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BIOPLASTICS FROM NATURAL RENEWABLE POLYMERIC RESOURCES: A REVIEW

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Abstract. Petroleum-based plastics are durable, flexible, cheap, and widely available, thus remain increasingly in demand by the growing global population. However, being non-biodegradable, conventional plastics (especially single-use products and materials) end-life scenarios pose continuous threats to the environment, including animal and human health. An estimated 20 million metric tons of disposable plastic litter are introduced into the environment annually. Despite recent global initiatives, recycling rates remain low due to underdeveloped infrastructure and a lack of international standardization. Only about 9 % of plastic waste has been recycled globally, primarily by mechanical recycling, and around 12 % is incinerated (quaternary recycling). About 79 % of the annual production volume of petroleum-based plastics, generated by both developing and developed countries, end up in landfills and oceans globally. Being manufactured from different natural renewable polymeric resources, bioplastics, as sustainable alternatives, have several advantages over their commodity fossil-based counterparts. In particular, bioplastics contribute to lowering carbon footprint, may show valuable and unique thermomechanical and physical properties and performance, are versatile, energy-efficient, and, most importantly, often possess inherent biodegradability. This review discusses the bioplastics from selected plant-derived biopolymers - celluloses, starch (and their derivatives), and plant proteins. Chemistry, advantages, and challenges, as well as some applications of resulting polymeric materials thereof, are assessed.

Keywords: bioplastics, renewable resources, sustainability, biodegradability, celluloses, starch, plant proteins.

1. Entering the Age of Bioplastics

Sometimes, historical periods are called by the name of explored materials that changed the lives of humans, such as stone, bronze, iron, glass, steel, plastic, or silicon ages. The first Industrial Revolution (1760-1840), which provided machinery and tools (operating on steam and coal), enabled the second Industrial Revolution (1870-1919) to happen. The latter is undoubtedly defined by a breakthrough in oil well drilling/refinery techniques, which gave access to numerous petroleum derivatives, thus allowing energetic and chemical industries to boom. Exploring petroleum hydrocarbon structures and revealing their potential in application as monomeric units further opened doors to the world of synthetic polymers. It still can be argued, though, which point of polymer recognition on history pages can be considered the first: the vulcanization of natural rubber by Charles Goodyear in 1830, modification of cellulose into Celluloid thermoplastic by John Wesley Hyatt in 1866, or the first synthetic thermoset - Bakelite, obtained from petroleum-based phenol and formaldehyde by Baekeland in 1907.[1]

Nevertheless, the latter exploration is considered a starting point of the Plastic Era, during which polymers became recognized as prominent and iconic materials, but unlike other historically significant breakthroughs, their reputation was compromised. Their popularity is explained by the ability to provide unique properties in one product, which cannot be attained with other manufacturing materials such as metals, wood, ceramics, and glass. The main benefits are: cost-effectiveness of production; lightweight (reduces energy consumption during transportation); design flexibility (can be shaped into very complex geometries); simultaneously strong and tough (can withstand mechanical damage with prolonged integrity); corrosion/water/chemically resistant (usually inert for interaction with the environment).[2,3] From electronics and packaging to medical and agricultural markets, the universal use of plastics has continued growing ever more quickly.[4]

Following the commercialization of Bakelite, synthetic polymer market was extremely rapidly filled with: polyvinylchloride (1926), melamine-formaldehyde resins (1930), polystyrene (1931), polymethyl methacrylate (PMMA) (1933), polyurethanes (1937), polyethylene and Nylon-6 (1939), unsaturated polyesters (1942), epoxy resins (1946), polypropylene (1957) and polycarbonates (1960).[1] All these polymers haven't lost

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any recognition or importance till nowadays, growing their annual worldwide production from 1.7M tons in 1950 to 390.7M tons in 2021.[5] With this production capacity, synthetic polymers have found their application everywhere (Fig. 1), integrating into our everyday lives so that we no longer perceive them as anything special.

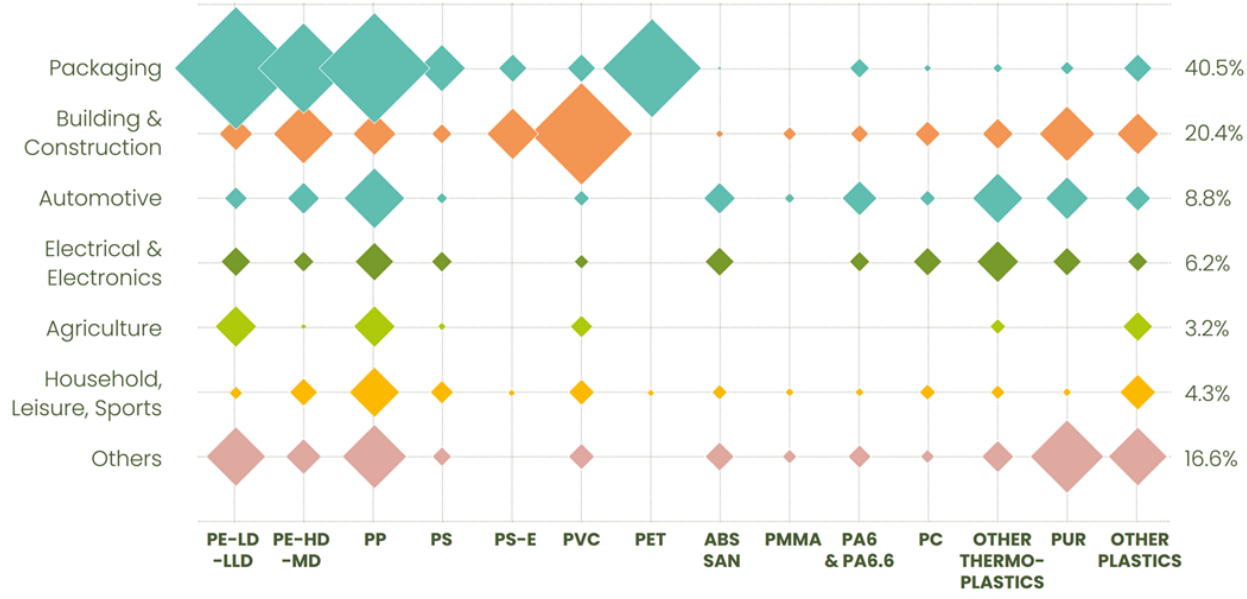


Fig. 1. Commodity petroleum-based plastics, their consumption volumes, and main application areas in the European Plastic Market in 2020 [6]

