the contact with food simulant, a change between crystalline and amorphous ratios of PBS and PBSA takes place that eventually affects the permeability behavior of these polymers (Siracusa et al., 2015). However, due to the dependency of the sorption (thermodynamic parameter) and diffusivity (kinetic parameter) processes of these polymer on different factors such as polymer segments and intersegmental packing, environment, temperature, and so on, there is no linear relationship between crys-

tallinity and permeability (George & Thomas, 2001; Siracusa et al., 2015). Another property that makes these polyesters a good candidate for food packaging application is that these materials are relatively resistant to degradation when exposed to e.g. heat and light (Siracusa et al., 2015).

### 2.3.2 | PBS and PBSA film preparation

The melt processability and mechanical properties of PBS and PBSA are closely comparable to those of widely used fossil-based plastics in food packaging sector, such as PE and PP (Vytejková et al., 2017). Moreover, the excellent melt processability of PBSs makes them very suitable for extrusion, injection molding, thermoforming, and film blowing. For PBS, the thermal stability of the polymer vastly depends on the water content, the presence of residual carboxylic acid terminals, and the molecular structure of the polymeric chain (Chen & Yan, 2019). Extrusion and injection molding process can be used to process PBS with molecular weight less than 100,000. However, for film blowing and casting high melt strength is necessary. Therefore, only PBS with higher molecular weight or long-chain structure can be used for film blowing and casting to ensure smooth processing (Xu & Guo, 2010). Long-chain branches intertwining brings high melt tension and enables the production of stretch blown bottles (Barletta & Cicci, 2019).

### 2.3.3 | Applications and commercial availability of PBS and PBSA

The mechanical properties and ease of processability have made PBSs a very potential candidate for different food packaging application, such as films and semi-rigid bowls (Vytejková et al., 2017). A copolymer of PBS and polybutylene adipate, polybutylene succinate adipate (PBSA), has also been identified as a suitable material for food packaging applications due to the high crystallinity and good thermal properties of the homopolymers (Puchalski et al., 2018).

PBS has a global capacity of over 140,000 t/a, which was 12.8% of global capacity of biobased degradable plastics reported in 2014 (Hu et al., 2015, 2019). Commercially available PBS and PBSA products include GSPLA® (Mitsubishi Chemicals), Bionolle™ (Showa Denko K.K.), Skygreen (SK Chemicals), and BIOPBS™ (PTT MCC Biochem).

It could be concluded that PBS exhibits excellent thermoplastic processability with high crystallinity and glass transition temperature below room temperature. Moreover, the mechanical properties of these polyesters are

desirable with 300% to 500% of elongation at break and are recognized as one of the most promising biodegradable plastics for different commercial application including food packaging. However, PBS has low impact strength and tear resistance, which might limit its application (Hu et al., 2019; Xu & Guo, 2010). Moreover, a considerable decrease in the shear viscosity of PBS takes place when the temperature is increased above 200 °C, which is not desirable in high-temperature processing and shaping. It is also important to mention that the presence of more than 0.1% water content in PBS can initiate hydrolysis reaction during thermal processing (Xu & Guo, 2010).

### 3 | NATURAL BIOBASED, BIODEGRADABLE POLYMERS FOR FOOD PACKAGING

In this section, the structure, properties, possibilities, and limitations of some of the most relevant and promising natural biopolymers for food packaging films are described, exemplified by the polysaccharides cellulose, starch, alginate, and chitosan as well as protein films.

#### 3.1 | Structure and properties of natural biopolymers

Polysaccharides, such as cellulose, starch, chitosan, alginate, agar, guar gum, carrageenan, and pectin, are abundant in nature as structural polymers. In general, they are composed of linear or branched chains of one or more types of alternating monosaccharides, for example, glucose, fructose, mannose, and galactose (Smidsrød & Moe, 1995). Many polysaccharides have good film-forming properties and in addition their high accessibility, low cost, biodegradability, and compostability make them desirable candidates for use in more sustainable food packaging systems (Cazón, Velazquez, Ramírez, & Vázquez, 2017).

Cellulose holds the title of being the most abundant renewable polymer in nature and can be isolated from wood, cotton, hemp, and plant-based materials as well as synthesized by microorganisms. Structurally cellulose is a linear glucan of  $\beta$  (1  $\rightarrow$  4)-linked glucose units (Table 1) (Cazón et al., 2017). Cellulose is organized in crystalline fibers and is insoluble in most solvents and is therefore not a filmogenic polymer without modification or functionalization (Cazón et al., 2017). In parallel to the current renaissance of paper-based materials for food packaging, research is focused on utilizing the vast cellulose resources for film preparation. For instance, films of the cellulose derivatives hydroxypropyl cellulose and methylcellulose have been shown to have good gas barrier ( $O_2$

and  $CO_2$ ) and good fat resistance, though poor water resistance (Villalobos, Chanona, Hernández, Gutiérrez, & Chiralt, 2005). Cellulose acetate is obtained by acetylation of cellulose. The polymer forms relatively clear films and is currently applied for dry foods and fresh produce, though its application areas are restricted due to moisture sensitivity (Cazón et al., 2017).

Starches are glucans composed of linear chains (amylose) and branched chains (amylopectin) of D-glucose units coupled by  $\alpha$ (1  $\rightarrow$  4) and  $\alpha$ (1  $\rightarrow$  6) glycosidic linkages (Table 1) (Jiménez, Fabra, Talens, & Chiralt, 2012). Starch in the form of granules is found in a variety of plants, including potatoes, corn, rice, beans, and wheat, as energy storage and is therefore highly abundant in nature. The short-branched chains of amylopectin form the crystalline regions of the starch, whereas the branching points of amylopectin together with the linear amylose chains compose the amorphous regions (Alcázar-Alay & Meireles, 2015). Starch film formation is dependent on the fraction of amylose (Liu & Han, 2005). Starch films have good oxygen barrier properties due to their highly ordered network structure of amylose and amylopectin in alternating crystalline and noncrystalline layers. Both the oxygen barrier properties and water sensitivity of starch films are improved with increased crystallinity, that is, increasing level of amylopectin (Cazón et al., 2017). The tensile strength of starch films is relatively high, whereas due to the crystalline structure, the elastic properties are poor. Addition of plasticizer can to some extent reduce the brittleness of the films (Thakur et al., 2019). Different strategies have been studied to overcome the brittleness of starch films, for example, blending with polyvinyl alcohol (PVA) (Gómez-Aldapa et al., 2020; Wu et al., 2017). Further, crosslinking the starch network is a common strategy to improve the films' resistance to water, while also contributing to better mechanical properties and thermal resistance (Reddy & Yang, 2010). Starch is not inherently thermoplastic, but thermoplastic starch (TPS) can be prepared by processing starch and plasticizer in an extruder at high shear and pressure at 140 to 160 °C (Abdul Khalil et al., 2018; Jiménez et al., 2012). Commercial TPS-based materials are on the market both in the form of rigid trays and flexible films, for example, the Mater-BI series of Novamont (Italy).

Alginates compose a versatile group of linear block copolymers of two uronic acids:  $\beta$ -D-mannuronic acid (M unit) and  $\alpha$ -L-guluronic acid (G unit) linked together by 1  $\rightarrow$  4 glycosidic linkages (Table 1). Alginate is an important structural polymer in the cell wall of marine brown algae (*Phaeophyceae*) and is also produced by some soil bacteria (Draget, Moe, Sják-Bræk, & Smidsrød, 2006). The fraction and sequence of M and G units in alginate are determinant factors for its properties (Smidsrød & Moe, 1995). In particular, the length and distribution of G-blocks is important

for its gel-forming potential as the G-blocks can align and form complexes with di- or trivalent cations (typically calcium,  $\text{Ca}^{2+}$ ), inter- or intramolecularly resulting in a gel lattice (Skjåk-Bræk, Smidsrød, & Larsen, 1986). The gel-forming properties of alginates can be utilized in film formation as crosslinking has shown to improve the water barrier properties and the mechanical properties of the resulting material after solvent evaporation (Benavides, Villalobos-Carvajal, & Reyes, 2012). Further, in comparison with films of other natural biopolymers, alginate films generally have relatively good tensile strength, flexibility, and mechanical strength and  $\text{O}_2$  barrier, while also being relatively fat resistant (Shahabi-Ghahfarrokhi, Almasi, & Babaei-Ghazvini, 2020). A benefit for food packaging is the organoleptic properties of alginates, being relatively tasty and odorless (Puscaselu, Gutt, & Amariei, 2019). Limitations to the use of alginate films as food packaging material are related to high permeability to water (Shahabi-Ghahfarrokhi et al., 2020).

Chitosans are a heterogeneous group of cationic polysaccharides prepared by alkaline deacetylation of chitin—the major structural polysaccharide of the exoskeleton of crustaceans and insects as well as in the cell walls of some algae and fungi. The main commercial source of chitosan is shellfish waste (Vårum & Smidsrød, 2005). Chitosan is a linear polysaccharide of  $\beta(1 \rightarrow 4)$ -linked 2-acetamide-2-deoxy-D-glucose (acetylated, A-unit) and 2-amino-2-deoxy-D-glucose (deacetylated, D-unit) (Table 1) (Smidsrød & Moe, 1995). Chitosans can be prepared with a wide range of acetylated units and chain lengths. The fraction of acetylated units, fraction of acetylation ( $F_A$ ), can vary from 0 (0% acetylated) to 0.7 (70% acetylated). The nonpolar acetyl groups convey hydrophobic properties to chitosan and based on the  $F_A$ , chitosan may be considered as mainly an ampholyte or mainly a polyelectrolyte (Nilsen-Nygaard, Strand, Vårum, Draget, & Nordgård, 2015). Chitosan-based films typically have good mechanical properties as well as low gas permeability (carbon dioxide and oxygen). However, as for most other natural biopolymers, the relatively high water vapor permeability (WVP) of chitosan films puts limitations to the applications (Elsabee & Abdou, 2013). Though most commercially available chitosans have low  $F_A$ , it has been demonstrated that chitosans of higher content of acetyl groups form more water-resistant films due to their more hydrophobic nature (Kim, Son, Kim, Weller, & Hanna, 2006). Another interesting aspect of chitosan is its inherent antimicrobial properties. The mode of action causing the antimicrobial activity is not yet fully understood; however, it is believed that electrostatic interaction between positively charged chitosan and negatively charged microbial cell membranes plays an important role. This interaction assumedly leads to increased membrane permeability and hydrolysis of the

peptidoglycans in the microorganism wall, inhibiting bacterial growth (Kong, Chen, Xing, & Park, 2010). Studies have shown that the antimicrobial effect is most pronounced at low  $F_A$  and at low pH (Younes, Sellimi, Rinaudo, Jellouli, & Nasri, 2014), that is, higher charge density of chitosan.

Protein-based films developed from, for example, wheat gluten, whey protein, soy protein, corn protein, and gelatine have gained much attention because of their abundance, excellent film forming properties, low cost, biodegradability, and compostability (Hassan, Chatha, Hussain, Zia, & Akhtar, 2018; Zubair & Ullah, 2020). The unique structure of the proteins, especially their high intermolecular binding potential via covalent bonds, confers a wide range of functional properties to the protein-based films, often exceeding the mechanical properties of polysaccharide and lipid-based films (Coltelli et al., 2016). Wheat gluten protein is an agricultural biopolymer containing water-soluble monomeric gliadins (molecular weight between 30 and 50 kDa) and a water-insoluble glutenin (up to 300 kDa), besides low-molecular-weight proteins such as albumins and globulins.  $\beta$ -Lactoglobulin is the major protein fraction in whey making up 57% of the total protein, whereas  $\alpha$ -lactalbumin is the second most abundant whey protein. Soy protein consists of two major globular proteins:  $\beta$ -conglycinin and glycinin (Song, Tang, Wang, & Wang, 2011). Derived from collagen in animal skin and bones (e.g., bovine and pork) and in fish by-products (Etxabide, Uranga, Guerrero, & de la Caba, 2017), gelatine consists of a pool of protein segments carrying different molecular weights (100 to 300 kDa), along with high-molecular-weight aggregates and peptide fractions (<100 kDa). Both the origin and the extraction process influence the average molecular weight of gelatine, hence its film forming capacity (Gómez-Estaca, Gavara, Catalá, & Hernández-Muñoz, 2016). Furthermore, the molecular weight of the protein and its distribution influence film mechanical properties, as seen in different soya protein fractions (Cho & Rhee, 2004), in addition to protein composition and their partial modifications as investigated for wheat gluten fractions (Hernández-Muñoz, Villalobos, & Chiralt, 2004).

