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Biobased Food Packaging Materials: Sustainable Alternative to Conventional Petrochemical Packaging Materials: A Review

Tipale Mayuri[1], R.N. Shukla[1], Jadhav Balaji[1]

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ABSTRACT

Plastics as a packaging material in the food industry is highly preferable, but due to environmental concerns and government legislation, there is a need to develop biobased biodegradable packaging materials. This review includes the sources of biopolymers, methods of productions, future and applications of biobased packaging materials in food packaging. As bioplastic materials have some limitations in terms of mechanical and barrier properties in comparison with synthetic plastics. These limitations can be overcome by using suitable fillers, nanocomposites and certain additives in bioplastics production. The development of bioplastic has a high potential to replace non-biodegradable plastic packaging. Biopolymer with synthetic polymers blends is also an emerging area of research. Bench casting and continuous casting are the popular methods for bioplastic production in the scientific community. According to the global production capacities of the bioplastics report, the present bioplastics production is only 2% of the production volume of the synthetic polymers, it will increase 4% until 2023.

Key words: Biobased plastic, Biopolymers, Food packaging, Renewable resource.

Petrochemical-based plastic materials have been widely employed for the packaging of food materials owing to their good mechanical, barrier and optical properties. Despite having many advantages of petrochemical plastic, the major disadvantage of this material is that it is non-biodegradable which is today's burning issue around the globe.

In the last 15 years, half of the plastics ever produced have been manufactured. Plastic output expanded at an exponential rate from 2.3 million tons in 1950 to 448 million tons in 2015, with production predicted to double by 2050 (Parker, 2019). According to a Central Pollution Control Board study for 2018-2019, India generates 3.3 million metric tons of plastic garbage every year. Concerns about the environmental impact of petroleum-based plastic packaging materials, such as natural resource depletion, energy crisis, global warming and ecological challenges, have sparked interest in the development of bioplastics materials based on biopolymers. Biopolymer is an organic polymer produced by living organisms and plants from natural resources. Biopolymers have the characteristics of being fully capable of biodegradation at accelerated rates and breaking down cleanly into simple molecules such as carbon dioxide, water and methane under the enzymatic action of microorganisms (Nase *et al*., 2014). They are categorized based on their production method or source: 1) Polymers taken or removed directly from biomass, such as polysaccharides and proteins, 2) Polymers produced using traditional chemical synthesis from sustainable bio-based monomers such as polylactic acid (PLA), 3) Microorganisms or genetically engineered bacteria create polymers such as polyhydroxyalkanoates and bacterial cellulose (Nase *et al*., 2014).

Film-forming biopolymer materials

Film-forming biopolymers can also be differentiated based on the type of film-forming material, which forms cohesive or continuous matrices. Biopolymers are differentiated into composite films, protein, lipid and polysaccharides and they are shown in the Fig 1.

[1]Department of Food Process Engineering, Sam Higginbottom University of Agriculture, Technology and Sciences, Prayagraj-211 007, Uttar Pradesh, India.

Corresponding Author: Tipale Mayuri, Department of Food Process Engineering, Sam Higginbottom University of Agriculture, Technology and Sciences, Prayagraj-211 007, Uttar Pradesh, India. Email: mayuritipale9@gmail.com

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Composite films

Composite films are made up of protein, lipid and polysaccharides blends and are noted for their mechanical qualities. Lipid-based coatings have poor mechanical characteristics and are less permeable to water vapor while permeable to oxygen. The characteristics of the composite film can be improved by adding the optimum number of components. Kim and Ustunol (2001) invented the film by combining butterfat, candelilla wax with protein and plasticizer. Water solubility (20°C, 24 h) and moisture sorption isotherms (0.18-0.90 (a_w), 25°C) of the films were determined. The film's solubility and equilibrium moisture content increased rapidly, however, the addition of lipid lowered the solubility and equilibrium moisture content of sorbitol and glycerol plasticized films (Kim and Ustunol, 2001).