Despite all the advantages mentioned and with respect to their history, petroleum-based plastics are already at the beginning of the end of their triumph. Being overwhelmed with success and synthetic plastics performance, society forgot to answer two questions: what is the cost of performance to our environment, and what should we do with polymers that are no longer used? The primary reason these questions have been raised is that we are facing the consequences of the non-biodegradable nature of synthetic polymers, linear production economy, "dirty" manufacturing, and the gradual depletion of petroleum as both a material and energy source.[7] Hydrophobicity, chemical structure, crystallinity, and high molecular weight - all that benefit synthetic polymers' application inherently lead to the fact that their carbon content cannot be converted by living organisms to CO_2, H_2O, methane, or biomass.[8,9] Around 79 % of the abovementioned annual production volume remains landfilled, threatening the ecosystem and human health.[5] Indeed, in 2018, the United States generated 35.7 million tons of plastic, of which nearly 27 million tons were disposed of in landfills, which makes up nearly 19 % of all municipal solid waste in the United States.[10] Of the total produced plastics, 14.5 million tons are attributed to packaging materials such as bags, sacks, and pouches, contributing to a significant portion of the total generated and landfilled plastics. Such numbers are alarming and further highlight the need for biodegradable bioplastic alternatives, especially for single-use applications of plastics such as packaging and food packaging, which make up a large portion of the plastic waste in landfills. The hydrocarbon chain of petroleum-based plastics can be considered biochemically inert, thus, the first step concern is the leaching of additives from polymeric matrices (plasticizers, fillers, pigments), which are typically toxic and harm water and soil systems.[11] Following that, hydrophobic surfaces of synthetic polymers can accommodate the growth of heterotrophs, autotrophs, predators, pathogens, and symbionts.[12] Under mechanical stresses and UV exposure, over time, the initial dimension of plastic waste is converted to powder form (microplastics, nanoplastics), where an extensive surface area can facilitate even more significant amounts of microbes and toxins. Due to their size, microplastics can quickly get into soil, water, and animal cells, harming the whole food chain system and, therefore, human health.

The problem of non-biodegradable nature could be significantly relieved if governments and manufacturers would develop more recycling facilities. However, the limited ability to sort plastics results in material heterogeneity; thus, similar recycling conditions cannot be applied, while cross-contamination of plastic wastes with organic matter and inert materials makes recycling costly and energy-intensive. Also, plastics can undergo very

limited use cycles while preserving mechanical properties. Therefore, incineration is most likely a way to eliminate waste. On the other hand, incineration of plastic waste is a primary source of air pollution, releasing furans, dioxins, polychlorinated biphenyls, and mercury into the environment as well as greenhouse gases.[13,14] Not currently in alert mode, but concerns about petroleum-based plastics also touch areas of pollution emission and energy source depletion. Nowadays, 10 % of extracted oil contributes to the worldwide production of synthetic polymers, while during the extraction of raw materials and manufacturing, 2 % of global CO_2 is released. The latter, however, indirectly, can rapidly progress due to plastic waste accumulation that kills animal and plant species responsible for CO_2 capture.[15]

Based on the above-reviewed issues, it seems that developing polymeric materials from renewable sources, but most importantly, those of a biodegradable nature, is the best thing society can do to prevent global pollution collapse. Bioplastic products are designed to be bioderived and perform similarly to petrochemical plastics, thereby reducing dependence on finite virgin resources and decreasing the environmental impact.[16] Even though the demonstration and recognition of macromolecule phenomena happened only in 1920, the application of polymeric materials (which ironically were derived from natural and renewable sources) can be traced back to history.[1] Nowadays, we can observe a historical loop when breakthroughs and accumulated knowledge can help us to re-explore the benefits of natural sources for biobased polymers. When talking about bioplastics, terminology confusion should be avoided. Bioplastics are polymeric materials produced from biological materials or originate from renewable sources and can be either biodegradable or not, while biodegradable plastics can originate from renewable or fossil feedstock (Fig. 2A).[17]

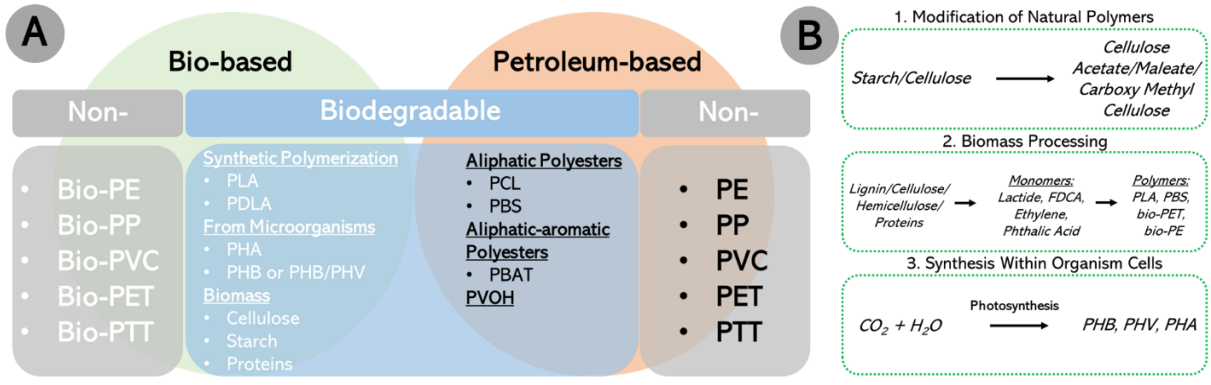


Fig. 2. Categorization of bioplastics based on origin and biodegradability potential (A); Principal routes for bioplastic production (B)