### 3.2 | Preparation of natural biopolymer-based films

Solvent casting is the most common film-forming method in which natural biopolymers are dissolved or dispersed in appropriate solvent (e.g., water, ethanol, methanol, and acetone or a combination of two) and mixed with plasticizer (e.g., glycerol, polyols, mono-, di-, and oligosaccharide, lipids, or mixed systems) depending on the

film-forming biopolymer. The resulting solutions or dispersions are casted on a plate and dried to form self-supporting films (Zhang & Mittal, 2010). However, solvent casting is demanding in terms of required area and drying times and therefore not suitable for industrial-scale film production and research effort is focused on developing preparation methods that are more suitable for commercial use. Dry processes, such as thermoplastic extrusion, take advantage of thermoplastic properties of biopolymers exhibited when they are plasticized and heated under low water content above glass transition temperature, for example, TPS (González, Iturriaga, González, Eceiza, & Gabilondo, 2020), and can be combined with film blowing (Brandelero, Grossmann, & Yamashita, 2011). Mendes et al. (2016) reported successful preparation of corn starch/chitosan films by extrusion followed by hot melt compression. Another emerging technology for film preparation is the electrospinning technique, which is particularly beneficial in preparation of functional nanocomposite films (Zhao et al., 2020).

### 3.3 | Limitations

Although films of natural biopolymers generally have relatively good gas barrier properties, overall their functional properties are inferior to those of conventional plastic materials, such that they are mechanically weaker, more brittle, and have higher WVP compared to fossil-based plastics (Cazón et al., 2017). Examples of selected physical properties of some natural biopolymers are displayed in Table 2. They are also more susceptible to lipid oxidation and microbial spoilage, the property ensuring their “biodegradability,” and thus limiting the shelf life of these packaging films as compared to petroleum-based plastics (Robertson, 2009). Their inherent hygroscopicity makes these films instable in humid environments and in contact with foods with a high water content, posing a challenge in optimizing their properties and identifying suitable applications. Further, their mechanical properties put limitations to their applications and processability at an industrial scale; in most cases, natural biopolymer films cannot be processed by industrial methods such as extrusion or film blowing. Modifications and techniques for improving properties of biodegradable materials are discussed in Section 4.

## 4 | MODIFICATIONS OF BIOBASED, BIODEGRADABLE MATERIALS

To overcome the challenges pinpointed in Section 2 and 3 related to the use of synthetic and natural biopolymers for

food packaging films, to this end, physical, chemical, and biochemical modifications, for example, plasticization, thermal, chemical, and enzymatic cross-linking, ionizing irradiation, pH alteration, incorporation of antimicrobial or antioxidant compounds, lipids, and nanoparticles, are applied during or after film formation to enhance the structural, mechanical, and functional properties of the polymers. This section gives a brief overview of some of the most established chemical modifications as well as novel and promising nanotechnology-based modifications of biodegradable materials. For an in-depth review on plasticization of biobased materials, we refer to the publication by Vieira, da Silva, dos Santos, and Beppu (2011). The physical and thermal treatment of packaging materials (e.g., plasma treatment and MW) through the means of innovative processing technologies is described in Section 6.

### 4.1 | Lipids

One of the critical factors limiting extensive use of biodegradable materials is their hygroscopic nature, causing them to absorb water and loose structural integrity. One way of increasing the hydrophobicity of biomaterials is addition of lipids, such as waxes and oils. The addition of a hydrophobic phase, often by surfactant-assisted dispersion, has been reported to reduce the WVP of the materials (Castro-Rosas et al., 2016). In a recent study, Syahida et al. studied effect of different concentrations of palm wax oil on the properties of fish gelatine films (Syahida, Ismail-Fitry, Zuriyati, & Nur Hanani, 2020). The study reported that at a concentration of palm wax oil of 15%, WVP of the films was significantly reduced compared to the pristine films. Similar effects have been reported for different combinations of film-forming polymer and lipids; PLA and beeswax (Lim, Kim, Ko, & Park, 2015), wheat gluten protein, acetic esters of mono and diglycerides and beeswax (Rocca-Smith et al., 2016), and essential oils (EOs; e.g., thyme oil, rosemary oil, and cinnamon oil) from plants contain volatile aroma compounds acting as antimicrobial or antioxidative agents (Ribeiro-Santos, Andrade, Melo, & Sanches-Silva, 2017). EOs are widely used in preparation of active films. The hydrophobic nature of these oils has been shown to have an additional effect on biodegradable films in terms of reducing WVP (Atarés & Chiralt, 2016). Active films and methods for incorporation of active compounds such as EOs are described in Section 4.5. Other described properties of films incorporated with lipids are increased opacity, rougher surface morphology, and poorer mechanical properties as the lipids do not contribute in polymer network formation (Castro-Rosas et al., 2016). These aspects imply careful optimization is necessary in incorporating lipids into biodegradable film matrixes.

## 4.2 | Mixed biopolymer systems: Blends

Blending of polymers is an extensively used, low-cost technique for tailoring the properties of the resultant packaging film by matching complementary properties of different polymer types. The microstructure of polymer blends depends on the net interaction between the polymers and blending conditions such as temperature, pH, and ionic strength. Blends of chitosan and PVA (Haghighi et al., 2020), starch and PVA (Domene-López, Guillén, Martín-Gullon, García-Quesada, & Montalbán, 2018), and chitosan and starch films (Luchese et al., 2018) are some of the promising polymer combinations recently studied for food packaging applications. Compatibilization through copolymerization is a collective term for different techniques and additives used to improve the miscibility and compatibility of polymers in blends, ensuring more homogeneous and stable films. For a comprehensive overview on compatibilization of biopolymer blends, we refer to the paper by Imre and Pukánszky (2013).

## 4.3 | Crosslinking

The crosslinking method to improve biomaterial properties involves formation of chemical bonds between different polymer chains, either intra- or intermolecularly, yielding stronger and more tightly bound three-dimensional networks. Type of crosslinking is often classified according to bond type/type of interaction (covalent, ionic, van der Waals, or H-bonds) or on the basis of mode of action—chemical, physical, or enzymatic (Garavand, Rouhi, Razavi, Cacciotti, & Mohammadi, 2017). Crosslinking may improve the properties of biodegradable films. For instance, tightening of the polymer lattice in biopolymer films can restrict the movement of water molecules through the film, resulting in a lower WVP. Wu and coworkers studied citric acid (CA) crosslinking of potato starch/chitosan and found that with 15% CA (per weight of polymer), the WVP decreased from  $3.03 \times 10^{-12}$  g·cm/cm<sup>2</sup> for noncrosslinked films to  $2.05 \times 10^{-12}$  g·cm/cm<sup>2</sup> and the degree of swelling decreased from 686.4% of initial film weight to 98.1%. However, tensile strength increased by 29%, which can be attributed to formation of shorter elastic segments within polymer lattice (Wu et al., 2019). Interestingly, the study also documented how an excessive amount of CA resulted in poorer mechanical properties. This can be attributed to the plasticizing effect of free crosslinker molecules (Garavand et al., 2017). Picchio et al. studied the effect of crosslinking of casein films with tannic acid on the mechanical, thermal, water resistance, and degradation properties. They reported that tensile strength of the crosslinked films increased while the

elongation at break decreased. Further, the water resistance of the films was significantly improved compared to the pristine casein films. Although noncrosslinked films dissolved within 24 hr in water, crosslinked films were still structurally intact after 7 days. Degree of swelling was also significantly reduced (Picchio et al., 2018). In a study by Cui and coworkers, enzymatic crosslinking of gluten by transglutaminase—enhanced by  $\alpha$ -polylysine—was applied to prepare gluten films with improved mechanical properties and better resistance to water (Cui et al., 2017).

## 4.4 | Reinforced biodegradable materials: Biocomposites and bionanocomposites

For industrial food packaging applications, multilayered structures consisting of different polymers are used for suitable barrier and mechanical properties (Anukiruthika et al., 2020). However, these complex structures increase cost, require use of supplementary additives and adhesives, and above all are mostly not recyclable or biodegradable. To overcome these challenges, there is a strong drive in the industry to develop more sustainable and green packaging solutions with improved mechanical and barrier properties. Polymer nanocomposites (PNCs) offer these desired functionalities. PNCs are formed by dispersing an inert, nanoscale filler throughout a polymeric matrix. These filler materials may include clay and silicate nanoplatelets, silica (SiO<sub>2</sub>) nanoparticles (Wu, Zhang, Rong, & Friedrich, 2002), carbon nanotubes (Chen, Tao, Xue, & Cheng, 2005; Zhou, Shin, Wang, & Bakis, 2004), graphene (Borriello et al., 2009), starch nanocrystals (Chen, Cao, Chang, & Huneault, 2008), cellulose-based nanofibers or nanowhiskers (Bilbao-Sáinz, Avena-Bustillos, Wood, Williams, & McHugh, 2010; Cao, Chen, Chang, Stumborg, & Huneault, 2008), chitin or chitosan nanoparticles (Lu, Weng, & Zhang, 2004), and other inorganics (Ma, Qian, Yin, & Zhu, 2002). A tortuous pathway is normally formed upon incorporation of nanomaterials in the polymer matrix. The tortuosity created by nanofillers affects diffusion rate of gas molecules (Nielsen, 1967) and is the primary reason for the improved barrier properties. Apart from tortuosity, polymer–nanoparticle interaction can also influence barrier properties by immobilizing polymer strands.

Khankrua et al. have studied the thermal and mechanical properties of biodegradable polyester/silica nanocomposites. Nanocomposites of PLA, PBS, and PHBV with hydrophilic fumed silica (0.1% to 5% per weight of polymer) were prepared by twin screw extrusion. They reported that although at low concentrations of silica (less than 0.5%), the tensile modulus was slightly increased however,

at the higher silica loading it deteriorated because of the interaction between the polar groups on surface of silica. On the other hand, both elongation at break and impact strength worsened at silica loading of more than 0.5% (Khankrua, Pivsa-Art, Hiroyuki, & Suttiruengwong, 2013). Zhang, Wang, and Cheng (2018) reported an improvement in the tensile strength as well as water and gas barrier properties with the incorporation of nanosilica in potato starch film.

A different approach using surface modification of nanoparticles has also been reported to overcome the challenge of agglomeration and to obtain more homogeneous distribution of particles in the polymer matrix. Lai and Hsieh (2016) have reported PE glycol methyl ether grafted silica particles via amino-silane. They reported better dispersion of modified silica particles in PLA matrix with improved tensile strength compared to polymer composite with unmodified particles. Similarly, Yan et al. (2007) and Zhu, Diao, Rong, and Cai (2010) have reported PLA composite with nanosilica grafted with lactic acid and oleic acid, respectively. Elongation at break of PNC with lactic acid modified silica was improved by eight times (Yan et al., 2007). Improved gas barrier properties have also been reported for vapor-deposited thin silica and alumina films on polymer substrates; however, these films are susceptible to cracking upon bending (Affinito et al., 1996; Leterrier, 2003). Apart from silica, other nanoparticles have also been reported, for example, magnesium oxide (MgO)/PLA composite with 25% improved gas barrier properties (Swaroop & Shukla, 2018).

Clays and other silicate materials are inexpensive, have high stability, and are supposedly nontoxic. Nanoclay-based PNCs are the foremost choice for food contact application. Typical clays, for example, montmorillonite (MMT), kaolinite, hectrite, and saponite, have been reported for PNC application (Yano, Usuki, & Okada, 1997). Maiti, Yamada, Okamoto, Ueda, and Okamoto (2002) have studied the effect of organic modifiers of various chain lengths in different types of clays, smectite, MMT, and mica on degree of dispersion of clay in PLA. Smectite nanocomposites have shown better gas barrier properties in comparison with MMT or mica nanocomposites. Swaroop and Shukla (2018) have reported nano-MgO-reinforced PLA films. Reinforced PLA films with 2% MgO showed maximum improvement in tensile strength and oxygen barrier properties (up to 29% and 25%, respectively) in comparison to pristine PLA films. Zahedi, Fathi-Achachlouei, and Yousefi (2018) reported a dramatic increase in Young's modulus from 25 to 40 MPa of MMT-reinforced carboxymethyl cellulose (CMC) nanocomposite.

## 4.5 | Active biomaterials

According to European commission regulation (EC) No 450/2009 "active materials and articles means materials and articles that are intended to extend the shelf-life or to maintain or improve the condition of packaged food". Active biomaterials are designed to deliberately incorporate components that would release or absorb substances into or from the packaged food or the environment surrounding the food. Furthermore, Framework Regulation (EC) 1935/2004 provides specific requirements for active materials and articles.

The manufacturing processes, including nanotechnology, have been discussed in the food guidance provided by the FDA. The approach of FDA toward nanotechnology is summarized as, "FDA does not categorically judge all products containing nanomaterials or otherwise involving application of nanotechnology as intrinsically benign or harmful. Rather, for nanotechnology derived and conventionally manufactured food products alike, FDA considers the characteristics of the finished product and the safety of its intended use" (FDA, 2014). Readers who are more interested in this topic and want to know more about the regulatory aspects of nanotechnology in different regions of the world, we would kindly refer them to articles in this area by Amenta et al. (2015) and He, Deng, and Hwang (2019).

In literature, active packaging is subcategorized into antimicrobial packaging and addition of labels, sachets, or pad. Antimicrobial packaging can be developed by direct addition of the antimicrobial components through, for example, solvent compounding, in the melt or as nanocomposites and so on. Active components can also be attached or coated to the surface (Ahmed et al., 2017).

