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Review

Bio-based active food packaging materials: Sustainable alternative to conventional petrochemical-based packaging materials

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ABSTRACT

In food industry, a growing concern is the use of suitable packaging material (i.e., biodegradable coatings and films) with enhanced thermal, mechanical and barrier characteristics to prevent from contamination and loss of foodstuff. Biobased polymer resources can be used for the development of biodegradable bioplastics. To achieve this goal, biopolymers should be economic, renewable and abundantly available. Bioplastic packaging materials based on renewable biomass could be used as sustainable alternative to petrochemically-originated plastic materials. This review summarizes the recent advancements in biopolymer-based coatings and films for active food packaging applications. Microbial polymers (PHA and PLA), wood-based polymers (cellulose, hemicellulose, starch & lignin), and protein-based polymers (gelatin, keratin, wheat gluten, soy protein and whey protein isolates) were among the materials most widely exploited for the development of smart packaging films. These biopolymers are able to synthesize coatings and films with good barrier properties against food borne pathogens and the transport of gases. Biobased reinforcements e.g., plant essential oils and natural additives to bioplastic films improve oxygen barrier, antibacterial and antifungal properties. To induce the desired functionality the simultaneous utilization of different synthetic and biobased polymers in the form of composites/blends is also an emerging area of research. Nanoscale reinforcements into bioplastic packaging have also been reported to improve packaging characteristics ultimately increasing food shelf life. The development of bioplastic/biocomposite and nanobiocomposites exhibits high potential to replace nonbiodegradable materials with characteristics comparable to fossil-based plastics, additionally, giving biodegradable and compostable characteristics. The idea of utilization of renewable biomass and the implications of biotechnology can firstly reduce the burden from fossil-resources, while secondly promoting biobased economy.

1. Introduction

Plastics are thought to be the most commonly used materials particularly in packaging applications. Currently, global production of plastics is about 320 million tons/year with an increasing demand which represent its broad-spectrum application in various fields (Paletta, Leal Filho, Balogun, Foschi, & Bonoli, 2019). Plastic industry is highly dependent on fossil resources therefore, increasing prices of natural gas and petroleum oil may influence the economy of the plastic market (www.european-bioplastics.org). For this purpose, commonly used polymers are polyvinylchloride (PVC), polyethylene terephthalate (PET), polypropylene (PP), polyethylene (PE), polyamide (PA), polystyrene (PS) and ethylene vinyl alcohol (EVOH) (Luzi, Torre, Kenny, & Puglia, 2019; Martínez-Abad, Lagaron, & Ocio, 2012). They are preferred because of their excellent barrier and mechanical properties, abundant availability and cost-effectiveness (Park, Koo, Cho, & Lyu, 2017). However, by the disposal of synthetic plastics, emission of greenhouse gases i.e., carbon dioxide and methane accumulation in the environment leads to major environmental consequences (Jain & Tiwari, 2015). Therefore, the use of fossil-based plastics is limited due to their non-recyclable or non-biodegradable nature. Therefore, it is very important to move towards the alternative raw materials for plastic production (Ahmed et al., 2018). This idea suggests the development and utilization of suitable, abundant and sustainable

Abbreviations: PHA, polyhydroxyalkanoates; PLA, polylactic acid; PHB, polyhydroxybutyrate; PHBV, 3-hydroxybutyrate co 3-hydroxyvalerate; BioPE, bio-polyethylene; CMC, carboxymethylcellulose; PUR, polyurethane; PF, phenol formaldehyde; LDPE, low-density polyethylene; TPS, thermoplastic starch; ASTM, American society for testing and materials; PVA, polyvinyl alcohol; PCL, polycaprolactone; PET, polyethylene terephthalate

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bioresources for the development of environmentally friendly alternatives to conventional resources (Bilal & Iqbal, 2018). The compostability is crucial for polymer resources because recycling requires energy (Al Hosni, Pittman, & Robson, 2019; Asgher, Arshad, Qamar, & Khalid, 2020a). Broadly speaking, compostability is a series of processes that exploit biodegradability of the organic matter to transform them into a defined product so-called "compost". Therefore, compostability allows degradation of the materials in soil by producing carbon dioxide, water and other inorganic compounds (Al Hosni et al., 2019).

Plastics produced by renewable resources are not necessarily biodegradable or compostable. The second one, biodegradable plastics do not necessarily base on renewable resources, because biodegradation is positively correlated to the chemical structure of the compound rather that its origin. Specifically, the type of chemical bonding present in molecules define the time period of biodegradation (Lambert & Wagner, 2017). Cellulose, starch, lignin, gelatin and keratin which are naturally carbon containing polymers are biodegradable or compostable whether obtained synthetically. At the same time, biopolymer-based bioplastics, can lose their biodegradability after chemical modification like polymerization, e.g. Nylon 9 obtained by the polymerization of oleic acid synthesized by the polymerization of castor oil monomers (Cotarca et al., 2001). Fig. 1 describes the different types of plastic from origin and biodegradability perspective. Plastics are chemical compounds synthesized by the polymers and numerous other chemicals e.g., additives, colorants, stabilizers, processing aids, etc. Optimization of each product must be studied with respect to its processing and future application (Matarneh, Sotnik, Deineko, & Lyashenko, 2018; Riza, Syaubari, Andriansyah, Dewi, & Ernita, 2019). Keeping in mind, synthesis of bioplastic using 100% renewable resources has not been realized. But the tendency is to use the maximum quantity of renewable resources is definitely possible (Asgher et al., 2020a). Until now bioplastic comprise more than 50% by weight of renewable resource. Several bioplastics are mixtures or blends comprising synthetic compounds, such additives and polymers, to boost up functional properties of final product to enhance applications (Marra, Silvestre, Duraccio, & Cimmino, 2016; Murphy & Collins, 2018; Saravanan, Leena, & Selvamurugan, 2016). If additives could be based on renewable resources, approximately a 100% biodegradable plastic can be made (Reinders, Onwezen, & Meeusen, 2017).

Bioplastics, like other fossil-based plastics, comprise a wide-range of applications e.g., collection bags, horticultures, agricultural foils, toys, nursery products, and textiles fibers (Marjadi, Dharaiya, & Ngo, 2010; Tortajada, Ferreira, & Prieto, 2013). For the packaging of edibles, expected performance from bioplastics is food quality preservation and protection from environment. Obviously to accomplish these functions is significant to control and alter their mechanical as well as barrier properties that consequently rely on the chemical structure of the packaging material. Additionally, it is important to consider the change in the characteristics of bioplastics during period of interaction with the food material (Jabeen, Majid, & Nayik, 2015; Jariyasakoolroj, Leelaphiwat, & Harnkarnsujarit, 2018). Literature study reveals that only a limited number of biopolymers are applied for food packaging applications. Unlike the common labels, films, wraps and laminates developed from fossil resources, the practice of biodegradable plastic material shows a right step towards the preservation of environment from hazardous substances. Important step is to understand not only the mechanical and physical properties of such compounds but also the compatibility with food material, that has been identified as a possible source of loss in the quality of food (Asgher, Urooj, Qamar, & Khalid, 2020b; Biscarat, Charmette, Sanchez, & Pochat-Bohatier, 2015). In this review, we have summarized the recent data related to biopolymers-based coatings and films for food packaging applications discussing the major sources, production methods, incorporation of active antioxidant/antimicrobial reinforcements, and their biodegradability and compostability perspectives.