Fig. 2B (Case I, III) shows that both plants and microorganisms provide us with ready macromolecules capable of further modifications to yield processable products. Additionally, the processing of the same macromolecules can be used to derive monomers applicable for polymerization *via* chain/step growth mechanisms (Fig. 2B Case II).[18] Implementation of bioplastic shouldn't be considered a panacea, but their production can indeed address the drawbacks of petroleum-based polymers, at least partially. The first and most apparent advantage of bioplastics is their origin from renewable feedstock that cut a portion of petroleum usage as a source of materials. However, manufacturing bio- or petroleum-based materials still requires energy, which currently comes from oil fuel; thus, only transfer to alternative energy sources can solve the petroleum feedstock depletion issue.[7] Nevertheless, commercialized bioplastics such as polylactic acid (PLA), polyhydroxybutyrate (PHB), and thermoplastic starch (TPS) are reported to require significantly less energy during extraction and manufacturing than low-density polyethylene (LDPE), polypropylene (PP), polystyrene (PS), and high-density polyethylene (HDPE).[19]

The topic of biodegradability and reduced greenhouse gas emissions remains tricky and is sometimes even used for advertising. As can be seen, polymers derived from both fossil and renewable feedstock can be biodegradable, meaning that origin does not define this property. During the development of biodegradable polymeric materials, one needs to consider that the main polymeric backbone should incorporate heteroatoms to form cleavable bonds *via* enzymatic and non-enzymatic hydrolysis.[20] In both cases, for synthetic and biobased polymers, the emission of gases is inevitable; however, for the latter, released CO_2 is adsorbed and stored as energy by plants, which is further used for bio-plastic production. Further, the idea of "emission-free" manufacturing is valid only when bioplastics undergo composting end-life

management to form a net-zero CO_2 cycle; otherwise, if landfilled, bioplastics have similar to synthetic polymers harming effect.[21] It is important to emphasize that biodegradation is only possible when bacteria can access oxygen and decompose waste to yield CO_2 and water. Solid waste, however, is merely dumped, forming layers of old and new plastics, greatly restricting oxygen access to the bottom layers. This triggers bacteria to switch to the anaerobic regime of decomposition, which releases methane by-products 25 times more potent in affecting the greenhouse effect than CO_2.[22] Additionally, the degradation of bioplastics *via* the environmental route also converts the initial size of waste to microplastic form, which has no difference in adverse effects compared to synthetic counterparts. Incineration is also possible; bioplastics have a calorific value similar to wood but still much lower than petroleum-based commodities, as well as air pollution-related concerns.[17] In this regard, composting (biodegradation under controlled conditions) is the best option to make bioplastics truly "green" alternatives, while collected by-products (*e.g.*, methane) can be converted to valuable energy. However, the ability to biodegrade only under specific conditions, without defined timeframes being exposed to the environment, is usually addressed as a drawback. On the contrary, this ensures that under normal conditions during application, no physical aging will occur, and initial properties won't have dynamic values. Thus, instead, the more appropriate question will be: can we control the rate of biodegradation during the design of polymeric products, or do we just have the courtesy to report it?

At this point, the research community has already succeeded in commercializing bioplastics (PLA, PHB, polybutylene adipate terephthalate (PBAT), polybutylene succinate (PBS), and others) (Fig. 2), but unfortunately, they contribute only about 1 % of the globally produced polymeric materials volume.[4] The real reasons for synthetic plastic domination are the high cost of bio-plastic manufacturing (including raw materials and investments in new production lines), competition with the food industry (same crops usage cannot cover the demands of both sectors), intensified farming (need of toxic fertilizers, use of transgenic energy crops, defo-restation) and lack of desired properties such as durability, strength, and barrier performance.[23,24] Bioplastics sound like an ideal solution, but research has yet to develop plant-based materials that demonstrate sufficiently competitive performance and cost compared to petrochemical alternatives.[25,26] Lastly, while we are in the initial steps of developing and commercializing something new - it's the best moment to establish rules and not repeat the mistakes of the past. Before generating industrial bioplastic volumes, we must ensure that recycling and composting facilities are ready to process generated waste. Therefore, it is important to continue researching and exploring more opportunities that nature can provide to address current issues.

2. Overview of Plant-Derived Polysaccharides for Bioplastics Production

Among the sources mentioned in Fig. 2B for bioplastics production, polysaccharides deserve special attention since they are both macromolecular and monomer raw materials. They can serve as food for microorganisms, making them biobased and biodegradable. Already commercialized, they lack many features of their petroleum-based counterparts. Thus, from an academic point of view, it's valuable to discuss in detail what makes them special, which drawbacks limit their performance, and how research can help speed up their journey to large-scale production.