Antimicrobial nanocomposites offer an extension in the shelf life of the product by suppressing the growth of microorganisms. Metal nanoparticles, for instance, silver, titanium, copper, and zinc, have shown a great potential for antimicrobial packaging application. Similarly, metal oxide-based, such as zinc oxide (ZnO), MgO, and titanium oxide (TiO<sub>2</sub>), antimicrobial packaging has been reported in literature (Shankar & Rhim, 2016). More recently, antimicrobial packaging based on natural antimicrobials such as EOs and their active components encapsulated in nanoclays or silica has shown a great promise (Melendez-Rodriguez et al., 2019; Shemesh et al., 2015).

Bashir et al. have prepared novel biodegradable films by blending guar gum, chitosan, and PVA with mint and grapefruit peel extracts and crosslinked with nontoxic tetraethoxysilane. The ultimate tensile strength and elongation at break (%) values of the modified films were reported to be 40 MPa and 104%, respectively.

Furthermore, the higher antioxidant activity was confirmed at 37 °C compared to 28 °C (Bashir et al., 2018). Zhu et al. have reported a novel antimicrobial bilayer films based on PLA/Pickering emulsions. The antibacterial activity was evaluated by agar disk diffusion method against *Staphylococcus aureus* and *Escherichia coli* (Zhu, Tang, Yin, & Yang, 2018). In a study by Mayorga et al., antimicrobial nanocomposites and electrospun coatings based on PHBV and copper oxide nanoparticles were investigated. Reduction of 5 log colony forming units (CFU)/mL of *Salmonella enterica* for the films prepared with 0.05% copper oxide (CuO) was reported (Mayorga, Rovira, Mas, Moragas, & Cabello, 2018). For profound reviews on active agents and active packaging applications, the reader is referred to Vilela et al. (2018) and Yildirim et al. (2017), respectively.

## 5 | BIODEGRADABLE MATERIALS FOR FOOD APPLICATIONS: EFFECT ON FOOD QUALITY AND SHELF-LIFE

In this section, an overview of recent studies on the use of biodegradable materials for real food applications is provided with emphasis on the effect on food quality and shelf life. The search has been conducted on the Web of Science, limited to a period of 5 years (2015 to 2020) with use of selected key words: “food packaging” and “shelf life”, combined with the different polymers. In Table 3, selected studies from this search are presented, describing applications of biodegradable packaging materials for packing of specific food substrates, where the biomaterial is used as main packaging material (i.e., not as coating on or in combination with conventional packaging materials), as well as the specific effect of these systems on food shelf life and/or quality.

Studies related to the application and effect of biodegradable films on real food products are rather scarce. Most of the studies on biomaterials resulting from the literature search stated potential applications in the food packaging field. In addition, among the studies addressing the impact on food products, almost none use pristine materials. Most of the scientific articles describe modified biodegradable films incorporated with, for example, nanoparticles, extracts, antimicrobials, and/or microencapsulation. Change in the organoleptic properties of food products packaged in active films can be a challenge due to the strong odor of, for example, EOs. This aspect is often not discussed in scientific studies.

### 5.1 | Synthetic biobased, biodegradable polymers

PLA is considered as a promising material for food packaging application. It can be processed as most conventional polymers to flexible films, extruded packages, containers, bottles, cups, and lunch boxes. As already mentioned in Section 2.1, some limitations (e.g., poor barrier properties) are reducing its wider application in food packaging (Auras et al., 2004; Mangaraj et al., 2019; Platt, 2006; Rhim & Ng, 2007). For this reason, most of the studies were related to the improvements of PLA properties by addition of nanoparticles and plasticizers and by application of advanced processing techniques or treatments (Yildirim et al., 2017). Moreover, it can be successfully incorporated with diverse antimicrobial substances (plant extracts, EOs, enzymes, etc.) (Radusin et al., 2019; Scaffaro, Lopresti, Marino, & Nostro, 2018). Patanè et al. (2018) reported that PLA is more suitable for packaging of fresh-cut long storage tomato, as compared to PP films. The PP film promoted a fog effect resulting in modification of the headspace atmosphere, whereas PLA prevented any visible water condensation (anti-fog effect). The potential of PLA as packaging material for red meat has also been reported (Panseri et al., 2017). The main finding in this study was that PLA-based packaging is a suitable material for preservation of red cherry color in refrigerated red meat during its entire shelf life, as compared to conventional packaging (PET). PLA-PHB films have been applied for packaging of oysters and compared to conventionally used high-barrier films of ethylene vinyl alcohol (EVOH). However, the reported extended shelf life was related to the addition of antimicrobial EOs rather than to the biomaterials used (Miao, Walton, Wang, Li, & Wang, 2019). Wen et al. (2016) reported an increase in shelf life of pork meat by 4 days (at 25 °C) compared to the control, by using a novel antimicrobial packaging material obtained by incorporating cinnamon EO/ $\beta$ -cyclodextrin ( $\beta$ -CD) inclusion complex into PLA nanofibers via electrospinning technique. Studies related to the application of PLA-based materials for food packaging applications (dry food, fruits, vegetables, fresh and processed meat, fish and sea products, etc.) have increased substantially over the last 5 years due to the growing awareness of environmental impact of plastic waste. Examples include baby formula packed in PLA/whey protein isolate (WPI) pouches for delayed lipid oxidation (Phupoksakul, Leuangasukrerak, Somwangthanaroj, Tananuwig, & Janjarasskul, 2017), minced fish or different fresh salads packed in PLA with addition of EOs (Llana-Ruiz-Cabello et al., 2015; Llana-Ruiz-Cabello et al., 2016; Zeid, Karabagias, Nassif, & Kontominas, 2019), and sliced salami sausage and cottage cheese packed in PLA nanocomposites

**TABLE 3** Biobased, biodegradable food packaging systems and effect on food quality and shelf life

<b>Material composition</b>	<b>Packaging method</b>	<b>Food product</b>	<b>Storage conditions</b>	<b>Benefit</b>	<b>Reference</b>
Synthetic biobased, biodegradable polymers					
PLA (compared with PET + PP)	Tray packed in compostable bags	Fresh cut "long storage tomato" (dipped in CaCl <sub>2</sub> )	4 °C, dark, 12 days	Suitable quality and shelf life, no condensation, less susceptible to microbiological spoilage	Patanè et al., 2018
PLA (compared with amorphous polyethylene terephthalate/PE trays wrapped with PVC)	Trays with lids packed in MAP (66% O <sub>2</sub> /25% CO <sub>2</sub> /9% N <sub>2</sub> )	Red fresh meat ( <i>Semimembranosus</i> muscle Piemontese beef)	4 °C, 8 days	Maintain red color, reduced content of volatiles (off-odor)-oxidation	Panseri et al., 2018
PLA/poly butylene adipate-co-terephthalate (PBAT) blend (compared with PE and unpacked)	Film wrapping	Passion fruit	20 °C, 21 days	Better preservation of sensory quality; overall flavor and edible quality, during storage time.	R. Zhang, W. Lan, et al., 2019
PLA/WPI/PLA (Control: PLA)	Pouch	Baby formula (dry product)	4, 25, and 35 °C, 50% to 59% RH, 19 days	Delayed lipid oxidation compared to pure PLA.	Phupoksakul et al., 2017
PLA incorporated with EOs (Control: PLA)	Fish burgers sandwiched between films and packed aerobically in LDPE sealed bags	Minced fish	4 °C, 8 days	Reduction in oxidation degree, increased shelf life.	Zeid et al., 2019
PLA films containing oregano essential oil ( <i>Origanum vulgare</i> L. virens) (compared with PP)	Extruded films placed inside PP bags with MAP (10% O <sub>2</sub> , 10% CO <sub>2</sub> , 80% N <sub>2</sub> )	Iceberg salad	4 °C, 7 days	Antimicrobial activity of EOs against yeast and mold.	Llana-Ruiz-Cabello et al., 2016
PLA/PBS with oregano EO (compared with PLA/PBS)	Packaging bags with MAP (10% O <sub>2</sub> , 10% CO <sub>2</sub> , 80% N <sub>2</sub> )	Lettuce	4 °C, 8 days	Prolonged shelf life by addition of EOs up to 8 days; EO allowed preservation of lettuce for up to 8 days.	Llana-Ruiz-Cabello et al., 2019

(Continues)



TABLE 3 (Continued)

Material composition	Packaging method	Food product	Storage conditions	Benefit	Reference
PLA/PHB with fennel oil (compared with EVOH)	Film wrapping	Oysters	4 °C, 16 days	Prolonged shelf life (2 to 3 days compared to EVOH films)	Miao et al., 2019
PLA-PHB with carvacrol and thymol (compared with PLA-PHB and EVOH)	Packaging bags	Salmon	4 °C, 18 days	Extended shelf life of 2 days	Ma, Li, & Wang, 2018
PLA-P3, 4HB-P3, 4HB-poly(3-hydroxybutyrate-4-hydroxy butyrate)/Angelica EO/ginger EO (control sample unwrapped peach and PLA-P3, 4HB)	Sealed bags	Peach	1 °C, 90% RH, 30 days	Better preservation-extended shelf life	Jiang et al., 2020
PLA/allium extract (control: neat PLA)	Film wrapping and packed in Oriented polypropylene (OPP) with MAP (10% O <sub>2</sub> , 10% CO <sub>2</sub> , 80% N <sub>2</sub> )	Ready-to-eat salads (lettuce)	4 °C, 7 days	Antimicrobial effect confirmed—Yeast and molds (more than enterobacterial and aerobic bacteria)	Llana-Ruiz-Cabello et al., 2015
PLA/nanochitosan nanocomposite (control sample neat PLA)	Pouches	Indian white prawn ( <i>Fenneropenaeus indicus</i> )	2 °C, 18 days	Antimicrobial films	Fathima, Panda, Ashraf, Varghese, & Bindu, 2018
PLA/PLS (polymer layered silicate) (control sample neat PLA)	Pouches (packed in vacuum)	Salami sausage, sliced	5 °C, 90 days	Reduction in lipid oxidation mainly due to enhanced water barrier properties	Vilariño et al., 2018
PLA/nanoclays (compared with neat PLA and conventional PET)	Trays (designed for fresh fruits) with lid + fork	Fresh-cut melons	10 °C, 80% RH, 7 days	Less environmental impact, extended shelf life and less food losses	Lorite et al., 2017
PLA; PLA/TiO <sub>2</sub> ; PLA/TiO <sub>2</sub> +Ag (compared to LDPE)	Pouches	Cottage cheese	5 °C, 25 days	Addition of TiO <sub>2</sub> and Ag are maintaining and/or prolonging cheese shelf life up to 25 days	Li et al., 2018.

(Continues)

TABLE 3 (Continued)

Material composition	Packaging method	Food product	Storage conditions	Benefit	Reference
Electrospun PLA nanofibers/ $\beta$ -cyclodextrin inclusion complex/cinnamom EO	Film wrapping	Pork meat	25 °C, 8 days.	Increase in microbiological shelf life by 4 days	Wen et al., 2016
PBSA/PLA/carvacrol or thymol	Film wrapping	Salmon slice	4 °C, 3 to 4 days	Increased shelf life by 3 to 4 days during cold storage	Yang et al., 2019
PBS/PBSA	Film wrapping	Raw chicken and turkey meat	4 °C, 5 days for raw poultry and 15 days for smoked poultry	Extended shelf life	Vytejčková et al., 2017
PHBV/chitosan/ZnO/Ag	Film wrapping	Chicken breast filets	4 °C	Sensory quality after 15 days of storage was significantly better compared to unpackaged chicken breast filets	Zare et al., 2019
PHB/polycaprolactone/nisin/organoclays	Ham sandwiched between films and vacuum packaged in pouches	Ham	5 °C	Extended shelf life from 7 to 28 days of Lactobacillus plantarum CRL691innoculated product	Correa et al., 2017
Natural biobased, biodegradable polymers					
Carboxymethyl cellulose with Thai rice grass extract	Sealed sachet	Green tea	10, 30, and 70 °C for 25 days	Adding Thai rice grass extract increased shelf life from 32 in CMC to 37 days. HDPE gave 44	Rodsamran & Sothornvit, 2018
Methylcellulose/tannic acid	Film wrapping	Cherry tomatoes and grapes	21 to 23 °C for 14 days	Reduction in weight loss (from 26.2% to 20.5%) and browning index (from 164 to 75) by adding tannic acid. Commercial cling film had 15.5% weight loss and browning index of 41.	Halim et al., 2018
CMC/gelatin	Film wrapping	Cherry tomatoes and grapes	Room temperature, 14 days	Decreased browning and weight loss adding CMC up to 75%	Samsi et al., 2019.