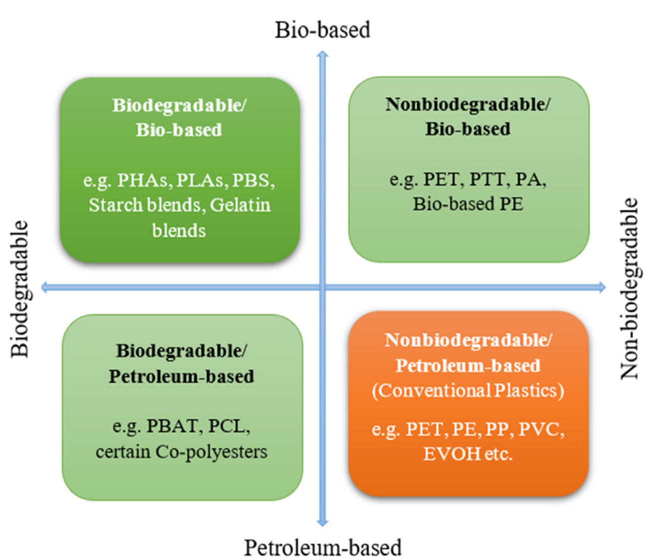

Fig. 1. Different types of plastics available in the market from origin and degradability point of view.

2. Biobased polymer resources

Polymers which are synthesized by the living body such as plants and microorganisms through metabolic engineering processes are called bio/natural polymers. Fig. 2 represents the general classification of biopolymers depending upon their origin (Iqbal, 2015). Carbohydrate polymers e.g. starch, chitosan, cellulose or lignin (Devlieghere, Vermeulen, & Debevere, 2004; Hassan, Chatha, Hussain, Zia, & Akhtar, 2018; Jiang, Duan, Zhu, Liu, & Yu, 2020; Priyadarshi & Rhim, 2020); proteins e.g. keratin, collagen or gelatin and polyhydroxyalkanoates e.g., polyhydroxybutyrate (PHB) and its copolymer 3-hydroxybutyrate co 3-hydroxyvalerate (PHBV) (Iqbal & Keshavarz, 2016; Iqbal, Rasheed, & Bilal, 2018). Lignocellulosic wood fibers contain high amount of cellulose and hemicelluloses. Their resulting films hold good toughness, tensile strength, high surface gloss and good transparency (Darni, Dewi, & Lismeri, 2017). Mostly, the cellulose is chemically altered during the procedure of dissolution to facilitate the breakage of polymer chains. Cellulose derivatives that are obtained after chains disintegration can be regenerate as coatings.

2.1. Microbially-originated polymers

Three major fermentation-based biopolymers are polylactic acid (PLA) polyhydroxyalkanoates (PHA) and exopolysaccharides (EPS) (Alshehrei, 2017; Asgher et al., 2020a; Chen, 2010). The monomers for the synthesis of PHA, PLA and BioPE can be manufactured from renewable biomaterials. Sugarcane and corn wastes are beneficent because their monomers exhibit minor degree of polymerization, hence, their extraction from plant source is easy.

2.1.1. Polylactic acid (PLA)

Bio-based lactide and lactic acid is synthesized by fermentation of sugars (commonly corn starch). Lignocellulosic biomass after hydrolysis looks an amazing pathway for lactide and lactic acid synthesis. Conventional thermoplastics have higher processing temperature as compared to PLA (de Kort, Bouvrie, Rastogi, & Wilsens, 2019). Purification of PLA requires electrodialysis, adsorption, solvent extraction, distillation and reverse osmosis (Huang & Ramaswamy, 2013). Cost of conversion processes can be reduced by increasing raw material, but this will enhance enzyme hydrolysis and fermentation expenses (Nampoothiri, Nair, & John, 2010). After fermentation, present proteins are eliminated by filtration processes. Bipolar electrodialysis leads towards bioconversion of lactate to sodium hydroxide and lactic acid.

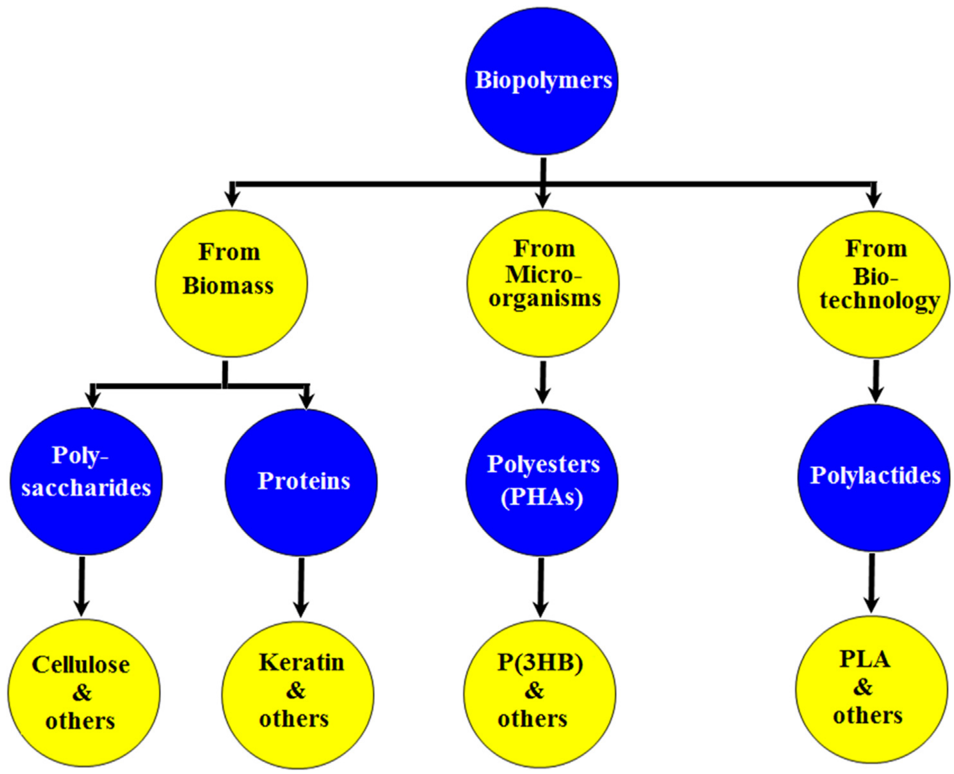

Fig. 2. General classifications of biopolymers (PHAs: polyhydroxyalkanoates, PHB: polyhydroxybutyrate, PHBV: 3-hydroxybutyrate co 3-hydroxyvalerate, PLA: polylactic acid). Reprinted with permission from, Iqbal (2015).