It has been reported that adding lipids (50% oleic acid), commercial vegetable oil and lecithin of sodium caseinates films resulted in an increase in water vapor permeability with increasing fatty acid concentration (4.88-22.56%, 12.20-48.14% and 61.59-15.85%) of the dry weight of casein (Hammam *et al*., 2019).

Lipid-based film

Lipids are good moisture barriers. Herbal wax, acetylated monoglycerides and surfactants are among the lipid-based substances used as film/coating. Paraffin and beeswax are the most basic lipid compounds. Because of their low polarity, lipid-based films and coatings are thought to be particularly effective at preventing moisture delivery. Lipid films and coatings are typically more brittle and thicker because of their hydrophobicity. Water vapor permeability decreases as the concentration of the hydrophobicity phase increases (Hassan *et al*., 2018). Different types of waxes are utilized in the formation of films and their ability to retain moisture varies. As paraffin wax is made up of a mixture of long hydrocarbon chains, whereas beeswax is made up of a mixture of hydrophobic compounds, it is more effective than beeswax. Fatty compounds have a high moisture retention capacity, substances such as plant fats, animal fats and waxes have been employed in the manufacture of the biobased film (Hammam *et al*., 2019). Microemulsion cast films made from glycerol plasticized mesquite seed gum and sonicated palm fruit oil emulsion was successful. The inclusion of palm fruit oil increased the hydrophobicity of the films. This film is suitable for a variety of food packaging applications that require low water vapor permeability and solubility (Rodrigues *et al*., 2016).

Protein-based film

Proteins are found in nature as fibrous or globular proteins. Proteins that can be employed in film production include lactic serum, casein, collagen and seine, soya protein and zein protein. Because of their unique structure, protein films have better mechanical qualities than polysaccharides. Modification of stearic acid soy protein isolate can be made from the cake leftover after extracting soya oil. Soy protein isolate can be remanufactured into plastic, stearic acid was used to improve the water-resistance of soy protein isolate films using the bioconjugation method and it considerably enhanced the film's water resistance (Ye *et al*., 2019). Schmid *et al*., (2013) used glycerol as a plasticizer to both hydrolyzed and non-hydrolyzed whey protein isolate films. This study found that increasing the content of hydrolyzed whey protein isolate greatly modifies the mechanical characteristics. The tensile and elastic characteristics of the film decreased (Schmid *et al*., 2013). Protein-based films outperformed lipid and polysaccharide-based films in terms of gas barrier and mechanical properties. The formation of a pumpkin oil cake protein isolate film with glycerol as a plasticizer at various pH values revealed that the film could be formed with a wide pH range (2 to 12) (Popovic *et al*., 2012). Zein, a protein derived from corn, is found in the grain's endosperm and is alcohol soluble. Zein films were created by combining oleic acid and glycerol as plasticizing agents in various ratios. The highest tensile strength was observed at an oleic acid to glycerol ratio of 3:1(Xu *et al*., 2012).

Polysaccharide based films

Plants and animal cells, the outer structure of insects all contain polysaccharides. Biobased and biodegradable materials have seen increased use in packaging, agriculture, textiles, medicine and other fields in recent years. There are numerous polymers blends available to replace the current synthetic plastic. The most important polymer is starch. Gums, cellulose derivatives, carrageen, chitosan, alginate and pectin are some of the examples of polysaccharides. Polysaccharides are inexpensive,