(Continues)

TABLE 3 (Continued)

Material composition	Packaging method	Food product	Storage conditions	Benefit	Reference
CMC/okra muilage/ZnO nanoparticles	Film wrapping	Chicken breast fillet	4 °C for 12 days	ZnO and okra increased shelf life from 6 days in CMC to 12 days. Total volatile nitrogen and lipid oxidation were also reduced (up to 40%)	Mohammadi et al., 2019
Chitosan/cellulose acetate phthalate/ZnO nanoparticles	Film wrapping	Black grapefruits	Room temperature, 9 days.	Cellulose/chitosan/ZnO extended shelf life from 3 in PE to 6 days.	Indumathi et al., 2019
Potato starch/butylated hydroxytoluene or green tea extract	Film wrapping	Beef	4 °C and room temperature, 10 days.	Reduced lipid oxidation (thiobarbituric acid-reactive substances [TBARS]) and protein oxidation (metmyoglobin formation)	Nisa et al., 2015
Tapioca starch film/chitosan nanoparticles	Film wrapping	Cherry tomatoes	10 °C for 10 days	Addition of chitosan inhibited gram-positive growth.	Shapi'i et al., 2020
Cowpea starch/maqui berry extracts	Film wrapping	Salmon	4 °C for 6 days	Antioxidant effect, almost inhibiting lipid oxidation by adding extracts	Baek et al., 2019
Cassava starch/EVOH/alginate	Film wrapping	Lettuce	6 °C for 8 days	Equal performance as PVC	Brandelero et al., 2016
Alginate/EOs/citrus extract and Ozonation	Film wrapping	<i>Merluccius</i> sp. filets	4 °C, 14 days.	Increased shelf life from 7 days to 21 days	Shankar et al., 2019
Alginate/EOs/citrus extract and Gamma Irradiation	Film wrapping	<i>Merluccius</i> sp. filets	4 °C, 21 days.	Increased shelf life from 7 days to 28 days	Shankar et al., 2019
Alginate/ $\beta$ -CD/carvacrol	Film wrapping	Mushroom	4 °C, 12 days.	Reduced weight loss, extended microbial and sensory shelf life	Cheng et al., 2019
Alginate/zinc oxide nanoparticles/ <i>Ziziphora clinopodioides</i> EO/apple peel extract	Film wrapping	Sauced silver carp filets	4 °C, 14 days.	Delayed microbial and chemical spoilage, increased shelf life	Rezaei & Shahbazi, 2018

(Continues)

TABLE 3 (Continued)

Material composition	Packaging method	Food product	Storage conditions	Benefit	Reference
Alginate/starch/polyvinyl alcohol (PVOH)	Film wrapping	Lettuce	6 °C, 60% RH	Better retention of color and eight times less mass loss	Brandelero et al., 2016
Alginate/Marjoram EO/clay	Film wrapping	Rainbow trout slice	4 °C, 15 days	Delayed growth of <i>L. monocytogenes</i> during the 15-day storage	Alboofetileh et al., 2016
Alginate/gelatin/oregano	Film wrapping	Rainbow trout	7 °C, 15 days	Increased shelf life	Kazemi & Rezaei, 2015
Chitosan/Chitosan/apricot kernel oil	Pouches	Bread slices	Room temperature, 10 days	Lower fungal growth compared to packaging in LDPE. Extended shelf life	Priyadarshi, Sauraj Kumar, Deeba, et.al., 2018
Chitosan	Pouches	Green chili	27°C, 7 days	Extended shelf life and lower moisture loss compared to unpackaged product.	Priyadarshi, Sauraj Kumar, & Negi, 2018
Chitosan with kombucha tea	Film wrapping	Minced beef	4 °C, 6 days	Extended shelf life with 3 days.	Ashrafi et al., 2018
Chitosan with EOs	Pouches	Peanut kernels	28 °C, 14 days	Reduction of fungal growth	Chein, Sadiq and Anal, 2019
Chitosan/ginger oil	Film wrapping	Barracuda fish fillets	2 °C, 20 days	Extended microbial and sensory shelf life compared to packaging in EVOH	Remya et al., 2016
Chitosan/corn starch/cinnamaldehyde EO	Film wrapping	Strawberries	20 °C, 65% RH, 14 days	Inhibition of fungi and <i>E. coli</i> . Extended shelf-life compared to PP film	Wang et al., 2019
Chitosan/ε-polylysine nanofibers	Film wrapping	Chicken	4 °C, 14 days 25 °C, 7 days	Reduction of inoculated <i>S. typhimurium</i> and <i>S. enteritidis</i> , improved sensory quality (appearance, color, flavor, juiciness)	Lin et al., 2018

(Continues)

TABLE 3 (Continued)

Material composition	Packaging method	Food product	Storage conditions	Benefit	Reference
Chitosan/montmorillonite/EOs	Film wrapping	Minced poultry meat	5 °C, 15 days	Extended shelf life compared to unwrapped product. Color preservation. Reduced lipid oxidation.	Pires, Souza & Fernando, 2018
Chitosan/montmorillonite/gi EO	Film wrapping	Poultry meat	5 °C, 15 days	Reduced lipid oxidation, extended microbial and physicochemical shelf life	Souza et al., 2018
Chitosan/montmorillonite/rosemary EO	Film wrapping	Poultry meat	5 °C, 15 days	Extended microbial shelf life. Reduction of TVC by 1.2 to 2.1 log CFU/g compared to unwrapped meat.	Souza et al., 2019
Zein/pomegranate peel extract (PPE)	Film wrapping	Fresh Himalayan cheese	Refrigerated storage, 30 days	PPE retarded cheese spoilage, oxidation of fats and proteins of cheese.	Mushtaq et al., 2018
Gelatin/catechin-lysozyme	Film wrapping	Minced pork	4 °C, 7 days	Reduced weight loss, retarded development of TBARS; retarded vital plate count, yeast, and mold compared to control in PVC wrap. Color preservation.	Kaewprachu et al., 2015
Fish myofibrillar protein/catechin-Kradon extract	Film wrapping	Bluefin tuna slices	4 °C, 10 days	Delayed lipid oxidation (PV, TBARS) and TVB-N, improved red color preservation. Sensory shelf life was 8 days compared to 2 days for LDPE wrapped control	Kaewprachu et al., 2017

(Continues)

TABLE 3 (Continued)

Material composition	Packaging method	Food product	Storage conditions	Benefit	Reference
Tilapia and squid skin gelatin film/nanoclay/ethanolic extract from coconut husk	Film wrapping	Mackerel meat powder	28 to 30 °C, 30% RH, 30 days	Retarded lipid oxidation; reduced loss of sensory quality	Nagarajan et al., 2015
Whey protein isolate/cellulose nanofiber/TiO <sub>2</sub> nanoparticle incorporated with rosemary EO	Film wrapping	Lamb meat	4 °C, 15 days	Doubled shelf life compared to plastic, and improved quality (retarded growth of spoilage bacteria, improved organoleptic quality)	Sani et al., 2017
Gelatin/ZnO nanorods/clove EO	Heat sealed pouch	Peeled shrimp	4 °C, 20 days	Complete inhibition of inoculated <i>L. monocytogenes</i> and <i>S. Typhimurium</i> after 7 days of refrigerated storage. No bacterial inhibition observed for neat gelatin films.	Ejaz et al., 2018
Soy protein/montmorillonite/clove EO	Film wrapping	Bluefin tuna	2 °C, 14 days	Retarded bacterial growth, lipid oxidation, no diffusion of nanoclay minerals (Si and Al), compared to PE.	Echeverría, López-Caballero, Gómez-Guillén, Mauri, & Montero, 2018.

(Li, Li, Zhang, Yuan, & Qin, 2018; Vilarinho et al., 2018) (Table 3).

The suitability of both PHA and PBS for food packaging applications has already been discussed in Section 2. Based on defined search criteria, just two articles reporting the practical use of PBS-based materials for food packaging have been identified, as well as two articles demonstrating food packaging with PHA derivatives (Table 3). The suitability of PBS and PBSA for vacuum packaging of raw chicken, turkey, and smoked turkey meat was evaluated by Vytejčková et al. (2017). It was suggested that although the physical, chemical, and mechanical properties of commonly used polyamide (PA)/PE and PBS packaging materials are not the same, no significant difference in the quality and safety of packaged food was observed. A shelf life test of salmon slices packed with PLA–PBSA bags (with and without active compound) was carried out by Yang et al. (2019). The shelf life of salmon slices was reported to increase by 3 to 4 days during cold storage due to reduced spoilage and deterioration.

Shelf life study of chicken breast fillets packaged in bionanocomposite films with a composition of PHBV/chitosan/ZnO/Ag gave significantly improved sensory quality after 15 days of storage at 4 °C compared to the control (unpacked product) (Zare et al., 2019), whereas PHB/polycaprolactone incorporated with nisin and organoclays extended the shelf life of ham from 7 to 28 days stored at 5 °C (Correa et al., 2017).

## 5.2 | Natural biobased, biodegradable polymers

Films prepared from modified celluloses have been applied in some studies for food packaging applications. For instance, shelf life of green tea was extended to 110 days in pouches made by CMC incorporated with Thai rice grass extracts microencapsulated powder (MP), as compared to 91 days in CMC pouches (Rodsamran & Sothornvit, 2018). The quality of green tea packaged in CMC–MP was found to be equivalent to that packaged in HDPE. In a study by Halim et al., tannic acid was used to increase the performability of methylcellulose films (Halim, Kamari, & Phillip, 2018). The ability to prevent browning and drip loss of grapes and cherry tomatoes was significantly increased by adding 15% (w/w) tannic acid. However, commercial cling film outperformed both modified CMC films. The same effect was observed by combining CMC and gelatine when wrapping cherry tomatoes and grapes (Samsi, Kamari, Din, & Lazar, 2019). Both browning and weight loss were slowed down compared to unwrapped products, and 75% CMC:25% gelatin was comparable in performance with commercial cling film. Adding okra mucilage

and ZnO nanoparticles is another way to increase the performability of CMC (Mohammadi, Kamkar, Misaghi, Zunabovic-Pichler, & Fatehi, 2019). The bionanocomposite films significantly restricted microbial growth, lipid oxidation, and total volatile nitrogen compared to a pure CMC film when storing chicken breast fillets at 4 °C. ZnO nanoparticles have also been used in combination with chitosan–cellulose acetate phthalate, where the shelf life of black grapes was increased threefold compared to control packaged in PE (Indumathi, Sarojini, & Rajarajeswari, 2019).

Cherry tomatoes wrapped in tapioca starch film exhibited significantly lower microbial growth at day 3 and 10 of storage at 10 °C than unwrapped products. Furthermore, addition of chitosan nanoparticles increased the antimicrobial effect (Shapi'i, Othman, Nordin, Basha, & Naim, 2020). Starch from cowpea containing maqui berry (*Aristotelia chilensis*) extract showed antioxidant effect and slowed down lipid oxidation of salmon compared to neat starch film (Baek, Kim, & Song, 2019). Using cassava starch combined with EVOH and alginate resulted in a film with equal performance to PVC film in a storage trial with minimally processed lettuce (*Lactuca sativa* L.), with a shelf life of about 4 days at 6 °C (Brandelero, Brandelero, & de Almeida, 2016). However, the mass loss was eight times higher using the biodegradable film rather than PVC. Nisa et al. (2015) showed a decrease in lipid oxidation and reduced discoloration of fresh beef using active starch-based films containing green tea extract.

Shelf life of hake (*Merluccius* spp.) fillets was increased from 7 to 21 days when wrapped with alginate films containing a mixture of EOs and citrus extract in combination with ozonation with no change in pH and color of the fish fillets during storage time at 4 °C (Shankar, Danneels, & Lacroix, 2019). However, a further extension in the shelf life to 28 days was observed when ozonation treatment was replaced with gamma irradiation. It was suggested that a desired shelf life of the fish fillets could be achieved by the optimization of the combined treatment dose. Alginate films containing carvacrol (CAR) microencapsulated by  $\beta$ -CD were used to extend the shelf life of white mushrooms against *Trichoderma* spp. (Cheng et al., 2019). The controlled release of CAR from the packaging film was responsible for the enhanced antifungal activity. Moreover, the senescence of the white mushrooms was delayed due to the increased activities of active free-radical scavenging enzymes of the  $\beta$ -CD–CAR/sodium alginate (SA) films to alleviate oxidative damage. CMC in combination with carrageenan and grapefruit seed extract has been reported to completely inactivate foodborne pathogenic bacteria, *Listeria monocytogenes* and *Escherichia coli*, within 3 to 9 hr of application (Shankar & Rhim, 2018). A gelatine–alginate film containing 75% fish gelatine and 25%

alginate containing oregano EO was developed to increase the shelf life of rainbow trout (*Oncorhynchus mykiss*) (Kazemi & Rezaei, 2015). Strong antimicrobial effect of oregano oil and restricted access of oxygen by the films delayed the bacterial growth by 15 days during storage at 7 °C. Sodium alginate–clay containing marjoram EO films was reported to reduce *L. monocytogenes*, aerobic mesophilic, and psychrophilic cell counts when applied to trout slices (Alboofetileh, Rezaei, Hosseini, & Abdollahi, 2016). Rezaei and Shahbazi (2018) incorporated *Ziziphora clinopodioides* EO (0% and 0.5%), apple peel extract (0% and 1%), and ZnO nanoparticles (0% and 0.5%) into sauced silver carp fillet using three techniques including direct addition, edible coating, and composite film based on SA/CMC to increase the shelf life (microbial, chemical, and sensory properties) and inhibit the growth of *L. monocytogenes* during refrigerated storage over a period of 2 weeks. It was concluded that the active coating and film could be used to extend the shelf life and maintain the quality of fresh fish fillets without adverse effects on the organoleptic properties.