Purification of lactide is through vacuum distillation. Lactic acid condensation and polymerization is carried out to obtain PLA (Pal & Katiyar, 2017).

Recently, various PLA-based technologies have developed to achieve desired mechanical, chemical and biological characteristics comparable to petroleum-based polymers. The need for physical and/or chemical modifications of PLA to attain suitable characteristics for its desired consumer applications demanded high scientific attention in last few years (Moustafa, El Kissi, Abou-Kandil, Abdel-Aziz, & Dufresne, 2017; Nazrin et al., 2020; Niu, Liu, Song, Han, & Pan, 2018). The development of environmentally friendly or “green” materials by blending with other fibers and polymers, e.g., natural fiber reinforced polymer composites is an emerging area of research. In addition, the formation of PLA-based nanocomposites by variety of different nanostructures for biomedical and industrial biotechnological applications is a valuable approach not only towards induction of desired functionality but also help in the reduction of experimental cost (Bilal, Rasheed, Nabeel, & Iqbal, 2020; Nazrin et al., 2020).

2.1.2. Polyhydroxyalkanoates

Polyhydroxyalkanoates (PHAs) comprises a broad group of biobased polymers. PHAs can also be thermally transformed as PLA and are synthesized by renewable raw materials (such as fatty acids, maltose, glucose) via biotechnological conversion by action of different microorganisms (Kawaguchi, Hasunuma, Ogino, & Kondo, 2016). Relying on microorganism choice, carbon source, additives and conditions provided, polymers with different building blocks and their different properties are obtained (Keenan, Nakas, & Tanenbaum, 2006). Different types of PHAs have been produced including polyhydroxybutyrate and its copolymer 3-hydroxybutyrate co 3-hydroxyvalerate. Their applications range from packaging industries to medicinal implants and textile sectors. There has not been significant application of PHAs as bioplastics and possible cause could be the high production and recovery cost of PHAs. Scientists are searching for the replacement with cost effective feedstocks for PHA production. For example, the use of wood-based raw material containing hemicelluloses which can be used for the development of bacterial PHAs (Asgher et al., 2020a, 2020b; Silva, Tobella, Becerra, Godoy, & Martínez, 2007).

2.1.3. Exopolysaccharides

Exopolysaccharides (EPS) are complex biopolymers, mainly composed of carbohydrates which are synthesized by various microbial species including bacteria (both gram-positive and gram-negative), fungi and blue-green algae, and are secreted outside the cell wall (Asgher et al., 2020a; Vidhyalakshmi, Vallinachiyar, & Radhika, 2012). There are several types of EPS such as alginate, glucans, dextrin, xanthan, Levan, etc. Among other types of EPS, Kefiran is a favorable due to its water soluble and biodegradable nature (Piermaria, Pinotti, Garcia, & Abraham, 2009). It is obtained during the fermentation of milk in the kefir synthesis (Maeda, Zhu, Suzuki, Suzuki, & Kitamura, 2004; Micheli, Uccelletti, Palleschi, & Crescenzi, 1999). Kefiran has been studied for various beneficial characteristics for human health e.g., attractive healing and antimicrobial activity (Leite et al., 2013; Rodrigues, Caputo, Carvalho, Evangelista, & Schneedorf, 2005), anti-inflammatory potential (Radhouani, Gonçalves, Maia, Oliveira, & Reis, 2018), etc. Moreover, its applications are broadly studied in food industry as emulsifier, stabilizer, and gelling agent (Piermaria, Mariano, & Abraham, 2008). Kefiran-based films are gaining much scientific attention due to its novel characteristics, e.g., biocompatibility, biodegradability, safety, emulsifying and stabilizing effects and excellent water vapor permeability and mechanical characteristics (Júnior, Vieira, & Anjos, 2020; Moradi & Kalanpour, 2019). In addition, kefiran-based films bioplastics have excellent visual aspects and could be effectively synthesized using edible plasticizers e.g., glycerol (Ghasemlou, Khodaiyan, Oromiehie, & Yarmand, 2011; Hassan et al., 2019). Therefore, the development of kefiran-based films can lead to suitable and environmentally friendly coatings and packaging materials with improved properties.

2.2. Wood-based polymers

2.2.1. Celluloses and hemicellulose

Lignocellulosic wood fibers comprise cellulose around 40–50% and hemicelluloses 25–30% by weight of it. Cellulose films hold good toughness, tensile strength, high surface gloss and excellent transparency (Agustin, Ahmmad, Alonzo, & Patriana, 2014; Guzman-Puyol et al., 2019). Mostly, the cellulose is chemically altered during the method of dissolution to facilitate the breakage of the polymer chains. Cellulose derivatives that are obtained after chains disintegration, that can be regenerate as bioplastic films for packaging production and other barrier applications. Hemicelluloses are amorphous and multi-faceted heterogeneous polysaccharides, structurally less ordered and are the wood-based hydrophilic polysaccharides with minimum thermal resistance (Bilal and Iqbal 2020b, 2020a; Hosseinaei, Wang, Enayati, & Rials, 2012). Hemicellulosic-based films are brittle; however, plasticizer addition improves the flexibility, toughness as well as low oxygen permeability have been described (Mendes et al., 2017; Zhang, Xiao, Chen, Wei, & Liu, 2020). These films have interesting role in packaging applications due to low oxygen permeability. Both the softwood hemicelluloses and the glucuronoxylan (hardwood-hemicelluloses) have been reported in composites/blends with bioplasticizer to synthesize packaging material with higher oxygen permeability and flexibility (Hartman, Albertsson, Lindblad, & Sjöberg, 2006; Sailaja, 2006; Silva, Silva, & Lucia, 2015). Though, the films were vulnerable to aqueous uptake. The higher polymer content (alginate or CMC) improved moisture uptake resistance with increased mechanical strength by reducing film flexibility.