Polysaccharides are a remarkable category of compounds that naturally exist in polymeric form and are the most abundant bio-based macromolecules on Earth. Structurally, they are composed of monosaccharides as monomeric units covalently bonded *via* a glycosidic bond to form a polymeric backbone. The main backbone of polysaccharides can contain anywhere from 40 to 3000 repeating units, while the side chains can have from 0 to 50 repeating units of sugars presented in Figs. 3A and B.[27,28] The length and branching degree of polysaccharides can vary depending on their origin and type, leading to a range of physical properties and applications. As shown in Fig. 3C, polysaccharides can be obtained from renewable sources such as plants/wood, algae, animals, bacteria, and fungi.[29] The production of polysaccharides from the sources, as mentioned earlier, is an industrially established process that includes extraction, separation, and purification steps.

Before the extraction step, raw materials are usually dried and ground into powder form to increase the surface area. Following that, powder soaking in organic solvent (ethanol, ether, petroleum, benzene, chloroform, or hexane) helps to remove lipids from biomass. Extraction further can be done by hot water treatment (for water-soluble polysaccharides - pectin and starch specifically) and alkali or acid solution treatment (for extracellular and non-water-soluble polysaccharides such as lignin, cellulose, and hemicellulose). In the latter method, the presence of either H^+ or OH^- ions helps destroy strong intermolecular hydrogen bonds; however, it can also lead to the degradation of polysaccharides.[30,31]

Having a wide range of sources for extraction, polysaccharides have found many application areas for the

following industries: ***food*** (thickening agents, gelation agents, emulsifiers, and antimicrobial/antioxidant agents);[32] ***pharmaceutical*** (antiviral, anticoagulant, antitumor, and immunomodulatory activities, drug carriers, tissue engineering);[28] ***cosmetics/personal care*** (active ingredients for wound healing and moisturizing effects, anti-aging/microbial/inflammatory agents);[33] textile; agriculture; biodegradable plastics/packaging and more. Combining biobased nature with abundance, biodegradability, sustainability, and cost-effectiveness provides polysaccharides with great potential to substitute currently used petroleum-based synthetic commodities.

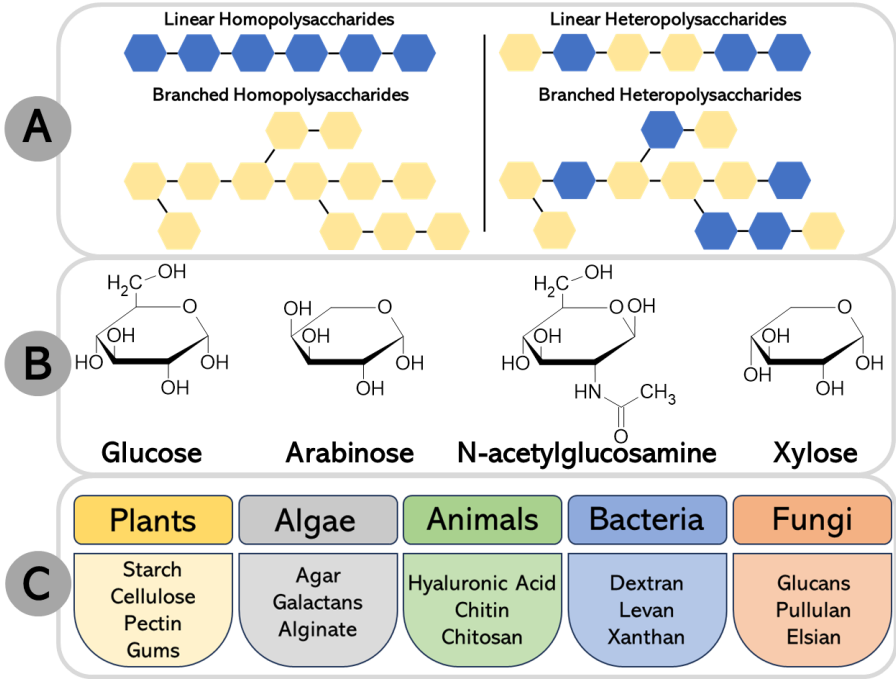


Fig. 3. Schematic representation of possible polysaccharides compositions and architectures (A); Chemical structures of sugars that are commonly found in polysaccharides (B); Natural sources for corresponding polysaccharides (C)

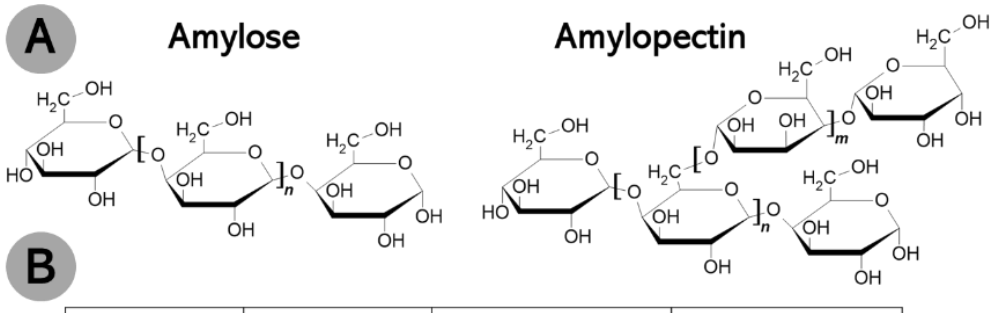
2.1. Bioplastics from Starch

Starch is one of the most abundant polysaccharides on Earth and originates in higher plant cells, where it is synthesized and stored for energy preservation. Naturally, it occurs within the intracellular medium as a spherical, oval, polygonal, disk, or rod-like granules ranging in size between 1 and 100 micrometers. Even though starch is referred to as a single type of polysaccharide, it is indeed comprised of both amylose and amylopectin, the ratio of which depends on the source used for extraction (Fig. 4).