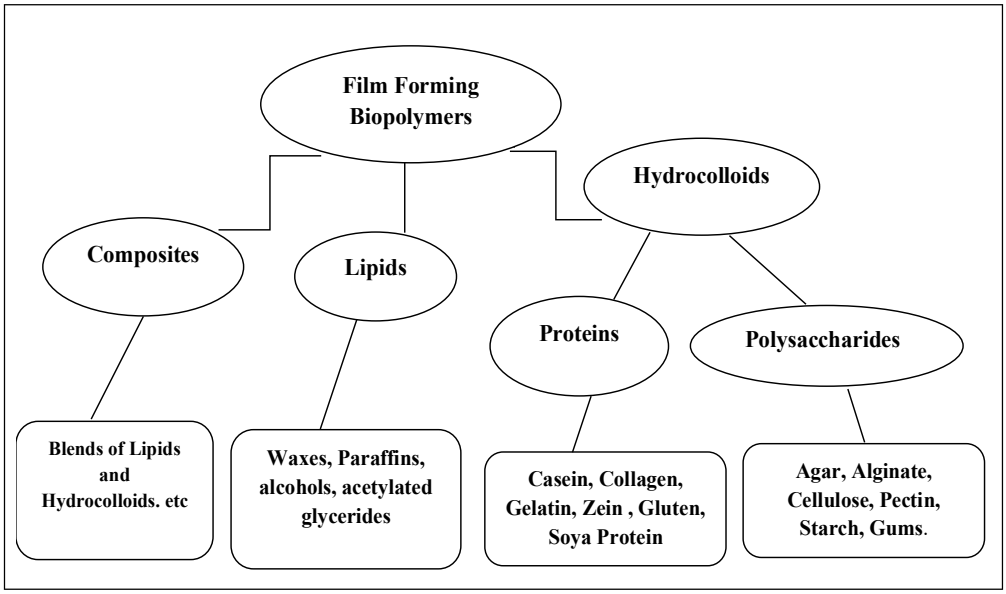

Fig 1: Film-forming biomaterials (Parreidt *et al*., 2018).

renewable and abundant in nature. They combine to form a film with superior mechanical properties. The polysaccharide-based packaging material is readily available and has some functional properties. Polysaccharide-based biodegradable films are an effective barrier to gas transfer, such as oxygen and carbon dioxide (Caz *et al*., 2017). Starch is made up of amylose and amylopectin, which are found in grains like corn, wheat and sweet potato. Amylose is in charge of forming a film. Increasing the amount of amylose in the films increases their elasticity (Hammam, 2019). Cellulose is the most common biopolymer found in nature. Because of its high hydrophilicity and crystal structure, it is not easily soluble in water. Cellophane films, which are used in food packaging, are made of cellulose (Pellicer *et al*., 2017). Alginate is a polysaccharide extracted from brown algae such as *Laminaria, Macrocystis* and *Azotobacter vinelandii* and bacterial alginate was extracted from *Azotobacter vinelandii*. It is a binary copolymer with carboxyl groups in every constituent residue (Daniela, 2012). Chitin is nature's second most abundant polysaccharide after cellulose. Chitin can be found in the exoskeletons of crustaceans and several insects, which is why chitosan derived from chitin is commercially available from abundant renewable sources (Caz *et al*., 2017). Chitosan is a biomaterial that is biodegradable, bio-functional, non-toxic and biocompatible (Dutta *et al*., 2009). Chitosan has the disadvantage of not being a thermoplastic polymer and thus cannot be extruded or molded at high temperatures (Caz *et al*., 2017). Pectin is a complex anionic polysaccharide composed of β-1, 4 linked d-galacturonic acid residues. Pectin contains the carboxyl group of uronic acid, based on the degree of esterification it is divided into two types: high methoxyl pectin DE>50% and low methoxyl pectin DE<50%. Low methoxyl pectins form a gel with divalent cations (Galus *et al*., 2013). Batori *et al*., (2017) made a biofilm from citrus peel waste for food packaging by combining the gelling ability of pectin and the strength of cellulosic fibers.

Microbial sources of bio-film

To improve the degradation and other properties of the films, biodegradable polymer blends have been developed. PHA, or Poly-b-hydroxy alkanoates, is a biocompatible and biodegradable thermoplastic with potential applications that have received a lot of attention. Poly 3- hydroxybutyrate (PHB) is used in bulk shrink packaging and flexible intermediate bulk containers (Jabeen *et al*., 2015). PHA and bacterial cellulose packaging materials have the potential. PHB is another natural polymer; both PHA and PHB are thermoplastic polyesters and have mechanical properties close to synthetically produced degradable polyesters for use in food packaging (Gurgel *et al*., 2011). Bacterial cellulose produces by bacteria *Acetobacter xylinum* and *A. pasteurianus*. Its chemical and physical structure is similar to the plant cellulose (Taylor *et al*., 2002). *Aureobasidium pullulans* synthesize pullulan, a microbial polysaccharide composed of maltotriose units linked by α (1,6) glycosidic units, from starch. Pullulan is water-soluble, has a low oxygen and oil permeable film, is odorless, colorless and heat-sealable (Mohamed *et al*., 2020). The bacteria *Xanthomonas campestris* produces xanthan gum, which is an exopolysaccharide. It forms a stable viscous solution in hot or cold water over a wide temperature and pH range (Mohamed *et al*., 2020).