2.2.2. Starch

Starch is the most common plant-based polysaccharide for the development of bioplastic films because of its cost-effectiveness, abundance, and significant film-forming properties (Hassan et al., 2018; Mose & Maranga, 2011). Different starches from previously unexplored resources, have been reported alone or in blended form with other natural polymers as biodegradable coatings or packaging films to extend the product's shelf life (Fakhouri, Martelli, Caon, Velasco, & Mei, 2015; Ojogbo, Ogunsona, & Mekonnen, 2020; Thakur et al., 2019). Fakhouri et al. (2015) reported composite coating based on corn starch/gelatin plasticized with sorbitol, which improved the post-harvest storage of Red Crimson grapes. The resultant biocomposite of starch matrixed with gelatin enhanced solubility characteristics and improved the mechanical characteristics. Moreover, the decrease in water vapor permeability of composite film as compared with waxy corn starch and modified blends was also significantly observed. Nawab, Alam, and Hasnain (2017) used mango kernel starch as a packaging material to improve the shelf life of tomatoes. The packaging material efficiently delayed the ripening process of tomatoes which was studied by analyzing variety of physical and chemical parameters e.g., soluble solids concentration (SSC), weight loss, and tartaric acid (TA) content and the firmness of fruit. The modification of this exceptional biopolymer has found applications beyond packaging and bioplastic sectors, as its applications have expanded to utilize in biomedical, pharmaceutical, tissue engineering scaffolds, drug delivery, and even in metallurgical sectors for the development of porous media (Ogunsona, Ojogbo, & Mekonnen, 2018; Thakur et al., 2019).

2.2.3. Lignins

Lignocellulosic wood contains 10–25% by weight of lignin (Yang, Ching, & Chuah, 2019). By an estimation, current lignin synthesis in paper and pulp industries is 50–60 million tons/year of which only 2% is commercially used and 98% is wasted as low-cost fuel to burn in chemical recovery boiler (Bajwa, Pourhashem, Ullah, & Bajwa, 2019; Sahoo, Seydibeyoğlu, Mohanty, & Misra, 2011). The potential utilization of lignin-based biomass as a cost-effective reinforcement would be used in polyurethane preparations to replace fossil-based polymers and consequently improving mechanical and thermal characteristics of the polyurethane (Christopher, 2013). Lignin is capable to replace phenolic compounds in phenolformaldehyde (PF) resins synthesis. Lignin can be applied as biodegradable solution for the phenols in the petrochemical-based PF resins (Kouisni et al., 2011). Phenolated lignin-PF resins reported better physical and mechanical characteristics than pristine lignins (Cetin & Özmen, 2002). Recently, the synthesis and utilization of aldehydes from lignin biomass was reported as an excellent formaldehyde alternative (Foyer, Chanfi, Boutevin, Caillol, & David, 2016).

2.3. Protein-based polymers

2.3.1. Collagen and gelatin

Both collagen and gelatin are obtained from animal sources. Collagen is the most abundant protein in nature (Fratzl, 2008). In animal, it constitutes about 20–25% of total body mass. Its structure consists of three cross-linked α-chains while denatured collagen derivative is called gelatin, composed of many polypeptides and proteins. Collagen is rich in methionine, hydroxyproline/proline, and glycine amino acids (Shoulders & Raines, 2009). Collagen-based bioplastics are synthesized by the extrusion process and comprises various applications (Oechsle, Bugbee, Gibis, Kohlus, & Weiss, 2017), while films production using gelatin requires wet process by the formation of film forming solution. Collagen-based bioplastic films comprise good mechanical properties e.g., hydrolyzed collagen films have been reported to possess excellent tensile strength (Fadini et al., 2013). However, Gelatin films possess poor mechanical and barrier properties which shows its hydrophilic nature (Ciannamea, Castillo, Barbosa, & De Angelis, 2018).

2.3.2. Wheat gluten films

Bioplastic films can be synthesized by plasticized wheat gluten by intensive extrusion followed by compression and molding (Zubeldía, Ansorena, & Marcovich, 2015). Film formation involves the synthesis of hydrophobic, hydrogen and disulfide interactions. Disulfide bonds are stabilized by sulfhydryl groups (Sharma, Khatkar, Kaushik, Sharma, & Sharma, 2017). Heating cause the denaturation of polymer as well as breakage of native hydrophobic and disulfide groups. During drying, new disulfide bonds form by the oxidation of gluten again. Film clarity is dependent upon the purity of gluten mass and the casting medium used e.g., alkaline or acidic (Chiou et al., 2020). Films produced exhibit homogeneity, excellent gas barrier characteristics and the mechanical strength (Mojumdar, Moresoli, Simon, & Legge, 2011).

2.3.3. Soy protein film

Soy proteins are also exploited for the synthesis of bioplastic films for packaging applications. Films based on soy proteins are smoother, transparent, flexible and cost-effective than other protein-based bioplastics (Otoni, Avena-Bustillos, Olsen, Bilbao-Sáinz, & McHugh, 2016). Moreover, they exhibit good oxygen barrier properties under low moisture conditions (Denavi et al., 2009). Major disadvantage of using these films are low mechanical strength, lack of heat stability and allergenicity as compared with low density polyethylene (LDPE). Soy protein isolates aqueous solution also have film formation abilities but on stainless steel plates at a high temperature. Film formation using other protein-based raw materials e.g. pea protein, canola protein, pumpkin oil cake, pistachio globulin protein etc. have also been reported by several scientists (Acquah, Zhang, Dubé, & Udenigwe, 2020; Popović, Peričin, Vaštag, Lazić, & Popović, 2012; Umaraw & Verma, 2017; Zhang, Liu, & Rempel, 2018).

2.3.4. Whey protein films

Whey protein has exceptional functional characteristics and film forming abilities (McHUGH, Aujard, & Krochta, 1994). Films based on whey have been studied for their excellent transparency, flexibility, oil/gas barrier properties at relatively low humidity (Coupland, Shaw, Monahan, O'Riordan, & O'Sullivan, 2000; Seydim & Sarikus, 2006). However, whey protein-based films show poor water barrier characteristics (Fang, Tung, Britt, Yada, & Dalgleish, 2002). Incorporation of essential oils/lipids has been shown to improve the water barrier and other active properties of whey-based films (Bahram et al., 2014; Seydim & Sarikus, 2006). However, determination of interaction between various biopolymers during coatings formation is necessary in order to develop packaging material with desired characteristic and functionality (Perez-Gago & Krochta, 2001). Çakmak, Özselek, Turan, Firatligil, and Güler (2020) developed edible films based on whey protein isolate plasticized with glycerol. They reported bergamot essential oils and lemon as active ingredients among bioplastic films. Results indicated excellent antimicrobial potential due to the incorporation of essential oils. In addition, significant oxygen permeability and water vapor permeability was reported.