As for other reviewed biomaterials, few studies are available on pristine chitosan films used in food packaging trials. Among them, Priyadarshi, Sauraj Kumar, and Negi (2018) investigated the effect on the shelf life of green chilis packaged in chitosan pouches, both a neat chitosan film and a chemically modified film crosslinked with citric acid. Compared to unpacked products, the chilis packaged in the chitosan material exhibited lower moisture loss and better color preservation, the overall quality being slightly improved in the crosslinked film. In another study, the performance of neat chitosan versus modified chitosan films incorporated with apricot kernel oil was studied for bread slices packaged in pouches during 10 days of storage at room temperature. The control was packaged in a LDPE pouch. The study reports lower growth of molds on the bread packaged with chitosan films, both with and without active compound, as compared to LDPE, and a longer shelf life. The effect was most pronounced for the bread slices packaged in the active films. The findings were attributed to antifungal effect of both chitosan and apricot kernel oil (Priyadarshi, Sauraj Kumar, Deeba, et al., 2018). Furthermore, several studies have been published on modified chitosan films. Among them, the less complex film compositions involve incorporation of EO or other active compounds into the chitosan films followed by solvent casting. For instance, this includes the use of kombucha tea in films for packaging of minced beef (Ashrafi, Jokar, & Mohammadi Nafchi, 2018), ginger oil in films for packaging of barracuda fish (Remya et al., 2016), and cinnamaldehyde in chitosan/starch films for packaging of strawberries (Wang et al., 2019). Looking into more complex bionanocomposite films, these involve incorporation of nanoparticles

in combination with one or more active compounds. An example of such is chitosan/MMT/rosemary EO films used for packaging of poultry meat (Souza et al., 2019). Lin, Liao, Surendhiran, and Cui (2018) studied electrospun chitosan/ε-polylysine nanofibers and performed shelf life studies on chicken fillets. Based on the findings in available literature, it is evident that the use of antimicrobial compounds in the films allows for a broader use of chitosan as a food packaging material.

Pristine protein-based films may not perform well enough to replace conventional packaging materials; however, by addition of active compounds, some of the protein films reviewed hereafter exhibit a performability exceeding that of the conventional polymer controls. Gelatine films incorporated with catechin–lysozyme were used for packaging of minced pork, with the control samples being packaged in PVC films. Analyses at selected sampling times during cold storage revealed that lipid oxidation and microbial growth were retarded for samples packaged in the active gelatine films, resulting in extended shelf life as compared to the control samples (Kaewprachu, Osako, Benjakul, & Rawdkuen, 2015). In a study by the same group, the effect of packaging Bluefin tuna slices in fish myofibrillar protein films incorporated with catechin–Kradon extract, with reported antimicrobial and antioxidant properties, was examined. The control samples were packaged in LDPE films. The active films performed better than the LDPE films in delaying lipid oxidation besides resulting in significantly lower total viable counts (TVC) and better preservation of the red color of the tuna, an important quality trait (Kaewprachu, Osako, Benjakul, Suthiluk, & Rawdkuen, 2017). Zein films incorporated with pomegranate peel extract (PPE) were found to retard lipid oxidation and microbial spoilage of the cheese. Interestingly, the study reports that the PPE in the films did not significantly alter the sensory profile of the cheese (Mushtaq, Gani, Gani, Punoo, & Masoodi, 2018). Ejaz, Arfat, Mulla, and Ahmed (2018) studied the effect of gelatine films incorporated with clove EO and ZnO nanorods for packaging of peeled shrimps inoculated with bacterial suspension of *L. monocytogenes* and *Salmonella* Typhimurium. The bionanocomposite films could be heat sealed due to the thermoreversible gelling properties of gelatine. The film composition with 2% ZnO rods and 50% clove EO (w/w) in the gelatine matrix was found to provide a complete bacterial inhibition after 7 days of cold storage. For the control, neat bovine skin gelatine films, TVC was significantly higher throughout storage, demonstrating the pronounced effect of the active compound. Other studies in literature on protein-based bionanocomposite films include combinations such as soy protein/MMT/clove EO for packaging of Bluefin tuna fillets (Echeverría, López-Caballero, Gómez-Guillén, Mauri,



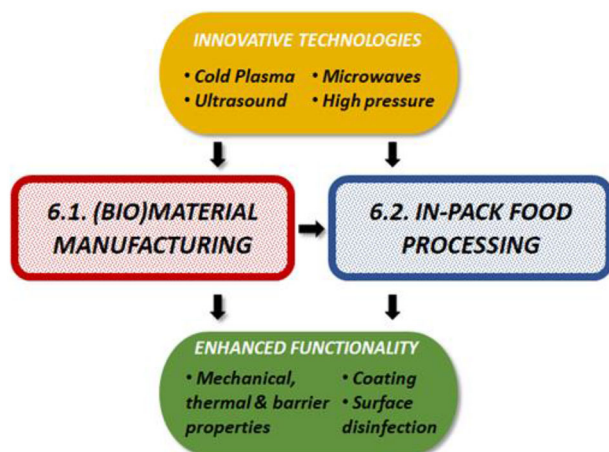


FIGURE 3 Intervention points for innovative technologies across the packaging supply chain

& Montero, 2018), gelatine/nanoclay/ethanolic extract of coconut husk for packaging of mackerel meat powder (Nagarajan, Benjakul, Prodpran, & Songtipya, 2015), and WPI/cellulose nanofiber/TiO<sub>2</sub> nanoparticles/rosemary EO for packaging of lamb meat (Sani, Ehsani, & Hashemi, 2017). Main observed effects on product quality and shelf life are listed in Table 3.

## 6 | COUPLING INNOVATIVE FOOD PROCESSING TECHNOLOGIES AND PACKAGING STRATEGIES FOR EXTENDED SHELF LIFE

In the last decade, a considerable and rapidly expanding body of literature, framed by a few comprehensive state-of-the-art reviews (please refer to Kourkoutas, Chorianopoulos, Nisiotou, Valdramidis, & Karatzas, 2016; Zhang, Wang, Zeng, Han, & Brennan, 2019), has extensively documented mechanistic/operational insight and competitive advantages of forefront processing technologies toward microbiological food safety and quality. The purpose of this section though is to assess their impact on material performance and subsequent interactions with food counterparts during in-pack processing and/or storage, as well as their potential toward enhanced manufacture and functionality of packaging materials (Figure 3). Such intervention criteria have therefore narrowed down the selection of relevant technologies for further review (i.e., CP, HP, MW, and US), besides shaping the section outline. To the knowledge of the authors, just a few review articles (Guillard, Mauricio-Iglesias, & Gontard, 2010; Morris, Brody, & Wicker, 2007) address comprehensively packaging implications of novel food processing technologies. In addition, biobased packaging systems have scarcely been studied in

relation to emerging processing; thus, conventional polymers, biomaterials, and composites will be jointly covered in a technology-dependent manner (5-year framework).

### 6.1 | Role of innovative technologies in production and safety of packaging materials

Besides food processing, innovative technologies have typically been integrated in production of packaging materials, including bio/nanocomposites, toward process optimization and enhanced material performance.

Otherwise referred to as the fourth state of matter, man-made plasma is typically sustained via an electric discharge in a gas subset; the partially or fully ionized gas assembles a bunch of subatomic and molecular entities (reactive oxygen and nitrogen species [RONS]) besides quanta of electromagnetic radiation (UV photons and visible light), all coexisting as nonplasma or thermal plasma upon thermodynamic equilibrium (Ashok, Dwivedi, & Vijay, 2008; Conrads & Schmidt, 2000). Although its commercial exploitation largely relies on upgraded surface features in advanced materials and a variety of usage domains in electronics, textiles, glass, or paper, CP induced at atmospheric pressure and room temperature has rapidly emerged as a value-added, niche opportunity for biobased applications (Ekezie, Sun, & Cheng, 2017).

Comprehensive reviews on CP (Ekezie et al., 2017; Hati, 2018; Pahwa & Kumar, 2018; Pan, Cheng, & Sun, 2019; Pankaj & Keener, 2017; Tolouie, Mohammadifar, Ghomi, & Hashemi, 2018; Zhang, Sang, et al., 2018) have outlined its multipurpose applications in material science, for example, surface ablation, cleaning and sterilization, surface activation (enhanced surface energy), etching, functionalization (improved adhesiveness, printability, wettability, sealing, and veneering), and enhanced bulk properties via chemical crosslinking and high-barrier multilayer depositional coating/grafting. Potential on conventional polymers, biobased films, and active and intelligent packaging systems has also been demonstrated. Such a broad “application–material” combination range is illustrated hereafter and in Section 6.2.

Sensitivity to water of fish protein films, one of their main limitations toward food packaging, has been reduced through CP-induced cleaning and etching (Romani et al., 2019). However, a bunch of literature has reported increased hydrophilicity of CP-modified substrates, for example, PA/PE, PET (more efficient than UV), and PVA thin films (Bauer et al., 2017; Kiruthika, Nivetha, & Shanmugavelayutham, 2019; Paneru et al., 2019), besides improved surface adhesion and deposition (e.g., antimicrobial, antioxidant, oxygen barrier, or biodegradable

coating and grafting), due to new oxygen-containing polar functional groups (higher surface polarity and lower water contact angle) and increased surface roughness: chitosan and chitosan/ZnO nanocomposite onto LDPE (Al-Naamani, Dobretsov, & Dutta, 2016); thymol onto PP carbonate (Bahramian, Chrzanowski, Kondyurin, Thomas, & Dehghani, 2017); sodium octanoate and Auranta FV onto the inner PE layer of CP-treated PA/LDPE using beef gelatine as carrier and coating polymer (Clarke et al., 2017); hybrid organic-inorganic Cu composite thin films onto CP-modified polycarbonate and c-Si (De Vietro, Conte, Incoronato, Del Nobile, & Fracassi, 2017); bilayer PP/CMC films with *Zataria multiflora* EO (Honarvar et al., 2017); eugenol, grape seed oil, and rosehip seed oil onto CP-activated (more efficient than  $\gamma$ -radiation) cellulose/chitin substrate (Irimia et al., 2017); alginate and chitosan onto LDPE containing summer savory extract (Rahmani et al., 2017); starch onto CP-modified PET films (Wiącek, Jurak, Gozdecka, & Worzakowska, 2017); chitosan (and DPPC) onto PET and PS films (Jurak, Wiącek, Mrocicka, & Łopucki, 2017; Suganya, Shanmugvelayutham, & Hidalgo-Carrillo, 2018); WPI onto PET, nylon film, and LDPE films (Joo et al., 2018); nisin onto PLA (Hu et al., 2018); nanofibrillated cellulose and nisin onto CP-treated biaxially oriented polypropylene (BOPP)/LDPE (Lu, Guo, Xu, & Wu, 2018); clove, argan oil, and chitosan onto PLA (Munteanu et al., 2018); acryl-coated PP film (Vukušić Pavičić et al., 2018); and CP surface modification of thyme EO encapsulated in silk fibroin nanofibers (Lin, Liao, & Cui, 2019).

Overall, potential of CP surface modification alongside antimicrobial coating has been demonstrated for diverse substrates in the abovementioned articles, toward active packaging and extended product shelf life. Furthermore, CP-induced polymer surface disinfection has largely been reported in literature; for example, 3 to 4 log/cm<sup>2</sup> reductions (*Salmonella* Typhimurium, *S. aureus*, and *E. coli* O157:H7) and >2 log/cm<sup>2</sup> reductions (*S. aureus*, *L. monocytogenes*, and *E. coli*) were achieved in glass, PE, PP, nylon and paper foil, and on PA/PE beef packaging films (Bauer et al., 2017; Puligundla, Lee, & Mok, 2016). Next to this, potential of CP and CP-activated liquids for biofilm prevention and removal has extensively been assessed on diverse food contact/processing surfaces (please refer to Flynn & Gilmore, 2018; Gilmore et al., 2018; Gupta & Ayan, 2019). Furthermore, solution plasma process (or sputtering) has been proposed for synthesis of metal nanoparticles and metal-polymer composites with a broad spectrum of antimicrobial activity, for example, Ag/polyacrylonitrile hybrid fibers and alginate/gold and alginate/silver bio-nanocomposites (Nam, MubarakAli, & Kim, 2016; Shi et al., 2011; Watthanaphanit, Panomsuwan, & Saito, 2013); for alginate depolymerization, due to enhanced bioactivity of low-molecular-weight forms (Watthanaphanit & Saito,

2013); to confer biocidal properties to alginate gels against pathogenic biofilms (Poor, Ercan, Yost, Brooks, & Joshi, 2014); and to develop alginate hydrogels with capacity for sustained release of RONS and cytotoxic potential (Labay, Hamouda, Tampieri, Ginebra, & Canal, 2019).