According to monomeric content, both are homo-polysaccharides whose backbone comprises α-(1-4)-D-glucopyranosyl repeating units. As shown in Fig. 4 - amylose has a linear backbone; thus, naturally, it is found in the granules as crystalline domains. On the other hand, the backbone of amylopectin has branches composed of α-(1-6)-D-glucopyranosyl, where the branching degree and branch length depend on the source of extraction.[34,35] The irregular architecture of amylopectin then disables the ability to close macromolecular packing (a precursor for the formation of lamellas), so this portion makes up amorphous regions of starch. It is well known that linear and branched architectures (even with the same composition and molecular weight) lead to different thermophysical performances of macromolecules. Separation of these two polysaccharides into pure forms is not feasible; therefore, raw starch is used as received for studies or production purposes. This brings some complications regarding results/properties consistency as they depend highly on starch composition and will be discussed further. Nevertheless, these polysaccharides have high molecular weights: 0.2–2 million Da for amylose and 100–400 million Da for amylopectin. Combining semicrystalline morphology and high molecular weights makes them great candidates for producing biobased plastics.[36]

Even out of the 1 % of commercialized bioplastics mentioned, thermoplastic starches (TPS) occupy the highest (up to 20 %) portion of production volumes and are mainly used in the packaging industry.[6] Indeed, it's understandable because the use of synthetic structural

plastics that can last for at least several years without losing performance could be excused for causing pollution issues. Flexible/rigid packaging materials usually last for a couple of months and sometimes hours (fast food, pastry, grocery bags, beverage cups/straws, meat/fruits/vegetable liners), thus contributing the most to waste pollution. Besides the production of TPS, starch is widely used as starch derivatives, where hydroxyl groups of native starch are substituted with more hydrophobic functional groups (acetate, hydroxypropyl, *etc.*). Also, combining starch with bio- and petroleum-based polymers; organic and inorganic fillers are often performed to enhance the properties of TPS, such as tensile strength and Young's modulus, and reduce WVP.



Starch Source	Amylose (%)	Amylopectin (%)	Crystallinity (%)
Corn	28	78	43-48
Cassava	16-21	76-84	13
Wheat	18	72	36-39
Rice	22	77	38
Potato	27	74	25-53

Fig. 4. Chemical structure of amylose and amylopectin (A); Sources for starch extraction and its compositions (B)

2.1.1. Thermoplastic Starch

Native starch macromolecules (amylose and amylopectin) contain an extensive system of hydrogen bonds, which must be disrupted to produce TPS. For that purpose, starch undergoes gelatinization, typically in an aqueous solution. Because starch is insoluble in cold water, an increased temperature is required for gelatinization. The dissociation of hydrogen bonds begins in amorphous regions of amylopectin, while cooling the starch solution causes retrogradation. During retrogradation, amylose macromolecules crystallize and form double helix structures. Most of the literature studies on TPS utilize casting from aqueous solutions as a primary method for film preparation. However, on the industrial scale, starch melting and re-crystallization are most commonly performed using extrusion, injection molding, compression molding, or blow molding.[37] A significant advantage of the extrusion process is the possibility of incorporating and homogeneously mixing additives. Like other polysaccharides, starch is extremely rigid and brittle when used alone and is not a true thermoplastic due to intra- and intermolecular hydrogen bonds. Therefore, plasticizers are often added, such as water, glycerol, or sorbitol (sucrose, fructose, glucose, glycols, urea, and amino acids also can be used).[38,39] Plasticizers partially disrupt polymer-polymer interactions and help to increase the flexibility and elongation of the plastic films, but they also reduce their tensile strength and increase water vapor permeability (WVP). Ideal TPS plasticizers must meet several requirements, such as low molecular weight, hydrophilicity, polarity, non-volatility, and high boiling point (needed to survive the TPS processing temperatures). Because the glass transition temperature, T_g of native starch is higher than its degradation temperature, adding plasticizers effectively lowers T_g and significantly enhances TPS processability. However, one should be aware of the possible adverse effects of plasticizers – antiplasticization effect and phase-separation. Even though the antiplasticization effect is not fully understood, it occurs at a critically low plasticizer content, decreasing TPS film elongation but increasing modulus. The phase separation occurs at critically high plasticizer content when the plasticizer-rich phase forms and starch macromolecular interactions reduce, causing a decrease in modulus.

The starch source also dramatically affects the processing parameters and properties of TPS: higher amylose content requires higher extrusion temperature because of high crystallinity and, thus, melt viscosity, but the tensile strength of such material is expected to be higher because of more ordered morphology.

In summary, TPS has satisfactory mechanical performance but is much lower than conventional petroleum-based thermoplastics such as PE or PP. In addition, TPS is a highly hydrophilic material with poor WVP but good O_2 barrier performance (at low plasticizer content). The advantages of TPS are its biodegradability, high abundance of native starch, biocompatibility, non-toxicity, and ability to tune the properties with additives. The WVP can be effectively reduced by including hydrophobic additives (waxes or fatty alcohols) or developing multi-layered materials.

2.1.2. Starch Derivatives

Despite its many intrinsic advantages, such as biodegradability, biocompatibility, film-forming properties, and sufficient mechanical performance, TPS has high hydrophilicity and, thus, poor water vapor barrier performance. Reactive hydroxyl groups of starch can be used in esterification reactions to reduce its WVP and enhance its mechanical properties. The chemical structure of native starch and various starch derivatives is represented in Fig. 5.