3. Processing of biobased packaging materials

Development of biobased packaging material follows a multistep methodology e.g., (i) breakage of intermolecular linkages; (ii) synthesis of new molecular arrangement and (iii) the development of 3D polymeric network by the newly formed linkages (Galić, Ščetar, & Kurek, 2011). Establishment of new molecular linkages lies upon processing conditions and the polymer shape (*viz.* length, width, and ratio) (Ballesteros, Michelin, Vicente, Teixeira, & Cerqueira, 2018; Galić et al., 2011). The newly made films are stabilized by the interactions as a result of hydrophobic, electrostatic, covalent and H-bonding (Liu et al., 2019; Zubair & Ullah, 2020). Two basic processing methodologies i.e., (i) wet processing and (ii) dry processing has frequently been reported for the development of biobased plastic materials. Dry processing lies on thermoplastic properties of polymers in which thermo-mechanical treatment result in the initiation of sulfhydryl/disulfide conversion reaction, however, wet processing lies on dissolution and the type of solvent used, the pH of solvent that can change the polymer confirmation (Blanco-Pascual, Fernández-Martín, & Montero, 2013). Fig. 3 represents the processes used for the synthesis of biobased plastic materials.

3.1. Wet processing

Casting, continuous spreading or wet processing method has frequently been used for the development of biobased plastics from renewable resources e.g., lipids, proteins and carbohydrates (Fig. 3A). Wet processing depends upon biopolymer dissolution in an appropriate solvent for the development of film forming solution. Several substances are used as additives to bioplastic films, which may be in the form of antimicrobial agents, cross-linking agents, micro/nanostructures, antioxidants, plasticizers and fillers (Felix, Perez-Puyana, Romero, & Guerrero, 2017). This method can be categorized by the casting of film forming solution, and the solvent evaporation. Addition of plasticizer is effective as it reduces intermolecular linkages and the hardness, ultimately giving smoother and flexible material (Lin & Krochta, 2003; Sanyang, Sapuan, Jawaid, Ishak, & Sahari, 2016; Sothornvit & Krochta, 2001). Wet processing is useful for packaging industries as it enhances the mechanical characteristics of bioplastic films (Farris, Introzzi, & Piergiovanni, 2009).

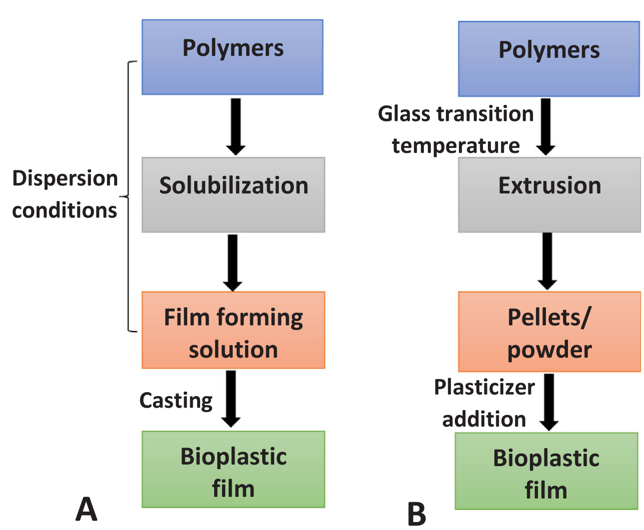

Fig. 3. (A) Wet processing (B) dry processing for the development of bioplastic films.

3.2. Dry processing

Whereas, dry processing is based upon thermoplastic characteristics of polymers which exhibit excellent role in the development of packaging material. This process can be linked with the theory of glass transition, in which a glassy material is converted into semi-solid condition at particular temperature. This transition (semi-solid) state basically cause molecular mobility and disorderness and alters the mechanical as well as physicochemical characteristics of polymers. Intermolecular linkages between proteins molecules break causing denaturation, and the new linkages and bonds establish causing modifications in material properties (Cuq, Gontard, Cuq, & Guilbert, 1998; Khwaldia, Perez, Banon, Desobry, & Hardy, 2004). Polymer-based packaging can be synthesized by different approaches e.g., (i) extrusion and (ii) thermal processing (Fig. 3B). Both processes can be applied independently or simultaneously in which extrusion is used for minor modifications and mixing while, thermal processing for the development of final product.

4. Reinforcements to bioplastics films

4.1. Bioactive reinforcements with antioxidant capacity

Currently, consumer demand for safe and healthier foodstuff has encouraged the scientists to develop novel preservation strategies. Several techniques have been applied to minimize lipid oxidation e.g., the addition of antioxidants directly to foodstuff or their incorporation into packaging material (Domínguez et al., 2018; Kusznierewicz, Staroszczyk, Malinowska-Pańczyk, Parchem, & Bartoszek, 2020). Foodstuff such as fish products or fresh red meat cannot be packed in the absence of oxygen. Direct addition of antioxidant material to foodstuff, encounter limitations, as when the active ingredients are chemically consumed, the protection stops and the food start degradation at an increased rate (Mastromatteo, Mastromatteo, Conte, & Del Nobile, 2010; Navikaite-Snipaitiene et al., 2018). Presently, antioxidant packaging is being developed which is based on the addition of antioxidant in the packaging material to improve stability of oxidation-sensitive foodstuff. For this purpose, the use of natural antioxidants has been widely studied, particularly plant essential oils (Table 1). Essential oils exhibit excellent antioxidant and antimicrobial properties (Deng et al., 2020; Heredia-Guerrero et al., 2018). However, the utilization of essential oils as food preservatives is restricted due to stronger flavor (Zeng, Zhang, Wang, & Piao, 2015). In order to overcome this problem, edible films are made with bioactive agents to induce desired functionality (Fig. 4) (Vilela et al., 2018). In previous few years, essential oils have been broadly reported as incredible reinforcements in biobased and biodegradable coatings and films. They are predicted to aid in the reduction of aqueous uptake properties, because of their lipidic nature. Besides, they help in the improvement of mechanical properties of polymeric films e.g. optical structure, tensile strength, by providing antimicrobial and antioxidant properties as well (Iamareerat, Singh, Sadiq, & Anal, 2018; Moradi, Tajik, Rohani, & Mahmoudian, 2016).

Foline-Ciocalteau (F.C assay) is used to study the phenolic content

Table 1
Raw materials for bioplastic/biocomposite films development with their origin, active reinforcements, advantages and disadvantages.