MW process, which refers to electromagnetic waves with a frequency of 300 MHz to 3000 GHz and a wavelength of 0.1 mm to 1.0 m, has been widely employed in materials processing, polymer synthesis, energy industry, separation process, food industry, nanomaterials synthesis, and so on (Huang, Tang, Zeng, & Xu, 2020). MW process can cause selective molecular vibration while transferring energy to the materials. Compared to conventional methods, MW method offers shorter reaction time, precise process control, and homogenous heat up of the reaction solution. Due to the ability to heat up the reaction solution homogeneously, this process enables uniform nucleation and rapid crystal growth, which allows formation of crystallites in polymers with narrow size distribution. In recent years, MW irradiation has been widely used as a powerful tool for rapid and efficient synthesis of different biopolymers. MW heating process has also been reported as a faster and efficient method for biomass extraction, hydrolysis, and pyrolysis process with improved product quality (Yuan & Macquarrie, 2015).

Electromagnetic radiation created during MW irradiation enhances structural stability of polymeric chains that increases thermal stability, mechanical properties, migration ability, and gas permeability properties (oxygen and nitrogen) (Wang et al., 2016; Yang et al., 2017; Zhong et al., 2019). MW radiation can efficiently alter starch molecule structure and change crystallinity and morphology of the polymer (Zhong et al., 2019). Moreover, MW treatment can improve the physicochemical properties of starch, such as water absorption ability, gelatinization temperature, swelling ability, and so on, which are considered important for efficient food packaging application (Zhong et al., 2019). Lin, Zhou, et al. (2019) studied the preparation and physicochemical properties of hydroxypropylated starch under MW assistance. They reported that the single chemical modification takes a very long time to introduce the hydroxypropyl group to the starch derivatives as compared to the MW process. They also reported that MW radiation not only reduced the reaction time, but also significantly improved the product quality. El Knidri, El Khalfaouy, Laajeb, Addaou, and Lahsini (2016) and Sebastian, Rouissi, Brar, Hegde, and Verma (2019) reported the extraction of chitosan and chitin from, respectively, shrimp shell waste and fungal biomass using MW irradiation with huge reduction in extraction time and high degree of deacetylation in few minutes. It has been reported that the MW-assisted polymerization process of lactic acid to produce PLA is 100 times faster than the convention heating method

(Bakibaev et al., 2015; Singla, Mehta, Berek, & Upadhyay, 2014). However, the MW radiation did not cause any significant improvement in the mechanical, thermal, and optical properties of PLA.

Xing, Zhang, Ju, and Yang (2006) also reported shortened reaction time during preparation of esterified starch molecules. Moreover, it has also been suggested that the polar hydroxyl group on the starch molecules absorbs the MW energy and rotates and moves in the electric field, resulting in chemical bond cleavage (Yao, Li, Liu, Wu, & Jin, 2016). Yang et al. (2017) treated the waxy maize starch with MW irradiation and reported that MW treatment can destroy the crystal structure of the starch and thereby increase the contact area between the surface of the particles and the reagent.

Graft copolymerization is a well-known technique to enhance specific properties of natural polymers without affecting its intrinsic properties (Shavandi & Ali, 2019). MW-assisted grafting method can produce faster free radicals to initiate and propagate the grafting reaction more effectively and thus results in higher polymer yield with better physicochemical properties as compared to the conventional grafting method where thermal heating is utilized. Several researchers have reported MW-assisted graft copolymerization of different natural polymers such as alginate, chitosan, starch, cellulose, and so on with a very precise control over the graft polymer, high yield rate, and with superior properties (Akin & Işıklan, 2016; Huacai, Wan, & Dengke, 2006; Singh, Tiwari, Pandey, & Singh, 2006).

Wang et al. (2016) developed MW-modified soy protein isolate and zein blend edible films for food packaging application. MW-modified films showed 5% to 25% increased breaking strength as compared to native films. It was suggested that the improved mechanical properties were due to increased  $\beta$ -sheet content and decreased  $\alpha$ -helix,  $\beta$ -turn, and random coil contents in the blended films caused the MW treatment. The surface of MW-modified films was more homogenous and showed fewer pores. Moreover, significant improvement in glass transition temperature and melting temperature was observed in MW-modified films. The improved thermal properties were due to change in molecular structure of protein films caused by MW treatment, driving hydrogen bonds, and hydroxyl groups to form a more compact structure.

Within the food packaging area, research on nonthermal HPP application has been mainly focused on the compatibility of this technology with existing or new packaging materials, where HPP is applied to the formed film rather than the film-forming solutions. Nonetheless, few studies have shown that HPP studies have shown that HPP can alter and improve the functionality of treated

biopolymer-based solutions and gelled systems prior to forming food packaging films (Montero, Fernández-Díaz, & Gómez-Guillén, 2002).

For protein dispersions, HPP may lead to structural changes by promoting protein unfolding, increasing protein surface hydrophobicity and free SH content, and thus increasing the degree of crosslinking and thereby obtaining denser and more uniform films with improved mechanical properties, lower water solubility, and WVP of amaranth protein films (Condés, Añón, & Mauri, 2015). Similarly, formation of new hydrogen bonds and shorter critical helix lengths endorsed increased resistance to the diffusion of water molecules in HPP-treated pigskin-derived, gelatine film at the optimal combination of pressure, holding time, and temperature (600 MPa for 30 min, at 20.5 °C) (Molinero et al., 2015). HPP treatment of PVA/chitosan films combined with 0.1% nano-TiO<sub>2</sub> enhanced the interaction among PVA, chitosan, and TiO<sub>2</sub> nanoparticles, which achieved higher film density and homogeneity especially at 600 MPa, thus providing improved mechanical and barrier properties (Lian, Zhang, & Zhao, 2016). HPP decreased molecular chain mobility of PLA chains and tightened the network structure in PLA/silver nanocomposite films enhancing the stiffness and tortuosity and preventing water vapor or oxygen transfer and migration of nano-Ag from the films (Chi et al., 2018).

Reducing sizes of biopolymer aggregates/particles to improve uniformity in film matrix might be a key element in developing biodegradable thermoplastic polymer or biodegradable particulate filler for food packaging. To this end, potential of high-pressure homogenization has been investigated in recent years toward utilization of plant-based by-products such as starch (Fu, Wang, Li, Wei, & Adhikari, 2011). High-pressure homogenization also produced films with better moisture barrier property, film transparency, and higher tensile strength when compared to ultrasound (US) and irradiation applied to potato peel solutions (Kang & Min, 2010).

Ultrasonication is a high-energy-based technology that operates in the region of 20 kHz to 1 MHz and used to disperse the reactants and initiate as well as accelerates reactions through collisions (Bera & Mondal, 2019). Sonochemical reactions are independent of acidity, basicity, and dipole moment. Nowadays, sonication is also widely used for the fabrication, surface modification, and dispersion of nanomaterials such as Au, Ag, Pt, and so on, with various sizes, shapes, and chemical and physical properties (Bera & Mondal, 2019). US treatment can significantly increase the tensile strength and puncture strength of protein films as compared to the untreated films (Cruz-Díaz, Cobos, Fernández-Valle, Díaz, & Cambero, 2019). These improvements are mainly attributed to the increment of the

molecular order of the polymer chain due to US treatment and to a more rigid structure (Cruz-Diaz et al., 2019). Moreover, it has also been mentioned by several researchers that US treatment could produce an increase in the exposition of hydrophilic and hydrophobic groups by unfolding the protein chains; exposed groups could form new bonds resulting in better mechanical properties of the films (Yilmaz, Turhan, Saricaoglu, & Tural, 2020). Liu, Wang, Lan, and Qin (2019) reported that US can significantly improve solubility, thermal performance, barrier properties, and bacteriostatic activity of PVA–tea polyphenol composite films. It has been suggested that sonication can loosen the polymeric structure and promote release of active substances that could be a possible reason for improved antimicrobial properties (Zhang, Wang, Ma, et al., 2019). Mechanical and structural properties of chitosan can be altered through controlled sonication time and mixing between sonicated and nonsonicated components due to the altered viscoelasticity and morphological aspect of the mixture (Gomes et al., 2016). Cruz-Diaz et al. reported that WVP of US-treated edible films made from whey protein was lower than the heat-treated ones due to better distribution of WPC lipids in the film (Cruz-Diaz et al., 2019). US treatment can also improve emulsifying properties of whey protein resulting in better lipid distribution within the films. Moreover, polymeric films with denser film networks generally exhibit lower WVP and US treatment can significantly increase the density of the films as compared to the heat-treated films (Schmid & Müller, 2019). Selected examples on the implementation of innovative technologies for (bio)material production are illustrated in Table 4, with special focus on their quantitative impact on (bio)material functionality and safety.

## 6.2 | Impact of innovative food processing on package performance and product shelf life

This subsection addresses the impact of innovative “in-pack” processing and/or subsequent storage on material performance indicators and eventually food safety and stability.

CP exerts varying effects on structural, mechanical, thermal, and barrier properties of packaging materials, with power, holding time, carrier gas, and polymer nature standing as the most influential parameters on surface modification: no adverse effect on glass transition temperature, overall migration and oxygen/WVP, and improved thermal stability of PLA (Pankaj et al., 2014); structural changes and reduced barrier properties of PP due to etching and degradation (Vishnuvarthanan & Rajeswari, 2015); increased tensile strength, elongation, lightness,

printability (ink adhesion), moisture barrier properties, glass transition temperature and biodegradability of defatted soybean meal (DSM)-based edible film, and limited oxygen availability due to CP polymer crosslinking (Oh, Roh, & Min, 2016); no adverse effect on surface temperature, optical characteristics, tensile strength, and strain-induced deformation of glass, PE, PP, and nylon and paper foil (Puligundla et al., 2016); lower WVP and higher tensile strength of bilayer *Zataria multiflora* EO-coated PP/CMC films (Honarvar et al., 2017); increased tensile strength of alginate–chitosan/LDPE, with lower solubility for chitosan-based films (Rahmani et al., 2017); significant changes in elongation at break and crosslinking, but unchanged thermal stability and flexural properties of starch/PET films (Wiacek et al., 2017); improved oxygen barrier properties, tensile strength, and elongation at break of whey-protein-coated PET (Joo et al., 2018); satisfying mechanical properties and transparency of BOPP/LDPE films coated with nanofibrillated cellulose and nisin (Lu et al., 2018); antifogging and highly transparent properties of PVA thin films (Paneru et al., 2019); and decreased WVP and solubility of fish protein films Romani et al., 2019.

Besides surface sanitization and enabling of microbicide coatings/gels (Section 6.1), enhanced microbiological food safety and quality have also been recorded for CP-treated polymers: retarded lipid oxidation and reduced hardness of smoked salmon packaged in CP-treated DSM films during storage at 4 °C (Oh et al., 2016); unaffected color, lipid peroxidation, sarcoplasmic protein denaturation, nitrate/nitrite uptake, or myoglobin isoform distribution of CP-treated vacuum-packaged beef loins after 10 days followed by a 3-day aerobic storage at 3 °C (Bauer et al., 2017); and improved release of thyme EO (TO) after CP treatment of TO/silk fibroin nanofibers, leading to approximately 6.1 log/g reductions of *S. Typhimurium* in chicken and duck meat (Lin, Liao, et al., 2019). Furthermore, in-package CP technology, namely, the ability to generate CP inside a sealed package, and its combination with modified atmosphere packaging (MAP) have gained growing interest in recent years as a food surface decontamination technology (please refer to Ekezie et al., 2017; Misra, Yepez, Xu, & Keener, 2019).