Binding agents

Binding agents are hydrocolloids that are added to improve the physical properties of the film; the addition of biomacromolecules increases the film's cohesiveness. The chemical structures of the hydrocolloids are expected to have the ability to form films. As binding agents, pectin, cellulose, carboxymethylcellulose, methylcellulose, gelatin, collagen, chitosan, alginate and carrageenan are used. Sometimes more than one binding agent is used in films (Otoni *et al*., 2017).

Plasticizers

Plasticizers are low molecular weight agents that are incorporated into polymeric film-forming materials to increase the polymers' thermoplasticity. Plasticizers increase the free volume of polymer structures or polymer molecular mobility, as well as their flexibility and processability (Han *et al*., 2014). Natural plasticizers with low toxicity and migration include epoxidized triglycerides, vegetable oils derived from soyabean oil, linseed oil, castor oil, sunflower oil and fatty acid esters (Gurgel *et al*., 2011). The compatibility of the plasticizer and the polymer is important for effective plasticization. Polarity, hydrogen bonding, solubility parameters and dielectric constant are all examples of parameters. Glycerol, also known as vegetable glycerin, is a common plasticizer that is suitable for water-soluble polymers. Glycerol has a higher plasticizing efficiency than sorbitol (Otoni *et al*., 2017). Water is the most common and effective plasticizer; however, the plasticizing effect of water in hydrophilic biopolymers is difficult due to environmental conditions such as relative humidity and temperature (Parreidt *et al*., 2018). To overcome the brittleness of alginate films, some studies combine more than one plasticizer, such as glycerol, palmitic acid, β-cyclodextrin and glycerol monostearate used in film formulations (Fan *et al*., 2009). Miller *et al*. (2021) investigated the effect of glycerol and sorbitol plasticizers on potato-based film and discovered that as plasticizer concentration increased, so did the film's water vapor and oxygen permeability. A glycerol-containing film had higher water vapor and oxygen permeabilities than a sorbitol-containing film (Miller *et al*., 2021).

Fillers

In comparison to petrochemical-based plastic, biobased packaging films have lower mechanical resistance, barrier and thermal properties. A biobased nanocomposite is a multiphase material formed by the combination of two or

more components, including a matrix in the continuous phase and a matrix in the discontinuous phase at the nanometer scale (Ramos *et al*., 2018). Polymer nanocomposites are polymer-nanoparticle mixtures; polymer nano reinforcement fillers are added to polymers to produce nanocomposites with improved mechanical and other properties. Fillers include polysaccharide nanoparticles such as cellulose nanocrystals (CNC), microcrystalline cellulose (MCC), montmorillonite (MMT) and nano clays (Otoni *et al.,* 2017). Eco flex includes 1.0 wt. % biobased silica/carbon hybrid nanoparticles (SCNPs). The TGA and tensile test analysis data revealed that the thermal and mechanical properties of the film have improved significantly (Biswas *et al*., 2017). Clay can be used as a reinforcement agent because it improves the physicochemical, mechanical and barrier properties of several biopolymers. Clay is a natural, non-toxic and abundant resource (Ramos *et al*., 2018).

Other additives

Additives used in biobased packaging film are cross-linking agents, antioxidants and antimicrobials. These additives are also known as functional additives, it provides improved characteristics to the packaged product or packaging material.