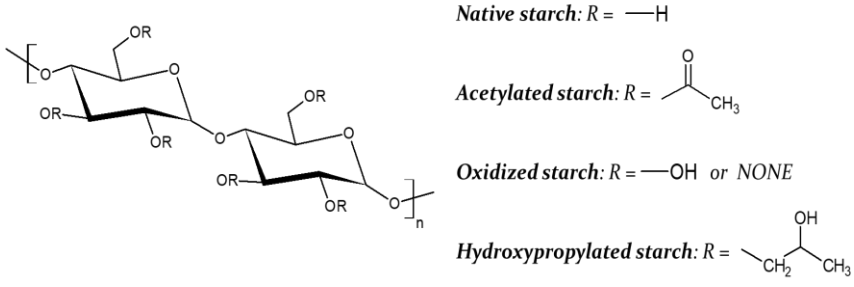


Fig. 5. Structural formulas of native starch and starch derivatives

Starch acetate (SA) can be obtained by reacting native starch with acetic acid and acetic anhydride. The degree of substitution (DS) dramatically affects the properties of SA and defines its further use. The low DS (less than 0.2) SA is a highly hydrophilic material primarily used in the food industry as a thickener and texturizing agent and is approved by the FDA.[40] High DS (0.6-2.9) significantly improves SA hydrophobicity.[41] Because of increased hydrophobicity, lower WVP, biodegradability, and film-forming nature, SA with high DS is used to make edible coatings to enhance food storage. In another study, SA was coextruded with native starch, and the optimum native starch content was 46 %.[42] The lowest water absorption index and mechanical energy for extrusion were obtained at this concentration.

Oxidized starch (OS) is obtained by substituting hydroxyl groups for carbonyl or carboxyl groups, which increases its hydrophobicity, although not as significantly as in the case of starch acetate. Starch can be oxidized using hydrogen peroxide, hypochlorite (the most used industrially), chlorate, permanganate, periodate, dichromate, persulfate, and ozone.[43] It has excellent film-forming properties and can be used for textile finishing and improving paper printability.[44] The high degree of oxidation (over 56 %) disrupts starch crystallinity and makes it amorphous while lowering molecular weight and thermal stability.[45] OS with a high oxidation degree can be used as a plasticizer in TPS production instead of water or glycerol. Incorporation of 5-10 wt. % OS effectively improves TPS toughness and rheological properties while preventing its leaching from the final product.[46]

Hydroxypropylated starch (HPS) is prepared by the reaction of native starch with propylene oxide. The crystallinity of HPS remains similar to that of unmodified starch. However, HPS has decreased retrogradation efficiency, lower T_g, higher water swelling, and solubility.[47] Although hydroxypropyl groups are more hydrophobic than hydroxyl groups, the higher water solubility of HPS can be explained by disrupting the original system of hydrogen bonds and the effect of plasticization. Reduced retrogradation (amylose crystallization) is also explained by the induction of large hydroxypropyl groups with subsequent decrease in intra- and intermolecular interactions. A decrease in retrogradation is particularly important for the properties of HPS gels, which show lower turbidity and significantly higher freeze-thaw stability than native starch gels.[48] HPS also possesses excellent film-forming properties, and it was shown that the starch source doesn't affect tensile strength and elongation.[49] On the contrary, the HPS films from potato starch lose their flexibility during storage compared to wrinkled pea HPS films, which remain flexible even after 12 months of storage. It is important to emphasize that HPS films show lower tensile strength than native starch due to reduced inter- and intramolecular interactions and higher WVP than starch acetate.

2.1.3. TPS Blends and Composites

Combining TPS with hydrophobic polymers and fillers benefits its mechanical performance and water resistance. For example, TPS can be blended with syn-

thetic biodegradable polymers,[50] such as polylactic acid (PLA), polycaprolactone (PCL), polyvinyl alcohol (PVA), polyhydroxybutyrate (PHB), polybutylene succinate (PBS), and polybutylene adipate-*co*-terephthalate (PBAT) or synthetic non-biodegradable polymers, such as PE, PP, PS, terpolymer of acrylonitrile, butadiene and styrene (ABS).[51] However, because of the high hydrophilicity of native starch, its blending with hydrophobic counterparts is challenging. These blends are thermodynamically immiscible, and phase separation occurs due to poor interfacial adhesion.

Four main approaches are identified to provide compatibilization of starch and PLA blends: chemical cross-linking, amphiphilic bridging, componential modification, and interfacial transitioning.[52] The first method utilizes a coupling agent (for instance, diisocyanate) to covalently attach starch macromolecules to the hydrophobic polymer. Amphiphilic bridging relies on adding amphiphilic compounds that have an affinity to both TPS and hydrophobic counterparts (most commonly, grafted polymers). Chemical modification of TPS (acetylation) increases its hydrophobicity and, thus, compatibility with hydrophobic counterparts. Lastly, interfacial transitioning uses the third component (PCL, ABS, *etc*.) to create a new transition phase and improve interfacial adhesion. As such, it was demonstrated that starch can be compatibilized with PLA using diisocyanate cross-linking, peroxide cross-linking, or the incorporation of amylose-grafted-PLA.[53] The latter two methods showed a 60 % increase in TPS tensile strength without a loss in elongation. At the same time, it is essential to emphasize that the type of starch used to prepare the blend may significantly affect biodegradability. It was previously shown that composting the blends of acetylated starch and PCL results in only 25-30 % biodegradation, while blends of native starch and PCL degrade entirely.[38] This can be attributed to the much higher hydrophobicity of acetylated starch and the lower access of microorganisms to the materials.