Microwave-assisted thermal sterilization (MATS) is accepted by the FDA for commercial sterilization of prepacked homogeneous and nonhomogeneous foods. MATS has the potential to produce food with higher quality, flavor, and longer shelf life (Zhang, Tang, Rasco, Tang, & Sablani, 2016). The shorter processing time associated with MATS is mainly responsible for the better food quality based on reduced shrinkage, less loss of texture, and greater retention in fresh-like appearance. The gas barrier and morphological properties and free volume of the polymer films used in food packaging application can

**TABLE 4** Selected examples on the implementation of innovative technologies towards enhanced (bio)materials

Technology	Material composition/substrate	Processing conditions	Impact on material properties	Reference
Cold plasma	CP surface modification of PVA thin film (antifogging)	CP: Dielectric Barrier Discharge (DBD); Argon; 4.75 kV; 30 kHz (at 6.13% duty cycle); 10 min	↓Contact angle (29.6 to 14.5°), ↑hydrophilicity (polar functional group), ↑roughness (0.26 to 0.65 nm), ↑antifogging and transmittance (86%)	Paneru et al., 2019
	Grafting of eugenol and rosehip/ grape seed oil onto CP activated cellulose/chitin blend fibers	CP: Glass reactor (50 Pa vacuum); Air or N <sub>2</sub> (10 cm <sup>3</sup> /min); 1.3 MHz; 30 W; 15 min γ-Irradiation dose: 5, 10, 20 kGy	Air CP (31.1% to 58.7% grafting) more efficient than γ-irradiation (9.7% to 22.8% grafting). Higher grafting degree for vegetable oils.	Irimia et al., 2017
	CP surface disinfection of glass, PE, PP, nylon and paper foil	CP: DBD; Room air; 10 kV (pulsed); 1.25 A; 35 kHz; 208 W; 10 min	>4 log/cm <sup>2</sup> ↓ for <i>E. coli</i> O157:H7 in 10 min; 3.0 to 3.5 log/cm <sup>2</sup> ↓ for <i>S. Typhimurium</i> and <i>S. aureus</i> No significant differences ( <i>p</i> > 0.05) in physicomechanical properties as compared to controls	Pulgundla et al., 2016
Microwave	Soy Protein Isolate and Zein Blended Film and Zein Blended Film	60 °C and 350 W for 2 min	↑Breaking strength by 5% to 25%, ↑glass transition temperature by 50%; ↑melting temperature by 3.5%	Wang et al., 2016
	Hydroxypropylated starch	150, 200, 250, 300, 350, and 400 W; 6 min	↑Transparency (4% to 8.81%); ↑the volume ratio of the condensing volume (21.11% to 33.11%); ↑hydrophilic (0.73 to 2.46 g/g), ↑hydrophobicity (0.67 to 1.01 g/g); ↓water evolution rate (46.91% to 36.16%)	Lin et al., 2019
	Waxy maize starch	1,600 W (160 W/g) for 5 min; 2,450 MHz frequency and repeated seven times	↓Molecular weight (21 × 10 <sup>7</sup> to 1.82 × 10 <sup>7</sup> g/mol) and the relative crystallinity (19.58 to 2.91%); ↑onset temperatures (69.90 to 75 °C); ↓DH (13.40 to 10.20 J/g) and viscosities	Yang et al., 2017
Ultrasound	US treatment of whey protein edible films	35 kHz ultrasound Bath; power 820 W (50% power setting); 20.8 W ultrasonic wave; 60 min	↑Firmness 3.7 to 5.7 N (US 10 min); after storage for 15 days, total number of colonies for control, US5min, US10min, and US15min was 6.72, 5.98, 5.28, and 5.14 log CFU/g; mold and yeast for control, US5min, US10min, and US15min was 4.77, 4.17, 3.85, and 3.69 log CFU/g; ↓ascorbic acid content by 49.55% for control, 41.14% for US5min, 32.83% for US10min, and 44.24% for US15min. ↓water vapor permeability (0.1 to 0.75 gm mm K Pa <sup>-1</sup> hr <sup>-1</sup> m <sup>2</sup> ); ↑tensile strength (0.7 to 0.9 Mpa); ↑puncture strength (0.5 to 1.2 MPa)	Cruz-Diaz et al., 2019

(Continues)

influence the shelf life of shelf-stable foods (Dhawan et al., 2014a). However, the gas barrier properties in polymer packages may deteriorate under high-temperature and high-moisture processes. Zhang et al. (2016) studied effects of barrier properties (oxygen transmission rate [OTR] and

water vapor transmission rate) of packaging materials on shelf life of mashed potato model food processed by MATS. They reported that the gas barrier properties of the polymers films were less affected by MW-assisted processing compared to conventional retort process

TABLE 4 (Continued)

Technology	Material composition/substrate	Processing conditions	Impact on material properties	Reference
	Polyvinyl alcohol/tea polyphenol composite films	Ultrasonic frequency of 40 kHz and power of 50 W; 30 min	↑Bacteriostatic rate ( $95.5\% \pm 4.2\%$ ) and ( $91.8\% \pm 3.7\%$ ) against <i>Staphylococcus aureus</i> and <i>Escherichia coli</i> ; ↑swelling capacity ( $740.19\% \pm 64.67\%$ ) and ↓solubility ( $5.26\% \pm 1.31\%$ )	Liu et al., 2019
	Polyvinyl alcohol with graphene oxide and nano-zinc oxide films	Frequency of 40 kHz; power of 50 W; 30 and 60 min	↑Growth inhibition of <i>S. aureus</i> and <i>E. coli</i> to $99.59\% \pm 1.58\%$ and $98.89\% \pm 1.16\%$ (after 1 hr US treatment)	Zhang et al., 2019
High-pressure processing	Aqueous dispersions of amaranth protein isolate (5% w/w)	0.1 (control), 200, 400, and 600 MPa for 5 min, at 6.5 MPa/s followed by pressure release at 20 MPa/s, with the adiabatic heating up to 33.5 °C; a mixture of propylene glycol and water (30:70) as pressure-transmitting medium	Up to 165% ↑tensile strength; up to 74% ↑elastic modulus Up to 42% ↓water solubility; 43% ↓water vapor permeability ↓Contact angle (<50°) indicating ↑hydrophilicity	Condés et al., 2015
	1:1 mixture of PVA and chitosan solution, with TiO <sub>2</sub> nanoparticles at 0.10% (w/v)	200, 400, and 600 MPa for 15 min at room temperature ( $23 \pm 2$ °C), with water as the pressure medium	Up to 38% ↓film thickness and 6% ↓density (especially at 400 MPa) Up to 20% ↓water vapor permeability; up to 7% ↓oxygen permeability 4.78 to 5.65% ↑elongation at break; up to ca. 100% ↑tensile strength Up to 28% ↓migration ratio of TiO <sub>2</sub> nanoparticles after 11 hr	Lian et al., 2016
	PLA film-forming solution with Silver nanoparticles (AgNPs) at 3 wt.% or 5 wt.%	0, 200, and 400 MPa for 15 min at room temperature, with water as the pressure transmitting fluid	Up to 51.5% ↓water vapor permeability Up to 20% ↑tensile strength; up to 78% ↑elongation at break Up to 18% ↓migration level at 40 days of storage in film treated at 200 MPa	Chi et al., 2018

due to morphological and free volume change resulted from the MW process.

Pressure-assisted thermal pasteurization (PATP) and sterilization (PATS) extend use of HPP by taking advantage of self-generated compression heating to achieve the lethality of conventional thermal treatments, thus providing safe foods and superior quality in a shorter process with lower maximum temperatures (Barbosa-Cánovas, Medina-Meza, Candoğan, & Bermúdez-Aguirre, 2014). Both processes, especially PATS with high temperature, can cause structural and morphological alternation in the packaging film including increased crystallinity, aging of amorphous phase, delamination, and rubbery to glassy state transition, thus compromising mechanical and functional properties of, for example, PATS-treated EVOH films (Dhawan et al., 2014b), PLA films (Sansone et al., 2012), PATP-treated PP and PE films (Filimon, Borda, Alexe, & Stoica, 2016), and LDPE films (Yoo, Lee, Holloman,

& Pascall, 2009), besides affecting product quality of in-packaged foods such as color (Ayvaz et al., 2012). An extensive review by Mensitieri, Scherillo, and Iannace (2013) discusses both reversible and irreversible effects on polymer films, whereas requirements for flexibility, dimensional stability, head space, and heat transfer properties of packaging materials are also well dealt with by Marangoni Júnior, Cristianini, Padula, and Anjos (2019).

Overall, the extent of PATP/PATS-induced changes on materials may depend on the treatment temperature rather than applied pressure intensity. For example, PATP showed no significant effect on structure and morphology of treated PLA films and their thermodynamics and functional properties, compared to the detrimental effects observed following PATS (Sansone et al., 2012). The migration behavior of gluten/MMT after 800 MPa for 5 min at 40 °C showed no significant effects, whereas the film did not withstand treatment at 115 °C (Mauricio-Iglesias,

Peyron, Guillard, & Gontard, 2010). Furthermore, increasing HPP temperature may affect the rate of migration during subsequent storage, as observed for the antioxidant, Irganox 1076, in HPP-treated PP (Caner & Harte, 2005). Additionally, the extent of PATP- and PATS-induced changes differs between the type of materials subjected to the treatment. PATS affected the barrier properties of vapor-deposited oxide and nylon containing films, but not those of aluminum foil and polyvinylidene dichloride (PVDC)-MA containing films (Bull, Steele, Kelly, Olivier, & Chapman, 2010). No change in the mechanical and thermal properties of PE was observed after PATP, whereas PP/silicon oxide (SiOx) pouches underwent significant modifications (Galotto et al., 2008). Further, only the foil-laminated packaging materials showed increased seal strengths when several plastic-laminated (nylon/coextruded EVOH, nylon/PP, PET/aluminum oxide/cast polypropylene (CPP), PET/PE) and aluminum foil-laminated (PET/Al/CPP and nylon/Al/PP) pouches with vacuum-sealed scrambled egg patties were subjected to PATS (Koutchma et al., 2010). The OTR of nylon 6 and nylon 6/nanocomposite films increased following 800 MPa for 10 min at 70 °C, but not that of nylon 6/EVOH (Halim, Pascall, Lee, & Finnigan, 2009). Lastly, the composition of in-packed foods and liquids can influence HPP impacts as seen in significantly lower OTR of LDPE films treated in contact with 95% ethanol compared to water (Yoo et al., 2009). Similarly, changes in the mechanical, thermal, and gas barrier properties of the HPP-treated PLA/SiOx/PLA films were smaller when in contact with olive oil than with water, which probably acted as a plasticizer (Galotto, Ulloa, Guarda, Gavara, & Miltz, 2009).

US is an eco-friendly and nonthermal method that is widely applied to inhibit microorganisms and improve quality and shelf life of food products during processing and preservation. Zhang et al. reported that the US treatment can inhibit the growth of total number of colonies, mold, and yeast in fresh-cut cucumber during storage (Fan, Zhang, & Jiang, 2019). Moreover, US treatment is reported to reduce the loss of weight, firmness, total soluble solids, total color change, water mobility, and maintained cell wall integrity in fresh-cut cucumber during storage. Hashemi (2018), São José (2015), Pinheiro, Alegria, Abreu, Goncalves, and Silva (2015), and Vivek, Subbarao, and Srivastava (2016) reported the positive effect of US treatment on safe and stable quality and physicochemical properties of tomato, fresh-cut kiwifruit, Mirabelle plum, watercress, parsley, and strawberry during storage. The US treatment is also used in food membrane processing for increasing the flux in dairy products (Córdova, Astudillo-Castro, Ruby-Figueroa, Valencia, & Soto, 2020). The US treatment can act as a complementary technique for enhancing the physical stability of in-packed

food emulsions (such as mayonnaise) without the need of expensive packaging materials (Gavahian, Chen, Mousavi Khaneghah, Barba, & Yang, 2018). Ding, Zhang, Ahmed, Liu, and Qin (2019) evaluated the effect of sonication on the performance of PVA/chitosan bilayer films during strawberry preservation. Improved physicochemical properties of the strawberry indicated that the US treatment of the films led to better maintenance of the food quality. A few representative examples of the quantitative impact of innovative technologies on biomaterial properties during food processing are summarized in Table 5.

## 7 | LITTERING AND WASTE MANAGEMENT

Waste is a major global issue causing negative impact on our environment. Waste management systems vary widely in different parts of the world. In Europe, more than 40% of all municipal waste was recycled or composted in 2016 (European Parliament, 2018). In some EU countries (northern part of Europe), recycling alongside incineration is most common while landfilling is almost non-existent. Incineration combined with landfill is most common in countries like France and UK, while landfilling remains the most common practice in the eastern and southern parts of Europe. In US, landfill or incineration account for the majority of waste management system and approximately 75% of plastic (from municipal solid waste) is deposited in landfill (United States Environmental Protection Agency, 2020). Energy recovery through incineration result in reduction of volume of waste by 90% and weight by 75% of waste as in the US by 90% and 75%, respectively (Gupta & Bais, 2020). However, incineration and landfill are also recognized as environmental threats. In EU this is recognized as the most harmful option for the environment, although one of the cheapest, and target for landfilling of municipal household waste should be less than 10% in 2035 (European Parliament, 2018).

Plastics has been recognized as a global environmental problem due to having negative impact on ecosystems and human health. Littering, plastic pollution, and marine debris have in recent years received increasing attention. According to the Ellen MacArthur Foundation, it is estimated that as much as 8 tons of plastic enter the ocean annually (Ellen MacArthur Foundation, 2016) and one study (Jambeck et al., 2015) also indicates that the amount will increase 10-fold from 2010 to 2025.