Raw material	Origin	Active ingredient	Advantages	Disadvantages	References
Starch	Corn	Grape juice	Good mechanical properties Good oxygen permeability Higher glass transition temperature Improved transparency and surface properties	High water sensitivity	Yıldırım-Yalçın, Şeker, and Sadıkoğlu (2019)
Gum	Basal seeds	Oregano essential oil	Acceptable transparency Improved water vapor permeability Good antioxidant and antimicrobial properties	High water vapor permeability by increasing active agent concentration	Hashemi and Khaneghah (2017)
Starch	Potato	Citric acid and sorbate potassium	Cost effectiveness Increased food shelf life Good antifungal properites Decreased product loss	High sensitivity to moisture	Saraiva et al. (2016)
Starch	Corn	Cellulose fibers from rice/coffee husk	Good antibacterial properites Good antioxidant properties Improved mechanical properties Oxygen barrier properties Improved water vapor permeability	Low transparency	Collazo-Bigliardi, Ortega-Toro, and Chiralt (2019)
Carboxymethyl cellulose	Rice stubble	Thai rice grass extract	Extended food shelf life Good mechanical properties Good antioxidant properties	High water sensitivity	Rodsamran and Sothornvit (2018)
Carboxymethyl cellulose	Rice stubble	Olive oil	Improved film properties Improved moisture barrier Good mechanical and water vapor permeability properites	High sensitivity to aqueous uptake	Rodsamran and Sothornvit (2017)
Gum	*Eucommia ulmoides*	Nanocrystalline cellulose	Improved elongation at break Good water vapor permeability Improved tensile characteristics	Lower thermal stability Decreased glass transition (T_g) temperature	Sun et al. (2018)
Pectin/ gelatin	Citrus fruit/ fish	Olive oil	Reduced oxygen permeability Excellent for the storage of meat products	Sensitive to moisture uptake	Bermúdez-Oria, Rodríguez-Gutiérrez, Rubio-Senent, Fernández-Prior, and Fernández-Bolaños (2019)
Pectin/ gelatin	Citrus fruit/ fish	Olive phenol	Good film forming properties Good antioxidant properties Good antimicrobial properties Improved mechanical properties	–	Bermúdez-Oria, Rodríguez-Gutiérrez, Vioque, Rubio-Senent, and Fernández-Bolaños (2017)

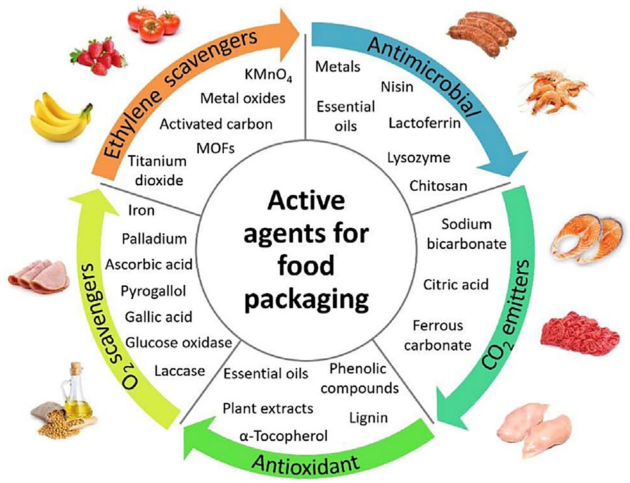

Fig. 4. Bioactive agents for smart food packaging applications. Reprinted from Vilela et al. (2018) with permission from Elsevier B.V.

in biopolymeric films. The 2,2-diphenyl-1-picrylhydrazyl (DPPH) is a classical synthetic radical used for phenolic antioxidants activity measurements. Disappearance of DPPH radical by the action of antioxidant is measure by spectrophotometer at 515 nm until constant absorbance attains. DPPH activity assay is restricted because radicals can develop interactions with other radicals e.g. alkyl, and the estimation curve is not linear, presenting various ratio of DPPH/ antioxidant (Frankel & Meyer, 2000). Green tea extract has been incorporated into chitosan films which showed good antioxidant capacity, and by increasing the concentration of green tea extract increased the antioxidant capacity of polymeric films (Siripatrawan & Noipha, 2012). The ferric cyanide reducing assay has been described by several scientists (Huang et al., 2011). Outcomes have described as mmol/g of ascorbic acid in bioplastic, higher the absorbance, higher will be the reducing power. Reduction strength of hake protein-based films has increased by the addition of thyme oil (Pires et al., 2011). Another study compared the hake protein reducing ability by incorporating with various essential oils and found that coriander oil was highly efficient among all (Pires et al., 2013). Moradi et al. (2016) positively correlated the addition of *Zataria multiflora* oil into the chitosan-based films with antioxidant activity.

The ferric-reducing antioxidant power (FRAP) directly determines the power of antioxidants to reduce Fe^{+3} into Fe^{+2} at acidic pH. Antioxidative potential of chitosan bioplastic incorporated with thyme oil has been determined using this technique by Ruiz-Navajas, Viuda-Martos, Sendra, Perez-Alvarez, and Fernández-López (2013), concluding that antioxidative potential depends on concentration of additive. FRAP assay was also used by other scientists to determine antioxidative potential of gelatin-based films with essential oils of rosemary/oregano. Results indicated that films incorporated with oregano essential oil were more active than those incorporated with rosemary essential oils (Gómez-Estaca, Bravo, Gómez-Guillén, Alemán, & Montero, 2009). Chitosan films present wonderful ferrous iron chelating (FIC) activity, which was increased after the addition of thyme oil, as with the increasing concentration, more the activity was observed (Ruiz-Navajas et al., 2013).

The substrate azinobis (3-ethylbenzothiazoline-6-sulfonic acid) or ABTS is a colorless chemical compound. Dilution of solution was made with ethanol, measured the absorbance at 734 nm and mixture is formed with antioxidant sample. Absorbance reduction after 6 min incubation is compared with the calibration curve. Using this technique antioxidant capacity of gelatin films have measured with the addition of citrus essential oils. It has also been concluded that lemon has excellent antioxidant properties (Tongnuanchan, Benjakul, & Prodpran, 2012). Other biobased materials have also been incorporated into bioplastic films having antioxidant capacities. Bioplastic films based on polystyrene have also been obtained having antioxidant capacity by the addition of olive leaf extract and α-tocopherol (Marcos et al., 2014). Soy protein films showed improved antioxidant capacity with the incorporation of red grapes extract, obtained by compression molding rather than casting (Ciannamea, Stefani, & Ruseckaite, 2016). This specifies the great influence of the technique used to obtain the film on the matrix release properties and ultimately the film activity. The film extract is used after dissolving in a suitable solvent by solidification, crushing and mixing. Phenolic compound availability is normally correlated with antioxidant capacity of the bioplastic films.