Starch has also been extensively studied in combination with inorganic fillers, such as clay nanoparticles, montmorillonite, graphene oxide, calcium carbonate, and natural fibers (various cellulose modifications) to prepare composites. The primary purpose of incorporating inorganic fillers is to enhance mechanical and barrier performance. The effect of the filler depends on the particle size, the aspect ratio, and its distribution within the matrix.[54] Two parameters are essential to achieve high interfacial area and interfacial adhesion: affinity of filler to matrix material and its homogeneous dispersion. They are closely related as affinity enhances the dispersion of fillers within the matrix. To increase compatibility, inorganic filler material often requires surface modification, which introduces functional groups or moieties that can interact with the polymer. A significant advantage of graphene oxide as an inorganic filler is the presence of functional groups (carboxyl, hydroxyl), which provides high compatibility with starch macromolecules.[55] Inorganic fillers increase the mechanical performance of the polymer-based films because of the stress transfer from the matrix to the filler, ultimately leading to higher tensile strength and Young's modulus. However, there is a limit filler loading, exceeding which particles tend to aggregate and reduce the mechanical properties of the composite.

At the same time, the presence of inorganic fillers significantly reduces water vapor and gas permeability in starch-based composites. Moreover, the decrease in particle size further enhances barrier performance by blocking the passages for water and gas molecules. Similar to the effect on mechanical performance, aggregation of the filler causes an increase in permeability due to the formation of large voids and diffusion of water molecules.

2.2. Bioplastics from Celluloses

Celluloses are the most abundant biopolymers found in nature and the main constituent of the plant cell walls. It typically constitutes about 40 % of carbon content in plant fibers, accompanied by hemicellulose and lignin. Plant fibers are the largest source of cellulose, but it can also be produced by fungi, algae (green, gray, red, yellow-green), bacteria (*Gluconacetobacter xylinus*), and some marine organisms (Tunicate, also known as "sea squirts"). To use cellulose for further modification, cellulose from plant fibers must be separated from the other components. Cellulose is typically extracted in a process called sulfate cooking (Kraft process), yielding wood pulp for the paper-making industry. Then, only about 2 % of wood pulp is processed into a dissolving pulp (93 % of cellulose) with the highest content of cellulose, while the remaining matrix components (hemicellulose, lignin) partially degrade and are typically used as an energy source.[56] The preference is given to cellulose because of its well-defined chemical composition, high crystallinity, reactivity, and high elastic modulus of cellulose nanoparticles. Independent of the source, it primarily consists of glucose monomer units linked by the β-(1-4)-glycosidic bonds. A highly developed system of hydrogen bonds in the cellulose structure leads to its unique properties – limited solubility in most solvents and high crystallinity. While β-(1-4)-glycosidic covalent bonds together with intramolecular hydrogen bonds make cellulose stiff and rigid, intermolecular hydrogen bonds provide the assembly of cellulose macromolecules into fibrils and their parallel stacking into layered structures.

In the natural state, the basic cellulose structure consists of fibrillar units that consist of nanofibrillated cellulose (NFC) and are further arranged into microfibrils.[57]

Microfibrils consist of crystalline domains called cellulose nanocrystals (CNC), connected by the amorphous regions. Purified cellulose is highly crystalline because the amorphous part of the cellulose is dissolved during hydrolysis. Processed cellulose can exist in various nanostructures, such as microcrystalline (MC), microfibrillated cellulose (MFC), and cellulose whiskers. Microcrystalline cellulose is obtained during basic aqueous solution of biomass treatment with further acid hydrolysis. The obtained product has a DP of 25-300, is crystalline, and is larger than the initial microfibrils. Cellulose whiskers are formed by hydrolysis (acid or enzymatic) and subsequent ultrasonication. They resemble rodlike particles (nanocrystals) of a few nanometers wide and hundreds of nanometers long. Microfibrillated cellulose is obtained by applying the high shear force to the wood fibers, which causes their cleavage and reduction into fibrils, microfibrils, and fibril aggregates. Ultrasonication and enzymatic hydrolysis allow the control of their size and decrease up to the nanoscale.

Despite possessing high elastic modulus, low density, and low thermal expansion coefficient, native cellulose has several disadvantages to be used solely for polymer production. First, cellulose has extremely low solubility in water and most organic solvents, which prevents it from effective dispersion in the medium, while its high hydrophilicity causes phase separation with polymers, which are typically highly hydrophobic. Secondly, it is non-film forming, which requires either modifying hydroxyl groups to provide film formation ability or mixing with the polymer capable of film formation.[58] Cellulose can be used for its homogeneous modification *via* the reaction of hydroxyl groups (3 OH groups per glucose unit) or as a reinforcing filler in polymer matrix composites (Fig. 6).