Crosslinking agent

Crosslinking of polymer matrices forms covalent bonds that connect adjacent chains. Crosslinking can be accomplished through two methods: physical, chemical, or radiation. Physical treatment entails the formation of a tridimensional network, whereas chemical crosslinking entails the addition of appropriate chemical agents (Otoni *et al*., 2017). Minerals such as Ca^{2+}, Mg^{2+}, Fe^{3+}, proteins, organic, amino and phenolic acids are the naturally occurring crosslinkers present in fruit and vegetable pomace, peels and seeds (Otoni *et al*., 2017). When citric acid was used as a cross-linking agent in agarose bioplastics film, the mechanical properties of the film improved and the degradation rate of the crosslinked film was greater than that of the non crosslinked film (Awadhiya *et al*., 2016). Chitosan film was thermocompression molded with citric acid as a crosslinking agent, a natural polycarboxylic acid. The reaction of crosslinking resulted in the formation of a homogeneous structure (Guerrero *et al*., 2018).

Antioxidants

Antioxidants are substances that are used to preserve food by preventing deterioration, rancidity, or discoloration caused by oxidation (Parreidt *et al*., 2018). Antioxidants, both natural and synthetic, are successfully incorporated into biobased films. Antioxidants that are used in the biofilm are butylated hydroxyanisole (BHA), butylated hydroxytoluene (BHT), Tertiary butylhydroquinone (TBHQ), tocopherol, catechin, carvacrol, lignin, citric acid, sorbic acid and propyl gallate. The addition of active components varies between 0.02 to 12% mostly in the range of 1-5% (Jariyasakoolroj *et al*., 2018). The stability of the active compound varies from compound to compound as a result of the various thermal and mechanical processes. Polymer coupling improves the thermal stability of the antioxidant. Catechin was used as an antioxidant in the soy protein isolate and carboxymethylcellulose film because CMC causes the film to become opaque. Catechin incorporation shows a synergistic free radical scavenging effect and this film can be used as a potent antioxidant packaging material in the food industry (Han *et al*., 2015).

Antimicrobials

The biofilm contains antimicrobial agents of both natural and synthetic origin. The majority of essential oils are used as antimicrobial agents. The protein-based film contained bioactive agents, formic acid and oregano essential oil. Antimicrobial activity was demonstrated by bioactive agents (Martinez *et al*., 2013). Cinnamaldehyde in concentrations ranging from 1.5 to 5% increases cross-linking and imparts antimicrobial activity to gliadin films. The films were composted after being subjected to simulated conditions of use in a food packaging application. Pau *et al*. (2015) found that the disintegration profile of the most cross-linked gliadin film was very fast.

Methods of bioplastic formation

Casting

It is the most common technique, which entails spreading a film-forming solution on a suitable surface. The film-forming solution allows for surface drying and peeling. Rapid drying of the solution should be avoided; rapid drying reduces polymer chain mobility and the development of intermolecular interactions in the film (Parreidt *et al.,* 2018). Casting is divided into two methods: bench casting and continuous casting, with the former being more popular in the scientific community (Otoni *et al*., 2017). Continuous film casting can be done on steel conveyors or coating lines. Continuous casting has the advantage of requiring less space and labor and is more suitable for industrial applications (Otoni *et al*., 2017). Table 1 shows the materials used for biofilms formation. Starch-based thermoplastic materials have a broader range of industrial applications, including extrusion, injection molding and film blowing (Jabeen *et al*., 2015).

Future of bioplastic

According to the global bioplastics production market, the compound annual growth rate of the global bioplastics packaging market is 15.2% over the forecast period (2021-2026). Fig 2 shows the global production capacities of bioplastics in 2018-2023. Bioplastic packaging materials are becoming more popular due to their low environmental impact, sustainability and growing consumer awareness. By report biobased building blocks and polymers-Global Capacity, Production and Trends 2018-2023, The total production of bioplastics biopolymers

Table 1: Materials used for biofilms (Han *et al.*, 2014).