The global food packaging market has experienced massive growth over the last decades. Attention has in many cases been focused on optimization in packaging process for improved efficiency and reducing the thickness

TABLE 5 Selected examples on the impact of innovative technologies on biomaterial properties during food processing

Technology	Material composition/substrate	Processing conditions	Impact on biomaterial properties	Reference
Cold plasma	In-package CP treatment of PLA	CP: DBD; Room air; 50 Hz; 70 and 80 kV; 0.5, 1.5, 2.5, and 3.5 min	↑Roughness (0.9 to 20.9 nm); ↑thermal stability, i.e., initial (295 to 337 °C) and max. (316 to 365 °C) degradation T; no changes ( $p > 0.05$ ) in oxygen/water vapor permeability; increased (≈doubled) overall migration (below legal limits)	Pankaj et al., 2014
	CP treatment of defatted soybean meal-based edible film	CP: Low pressure (667 Pa) MW-powered; 2.45 GHz; 400 to 900 W; 1 L/min of O <sub>2</sub> , N <sub>2</sub> , dry air, He, or AR; 10 to 40 min	Tensile strength, elongation, and moisture barrier property of DSM films increased by 6.8%, 13.4%, and 24.4%, respectively, under optimal CP conditions (Ar, 15 min, 400 W)	Oh et al., 2016
	CP pre-treatment of multilayer PET films with whey protein isolate coating layer	Corona discharge: 20 kHz; 20 mL/min CP; N <sub>2</sub> (300 L/min); 1 kW; 7 kV; 2 ethylene-acrylic acid (EEA) primer coating solution (0.5 g); 6 hr at room temperature	12.8% ↓ in water contact angle (CP vs. untreated) 211 times ↓Oxygen transmission rate (PET/WPI/nylon/LLDPE) Tensile strength and elongation at break (PET/WPI/nylon/LLDPE): Up to 21.8% and 29.7% higher than corona discharge and EEA coating	Joo et al., 2018
Microwave	Multilayer polymeric films consisting of PET	MW heating section was heated simultaneously with MW energy infringing from the four cavities and by circulating hot water (35 psig, 122 °C) through convection/conduction surface heating	↑OTR by 12-fold; ↑WVTR by 11 times; ↑free volume fraction by 15%	Dhawan et al., 2014a
	Multilayered polymeric films	Single mode microwave-assisted thermal sterilization pilot system (10 kW, 915 MHz)	No significant effect on the ORT and WVTR	Zhang et al., 2016
	High-amylose maize starch	1.2 kW power	↑Relative crystallinity from 20% to 25% (after 1 min treatment); ↓relative crystallinity from 20% to 17% (after 4 min treatment); ↑digestibility from 59% to 70% (after 4 min treatment); ↑viscosity and digestive hydrolysis	Zhong et al., 2019

(Continues)



TABLE 5 (Continued)

Technology	Material composition/substrate	Processing conditions	Impact on biomaterial properties	Reference
Ultrasound	US treatment of modified atmosphere packed fresh cut cucumber.	Power level 400 W; intensity of 226 W/cm <sup>2</sup> ; 5, 10, and 15 min with pulse mode (10 s on:5 s off)	↑Firmness 3.7 to 5.7 N (US 10 min); after storage for 15 days, total number of colonies for control, US5min, US10min, and US15min was 6.72, 5.98, 5.28, and 5.14 log CFU/g; mold and yeasts for control, US5min, US10min, and US15min was 4.77, 4.17, 3.85, and 3.69 log CFU/g. ↓ascorbic acid content by 49.55% for control, 41.14% for US5min, 32.83% for US10min, and 44.24% for US15min.	Fan et al., 2019
	US treatment of watercress, parsley, and strawberry	Frequency 45 kHz; 10 min	↓aerobic mesophiles from 0.9 to 6.5 log CFU/g. 0.9 to 6.3 log CFU/g, and 0.7 to 4.0 log cycles, respectively, for watercress, parsley, and strawberries	São José, 2015
	US treatment on tomato	Frequency of 45 kHz; 10, 15, and 19 min	↑TPC by 10% (at 80%, 15 min); ↓firmness by 17% (at 55%, 10 min); ↓mesophilic load from 3.8 to 2.55 and 2.95 Log <sub>10</sub> CFU/g (at 80%, 15 min and 100%, 19 min)	Pinheiro et al., 2015
High-pressure processing	Multilayer polymer films containing EVOH	680 MPa for 5 min at 100 °C after preheating (90 °C)	Up to 358% ↑O <sub>2</sub> transmission rate; up to 64% ↓water vapor transmission rates Change in crystallinity and 23%↑/9%↓ free volume depending on film	Dhawan et al., 2014b
	Commercial PLA, <i>Biophan 121</i> with water	200, 500, and 700 MPa for 5 min, after preheating at 90 °C in water bath, reaching maximum 115 °C during treatment	11% to 18% ↓water vapor permeability at 25 to 30 °C, RH 30% to 90% Ca 8% ↓CO <sub>2</sub> permeability and 7% ↓O <sub>2</sub> permeability at 33 °C	Sansone et al., 2012
	Nylon/Ethylene-vinyl alcohol /Ethylene-vinyl acetate (EVA); Nylon/EVA; MetPET/PE containing carrots	600 MPa and 110 °C, for 10 min	Ca 10 cc/m <sup>2</sup> ·day ↑O <sub>2</sub> transmission rate for Nylon/EVA at 25 °C for 4 weeks; >60 cc/m <sup>2</sup> ·day↑ for MetPET/PE at 37 °C for 4 weeks Ca 1 gm/m <sup>2</sup> ·day ↑water vapor transmission rate for MetPET/PE at 25 and 37 °C for 4 weeks	Ayvaz et al., 2012

of materials (down-gauging), rather than technologies and processes designed for circular economy. Thus, handling plastic packaging of food as linear models rather than striving for more holistic, circular systems contributes to that a significant portion of plastic waste in many countries ends up in landfill or is incinerated (Hahladakis & Iacovidou, 2018; Luijsterburg & Goossens, 2014; Radusin et al., 2020). It is recognized that recycling is one solution to reduce the need for production of virgin materials in addition to diverting waste from landfill; however, only 9% of the plastic produced since 1950 have been recycled (Geyer, Jambeck, & Law, 2017).

Lately, global attention on plastics has led to reassessment of plastic value chains, and legislation related to single-use plastics and plastic bags will be or has been introduced in several countries (Ellen MacArthur Foundation, 2016). The approach and targets are to some extent different in different parts of the world; in Australia, the target is to phase out single-use plastic packaging, whereas in China use of nondegradable plastic bags will be reduced and in some municipalities banned within 2020 (IHS Markit, 2020). In other parts of Asia (in countries in Southeast Asia), packaging regulations are more fragmented and without a detailed approach toward packaging and packaging waste. Several countries have targets for recycling/recovery and reduction of waste to landfill and ban on plastic bags, whereas fewer have limitation related to single-use plastic (Krishnan, Subramaniam, & Baecher, 2019). The European Union has committed to implementing the UN Sustainable Development Goals, and in this context all plastic packaging on the European market must be either reused or recycled by 2030 (United Nations, 2015).

Challenges related to traditional plastic materials have triggered the use of biobased and biodegradable packaging materials. However, according to Ellen MacArthur Foundation (2016) the reassessment of plastic value chains should focus on recycling and reuse of plastic as the first priority, and in that context, they indicate that biodegradable plastics are not a good fit to meet these defined ambitions. However, they recommend biodegradable/compostable materials for targeted application such as garbage bags for organic waste and in closed systems (such as food packaging for events and canteens) where risk of mixing with the recycling stream is low. However, biodegradation and composting may also contribute to direct waste away from landfills and lower methane emission. Further, biodegradable plastics on the market today, for example, PLA, are typically only compostable under controlled conditions. In relation to challenges with traditional plastics and littering/ marine debris, biodegradable materials are therefore not necessarily a quick fix to this problem, as pointed out by Ellen MacArthur Foundation (2016).

In addition to organic recycling and energy recovery, bioplastic packaging waste may also be suitable for reuse and mechanical recycling. Environmental benefits are achieved when, for example, virgin PLA is replaced by PLA recycle (Maga, Hiebel, & Thonemann, 2019). However, a separate recycling stream for PLA does not exist (Wojnowska-Baryła, Kulikowska, & Bernat, 2020). Maga et al. (2019) concluded that higher volume PLA input in the waste stream would ease and improve the technologies, through optimized collection, sorting, and recycling processes, and thereby encourage establishment for of PLA recycling streams. In recycling of both bioplastics and conventional fossil-based plastics, highest outcome (yield and quality) is achieved when impurities are reduced.

However, some challenges might be related to the recycling and sorting process, for example, identifying different types of plastics. The effect of, for example, PLA in conventional fossil-based polymer depends on the type of polymer. According to Bioplastic Europe, mixing of PLA in PP (3% PLA in postconsumer recycle) and PS (10% in PS regranulates) does not negatively impact the quality of the recycled material (European Bioplastics, 2017). However, only 0.1% PLA in PET results in opacification of recycled PET, 0.3% causes yellowing, and 2% and 5% result in agglomeration (Wojnowska-Baryła et al., 2020). PLA and PET are both transparent and similar in visual appearance and difficult to separate by the consumers. Thus, identification or extra labeling is needed if the sorting takes place at consumer level, as well as education of the consumers. A study performed in Norway has shown that central sorting of municipal waste results in higher recycling rates compared to different home sorting systems and waste collecting points (Deloitte, 2019).

## 8 | LEGISLATION

Food contact materials (all materials and articles intended for food contact) applied in Europe must meet the requirements set by the Framework Regulation (EC) No 1935/2004. Specific rules for plastic materials are set in Regulation (EC) No 10/2011 (with amendments), with the authorized list (positive list) for starting substance (intentionally added substance) given in Annex I with defined specific migration limits. Biobased plastic materials are also subjected to these regulations and amendments may include specification regarding biobased materials, such as Regulation (EC) no 2019/1338.

Legislation related to downstream handling of packaging materials must also be fulfilled: EU waste legislation including Waste Framework Directive and the Packaging and Packaging Waste Directive 94/62/EC. The main

priority of this legislation is preventing production of packaging waste. It comprises principles for recusing, recycling, and recovering packaging waste, thereby reducing the final disposal of waste. In recent revision of EU waste legislation, new and ambitious targets related to, for example, landfill and recycling are described, in which biodegradation is included. Specific requirements are set in Annex II related to composting and biodegradable packaging. The nature of the biodegradable packaging waste shall be such that it is capable to decompose to CO<sub>2</sub>, biomass, and water by undergoing chemical, physical, thermal, or biological decomposition. Biodegradable plastics can also be recognized in relation to recycling—and organic recycling. If a material is certified as compostable, it must be biodegradable under specific conditions according to international standards such as EN 13432 (“Packaging: requirements for packaging recoverable through composting and biodegradation industrial composting of packaging”) (European Union, 2000) and/or EN 14995 (European Union, 2006). The standard EN 13432 sets requirements to, for example, disintegration: after 12 weeks at maximum 10% of the material fragments can be larger than 2 mm.

## 9 | CONCLUSION

In the last couple of decades, the development of biobased and biodegradable food packaging materials has experienced a massive boost due to increased environmental focus and willingness to focus the research agenda in the direction of increased sustainability and toward a circular bioeconomy. However, in the same timeframe the use of conventional plastics has also increased, and most of this increase is as packaging material. Commercial use of biobased and biodegradable food packaging materials is still low compared to conventional materials, due to several factors including technical and mechanical properties, cost, legislation, safety, and after-life handling. Because the main purpose of the packaging material is to protect the food it surrounds, the biobased solutions must at least perform as good as conventional plastics, leading to more complex biomaterials including incorporation of active substances, biocomposites, and nanosized particles. Such developments make the biomaterials more suited to be combined with food processing technologies and better protect the foodstuff with increased technical properties, but also increase the complexity and cost and make them more durable and less biodegradable. Biodegradable biomaterials can already today replace conventional materials for many foodstuffs; however, for other foods further development is needed, including those involving processing with increased temperature and humidity. Biomateri-

als might not be a quick fix to plastic littering or to decrease the amount of fossil-based plastics used in short term. They still need to be collected in the waste streams and separated from conventional materials for controlled composting or recycling. There is still a need for more research within this field; conventional plastics have a century head start, but biomaterials are closing the gap rapidly, with the development leading us closer to a fully biobased food packaging future.

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## AUTHOR CONTRIBUTIONS

All authors contributed in development of the concept of the manuscript, organization of topics and subsections, proof-reading, and editing. Julie Nilsen-Nygaard: Corresponding author, coordination, final revision, references, Abstract, Introduction, and Sections 3, 4, and 5. Estefanía Noriega Fernández: Introduction and Section 6. Tanja Radusin: Sections 2 and 5. Bjørn Tore Rotabakk: Section 5. Jawad Sarfraz: Sections 4 and 5. Nusrat Sharmin: Sections 2, 5, and 6. Morten Sivertsvik: Conclusion. Izumi Sone: Sections 3, 5, and 6. Marit Kvalvåg Pettersen: Introduction and Sections 7 and 8.

## CONFLICTS OF INTEREST

The authors declare no conflicts of interest.

## DISCLAIMER

The author Estefanía Noriega Fernández is currently employed with the European Food Safety Authority (EFSA) at the Nutrition Unit that provides scientific and administrative support to the NDA panel in the area of safety assessment of novel foods. However, the present article is published under the sole responsibility of the author/s and may not be considered as an EFSA scientific output. The positions and opinions presented in this article are those of the author/s alone and are not intended to represent the views/any official position or scientific works of EFSA. To know about the views or scientific outputs of EFSA, please consult its website under <http://www.efsa.europa.eu>.

## ORCID

Julie Nilsen-Nygaard  <https://orcid.org/0000-0002-8384-6727>

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