4.2. Bio-based reinforcements with antimicrobial capacity

Antimicrobial packaging is the incorporation of antimicrobial agents into the packaging materials to retard the microbial growth in food products (Han, 2003, 2005). Such type of packaging provides improved quality, food safety and extended shelf life by preventing the growth of pathogenic microbes (Lavoine et al., 2014). Utilization of antimicrobial agents in food packaging has decreased the universal food-borne epidemics (Kim & Rhee, 2016). Moreover, the consumer's choice of fresh, least processed and additive free food products also made the antimicrobial packaging more interesting (Moon & Rhee, 2016). The antimicrobial agents consumed for the preservation are chemically produced or extracted from biomass of animals, plants and microorganisms (Table 1). These may include chitosan, enzymes, essential oils and natural extracts from different plant sources (Pereira et al., 2015; Qamar, Asgher, & Bilal, 2020). Essential oils extracted from different plants are the most abundant source of bioactive compounds such as phenolic compounds and terpenoids etc., which generally are named as antimicrobial agents (Ruiz-Navajas et al., 2013). Essential oils act on the microbial cells by a variety of different mechanisms; disrupting enzyme structures, damaging the phospholipid bilayer of cell membrane and compromised genetic makeup of microbes. Fig. 5 shows active packaging material based on bionanocomposites with excellent preservative effect against UV-irradiation and food borne pathogens (Kumar, Nehra, Dilbaghi, Tankeshwar, & Kim, 2018).

Plant-based bioactive reinforcements have prominent activity against bacterial and fungal species (Fardioui, Kadmiri, el kacem Qaiss, & Bouhfid, 2018; Khan, Huq, Khan, Riedl, & Lacroix, 2014). The antibacterial activity was confirmed by using different methods such as agar dilution method, disk diffusion method and broth dilution method. Antimicrobial activity of garlic oil was tested by mixing the oil and bacterial inoculums of *E. coli, B. cereus, S. aureus* and *S. typhimurium.* It was shown that the garlic oil retarded the growth of all bacteria and could be incorporated into the edible packaging (Pranoto, Salokhe, & Rakshit, 2005). Essential oils and other bioactive compounds have role in the chemical structure of film formation by interacting with the plasticizers and the polymers by decreasing the diffusion of antimicrobial agents into the food product (Ruiz-Navajas et al., 2013). Antimicrobial compounds released from bioplastic packaging are dependent upon a number of factors, which may include osmosis, electrostatic interactions between polymer and antimicrobial agents, structural changes caused by antimicrobials and ecological circumstances (Avila-Sosa et al., 2012). Despite advantages, it is recommended to use lesser amounts of antimicrobial compounds in edible films to gain a particular shelf life by their gradual release into the foods (Han, 2003; Malhotra, Keshwani, & Kharkwal, 2015).

Disk diffusion method considered to be the most appropriate method to screen the antibacterial activity of films. Studies have utilized this protocol to examine the antibacterial activity of edible films reinforced with different plant-based essential oils (Aminzare, Hashemi, Hassanzad Azar, Amiri, & Abbasi, 2017; Ruiz-Navajas et al., 2013). It was observed that the excellent antibacterial activity was exhibited against different gram-negative and gram-positive bacteria. By using same method, it was found that the essential oil containing films were

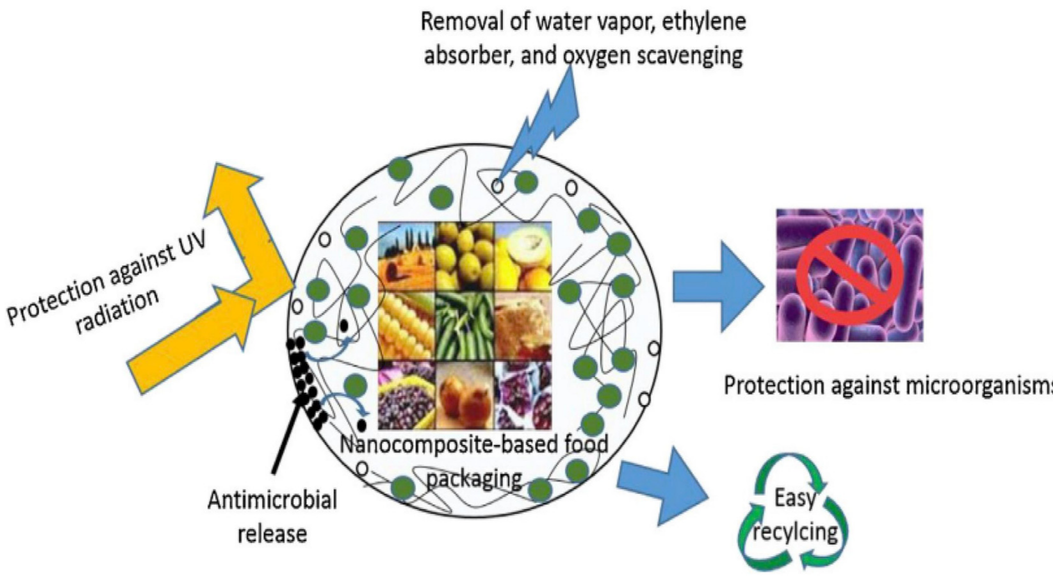

Fig. 5. Active food packaging based on bio-nanocomposites with excellent preservation ability against UV-irradiation and pathogens. Reprinted from (Kumar, Nehra, Dilbaghi, Tankeshwar, & Kim, 2018) with permission from Elsevier B.V.

considerably more efficient for gram-positive than for gram-negative species (Benavides, Villalobos-Carvajal, & Reyes, 2012). In recent studies; rosemary, oregano and garlic essential oils were incorporated into proteineous films and noticed that the oregano oil containing bioplastic have great antimicrobial activity, while the least activity was shown by rosemary oil (Aldana, Andrade-Ochoa, Aguilar, Contreras-Esquivel, & Nevárez-Moorillón, 2015).

The antimicrobial activity of neem extract was tested by Jagannath, Radhika, Nanjappa, Murali, and Bawa (2006) by incorporating it into starch-gelatin-based blends. They observed the decrease in its effectiveness against *S. aureus, S. typhimorium, E. coli, B. cereus* and *Pseudomonas* species. Metallic nanoparticles incorporated into biodegradable bio-packaging for the purpose of improving barrier and mechanical properties and enhancing product's shelf life. De Oliveira Pizzoli et al. (2016) studied the effect of silver nanoparticles into PLA/TPS/Gelatin films, which presented great inhibitory effect for wide range of fungi, bacteria and viruses. Silver nanoparticle's antimicrobial activity was successfully proven, the effectiveness depends upon release behavior of particles from packaging material in contact with foodstuffs (Cano, Jiménez, Chäfer, Gónzalez, & Chiralt, 2014).