Functional compositions	Materials
Film-forming materials	**Proteins:** Collagen, gelatin, casein, whey protein, corn zein, gluten, soy protein, egg white protein, fish myofibrillar protein, sorghum protein, pea protein, rice bran protein, cottonseed protein, peanut protein, keratin. **Polysaccharides:** Starch, modified starch, modified cellulose (CMC, MC, HPC, HPMC), alginate, Carrageenan, pectin, pullulan, chitosan, gellan gum, xanthan gum. **Lipids:** Waxes- beeswax, paraffin, carnauba wax, candelilla wax, rice bran wax. Resins- shellac, terpene, acetoglycerides.
Plasticizer	Glycerin, propylene glycol, sorbitol, sucrose, polyethylene glycol, corn syrup, water.
Functional additives	Antioxidant, antimicrobials, nutraceuticals, colors.
Other additives	Emulsifiers (Lecithin, Tweens, Spans), lipid emulsion-waxes, fatty acids.

Table 2: Applications of bioplastic for food packaging (Jabeen *et al.*, 2015).

Packaging application	Biopolymer	Company
Coffee and tea	Cardboard cups coated with PLA	KLM
Beverages	PLA Cups	Musburger (Japan)
Fresh salads	PLA Bowls	McDonald's
Fresh cut fruits and vegetables	Rigid PLA trays and packs	Asda (retailer)
Potato chips	PLA Bags	Pepsico's Frito-lay
Yogurt	PLA Jars	Stonyfield (Danone)
Bread	Paper bags with PLA window	Delhaize (retailer)
Milk chocolate	Corn starch trays	Cadbury Schweppes
Organic tomato	Corn-based packaging	Iper supermarkets (Italy) Coop Italia
Kiwi	Biobased trays wrapped with cellulose film	Wal-Mart
Potato chips	Metalized cellulose film	Boulder Canyon
Sweets	Metalized cellulose film	Quality street, Thomton
Organic pasta	Cellulose-based packaging	Birkel

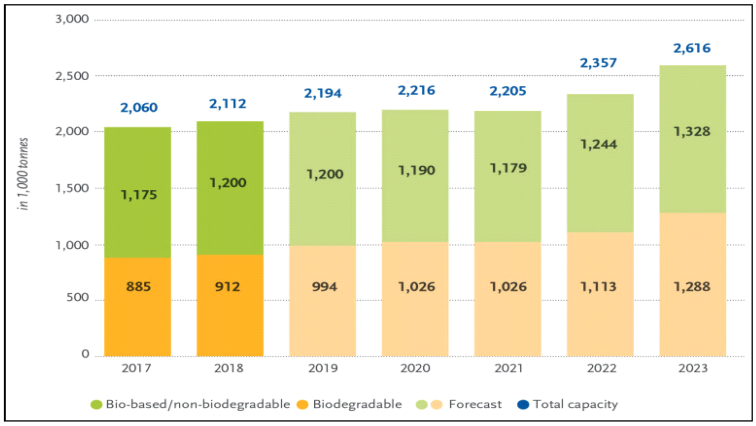

Fig 2: Global production capacities of bioplastics in 2018-2023 (Hong *et al.*, 2021).

has reached 7.5 million tons, accounting for 2% of total synthetic polymer production and is expected to rise by 4% by 2023.

Application of bioplastic in food packing

There are numerous biobased materials available in the market, with PLA being one of the most widely used. Bioplastics can be used to pack food products with both short and long shelf lives (Peelman *et al.*, 2013). The following are some of the biopolymer applications for various food products. Table 2 shows the packaging applications of biopolymers as food packaging.

CONCLUSION

Biobased packaging materials have the potential to reduce the use of conventional plastic in the food industry. Bioplastics have some mechanical and barrier properties limitations, which can be improved by physical and chemical modifications in biopolymers. The use of nanoparticles, crosslinking agents and specific additives yields desirable

results in the production of bioplastics. The development cost of bioplastics is high and it lacks the benefits of economies of scale. Bioplastics will be used more in the food industry, agriculture and pharmaceuticals in the coming years. There is a need and opportunity for research and development of bioplastics containing various biopolymer blends.

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