5. Compostability of bioplastic packaging

By the definition of ASTM-D6400, a compostable material is any material that degrades by biological means at a rate comparable to other known compostable substances. Hence, only substances which can degrade biologically in a composting environment can be labeled as 'compostable' (ASTM standards, 2004). Not all microbially degradable substances are compostable. Among all packaging types only few plastic packaging and papers are bio-degradable and thus, compostable. Microbes have ability to directly utilize biopolymers e.g., cellulose, lignin and starch-based polymers. Enzymatic degradation results in the reduction of molecular weight in surroundings i.e., outside microbial cell (Thakur et al., 2018). Enzymatic cleavage results in the formation of small segments, and the segment which is small enough as compared to microbial cell is transferred inside and consumed. Rate of biodegradation of these polymers can be enhanced by hydrolysis which results random breakage of polymers and ultimately reduction in molecular weight. The resulting low molecular weight compounds are more vulnerable to enzyme-based degradation (Emadian, Onay, & Demirel, 2017) (Fig. 6).

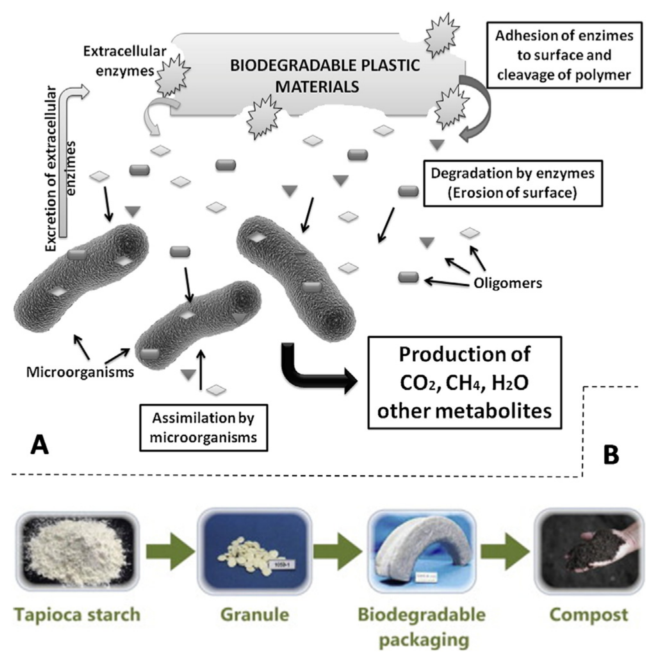

Fig. 6. (A) General degradation mechanism of plastic products via microorganisms. Reprinted from (Moustafa, Youssef, Darwish, & Abou-Kandil, 2019) with permission from Elsevier Ltd.; (B) Development of biodegradable packaging material from tapioca starch granules and its compost-ability in natural environment. Reprinted from Razza et al. (2015) with permission from Elsevier B.V.

Bacterial polyhydroxyalkanoates are another type of polyesters which have already been discussed in previous section. These are synthesized by various bacterial species as reserve materials, when they suffer with nutrient limited conditions in the presence of excess carbon (Raza, Abid, & Banat, 2018). Vinyl polymers, e.g. polyethylene, polypropylene, polyvinylchloride comprising carbon backbones, are generally not prone to biodegradation or hydrolysis. Polyvinyl alcohol (PVA) is an exception, which is degradable through biological means due to its high hydrolysis power. Enzymatic oxidation of hydroxyl group leads to the formation of carbonyl groups, cause cleavage in the polymer chain, ultimately decreasing molecular weight (Halima, 2016).

Fig. 6 shows the mechanisms of biodegradability and compostability of plastic packaging (Razza et al., 2015).

Hetero polymers have atoms other than carbon in their backbone, such as nitrogen and oxygen. Those atoms make polymers more susceptible to hydrolysis, and ultimately make them more susceptible to biodegradation (Asina et al., 2016). Nylons, polyesters, polycarbonates etc. are synthetic hetero polymers, which do not exhibit significant level of biodegradation. Currently, well known hetero polymers are PLA, polycaprolactone (PCL) and polyglycolic acid (PGA). Bacterial polyester e.g., PHAs can also studied in this category. Hydrolysis rate elevation leads to more available sites of microorganism to attack, causing increase in the rate of biodegradation. Polymeric hydrolysis is partially regulated by the diffusion rate of water in polymeric amorphous regions (Rowe, Eyiler, & Walters, 2016). Biodegradable polymers are mostly aliphatic polyesters e.g., PLA, PHA and PCL, having a hopeful tomorrow for packaging applications. It also includes polybutylene succinate (PBS), polybutylene adipate-co-terephthalate (PBAT), polybutylene succinate adipate (PBSA) and few polyesteramides, due to their comparable properties to petrochemical-based polymers mostly found in packaging applications. PHBV is hydrophobic in nature and also offers excellent gas barrier properties (Dasan, Bhat, & Ahmad, 2017).

Heat stability of PLA and its processability are similar to those of polystyrene; flavor barrier and grease/oil resistance properties are like PET; and also have low sealing temperature as compared to polyethylene and polypropylene (Auras, Harte, & Selke, 2004). These polymers contain minimum one hydrolysable bond in their backbone: ester, carbonate, ether or amide (Göpferich, 1996). Whereas C-C bonds enhance stability, the existence of hydrolysable groups in the backbone of polymer, vividly increased susceptibility to microbial degradation (Kale et al., 2007) it not only increases the susceptible sites for hydrolysis but also enhance polymer flexibility, hence polymer chains can easily be arranging themselves to fit into enzyme active sites. Degradation through microorganisms *via* enzymes, occurs at the polymer surface, whereas non-enzymatic breakdown can occur anywhere in polymer's bulk because diffusion of water takes place through polymer's amorphous regions. Both reactions mostly proceed at simultaneously (Auras et al., 2004).

6. Conclusions

The applications of bioplastic films in food packaging technology provide novel biodegradable and eco-friendly alternative to petrochemical-based plastics. This can solve the waste accumulation problem due to petrochemical-based plastics, which are nonbiodegradable in nature. The mechanical and barrier properties reported for natural-based biodegradable polymer films are away from the properties of most petroleum-based plastics as packaging materials in food industry. However, these low-quality films are being improved by the addition of several other biopolymers in the form of composites/blends. Erosion mechanism also play crucial role in determining biopolymeric applications. For example, if a polymeric material is used as drug delivering agent, active constituent existing in polymeric surface, undergoing surface erosion will release the drug faster, on the other hand, for a slowly releasing drug, polymer that experience bulk erosion is more suitable so active ingredient present in polymer can be released over longer time period. A great debate exists about the possible applications of bioplastic films on foodstuffs. Efforts are focused on the development of correct combination of materials mixed, because efficiency of biopolymeric films depend upon the polymers and other materials used as additives to enhance functional properties. Further research to improve the properties of bioplastics to achieve similar properties as petroleum-based plastics is required. The replacement of synthetic plastics with bio-based plastic is still in its fantasy and more in-depth understanding, awareness, and considerations are required in future studies.

Declaration of Competing Interest

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

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