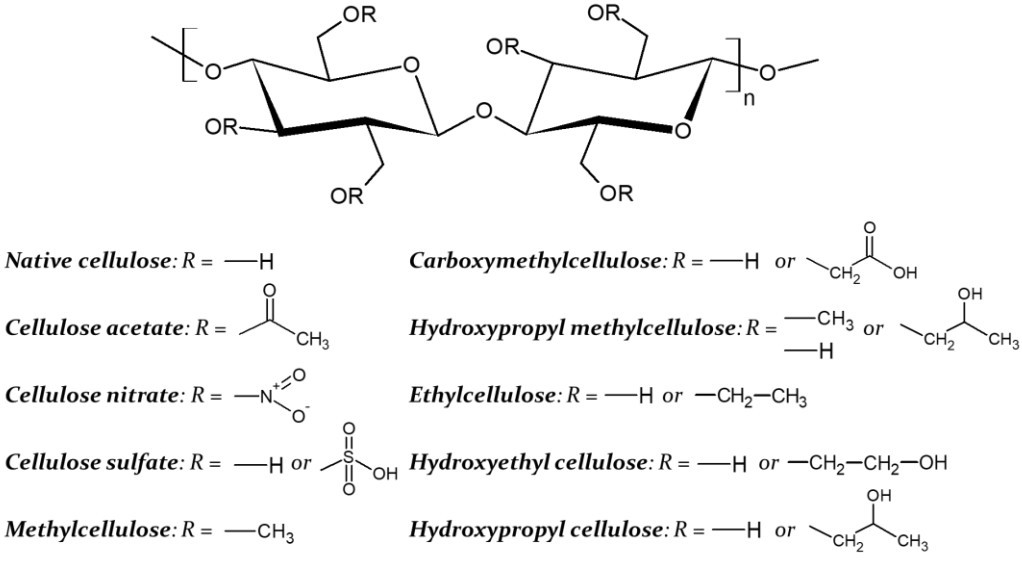


Fig. 6. Structural formulas of native cellulose and cellulose derivatives

2.3. Cellulose Derivatives

One of the examples of cellulose modification is *cellulose acetate* (CA). CA is obtained by the reaction of cellulose with acetic anhydride and acetic acid with a sulfuric acid catalyst, after which it becomes soluble in acetone, dioxane, methyl acetate, and acetic acid.[59] CA is readily biodegradable by microorganisms that use cellulase enzymes, and upon partial deacetylation, the cellulose polymer backbone can be degraded.[60] Also, photodegradation can occur due to exposure to UV light with wavelengths shorter than 280 nm but not as readily degrades in daylight. CA is widely used to prepare coatings, films, textile fibers (5.1 GPa modulus, 20 % elongation), membranes, and photo films, depending on its degree of substitution.[56,61] *Cellulose acetate-butyrate* (CAB) and *cellulose acetate-propionate* (CAP) are used to prepare membranes for ultrafiltration and water desalination due to the high tolerance of CAB to Chlorine, which is often used in water purification systems, and resistance to bio-fouling compared to pure CA.[62,63]

Cellulose nitrate (CN) was one of the first semi-synthetic polymers and immediately found wide application in coatings, films, explosives, and inks, depending on the substitution degree of hydroxyl groups with nitrate groups.[64] However, its high heat and light sensitivity significantly reduces its use in cinematographic films and photography. The reason for CN film degradation is extensive denitration with further scission of cellulose backbone and ring disintegration catalyzed by iron and iron-nitrogen oxide.[65]

Cellulose sulfate (CS) is a water-soluble cellulose derivative synthesized by sulfation in sulfuric acid, sulfur

trioxide, or chlorosulfonic acid. It demonstrates antibacterial and anticoagulant properties and can be used for drug delivery.[66,67] CS was also used to prepare edible and biodegradable films to prolong the shelf-life of fruits.[68] The water-soluble films demonstrate tensile strength up to 24 MPa and elongation at break up to 42 % depending on the glycerol content and high oil resistance, as well as significantly preserving bananas during storage for up to 15 days.

Methylcellulose (MC) is a water-soluble cellulose derivative (at higher DS, it dissolves in some organic solvents, and at DS=3, becomes insoluble in water) obtained by methylation with methyl chloride or dimethyl sulfide. Upon reaching a lower critical solution temperature (LCST) or 29±2ºC, it forms a thermo-reversible turbid gel.[69] At temperatures above 42.5ºC (varies with MC molecular weight, concentration, and heating rate), the phase separation occurs with the formation of a strong gel.[70] MC is Generally Recognized As Safe (GRAS) by the FDA and is widely used in the food industry as a thickener, protective colloid, and gelling additive.[71] It is non-allergic, non-toxic, and non-digestible, therefore it is widely used in biomedicine and cosmetic products. MC's low oil absorption and film-forming ability allow the production of films for food preservation. In addition, it can be used as an adhesive and viscosity control additive in cement- and gypsum-based materials, enhancing workability and adjustment time.

Carboxymethyl cellulose (CMC) is an anionic cellulose derivative synthesized by reaction of cellulose with monochloroacetic acid. Its solubility and rheological properties largely depend on the DS, molecular weight, and purity, which also define the final application – food grade, pharmaceutical grade, or industrial grade. The solutions of CMC exhibit shear thinning and pure viscous behavior at lower concentrations, while at concentrations above 4 wt. % linear viscoelastic behavior is observed.[72] Additionally, CMC is known for its time-dependence behavior (thixotropy), which is more pronounced at higher concentrations, attributed to the dominant disentanglement of the macromolecules over re-entanglement.[73] Because CMC is non-toxic, non-digestible, and has an excellent price-to-performance ratio compared to other analogs, it is widely used in many applications: thickener and sizing agent for textile printing; thickener, emulsion stabilizer and water retaining agent for food products; adsorbent for water treatment process; tissue engineering, drug delivery, 3D bioprinting and bioimaging in biomedical applications.[74,75]

Hydroxypropyl methylcellulose (HPMC) is a nonionic polymer soluble in cold water, which, similarly to MC, can form a thermoreversible gel at increased temperatures (50-80ºC). The gelation of HPMC happens due to polymer-polymer hydrophobic interactions in macromolecules containing methoxy groups, the same as in the case of MC.[76] As the temperature increases, partial dehydration occurs, enhancing polymer-polymer interactions and ultimately leading to gel network formation and viscosity increase. The HPMC's degree of substitution significantly affects the cloud point, onset of gelation, and viscoelastic properties of the gel.[77] It was observed that a higher content of hydroxypropyl groups results in weaker gels and lowers the temperature of the onset gelation. HPMC is a non-toxic, biocompatible, biodegradable, film-forming polymer that obtained GRAS approval from the FDA and is often used to make edible packaging for food preservation and as a thickener.[78] It has good oxygen barrier properties and oil resistance but high water vapor permeability (WVP), thus it is often mixed with additives (for instance, wax) that reduce WVP.[79,80]

Ethyl cellulose (EC) is obtained by reacting cellulose with ethyl chloride. EC is non-toxic, non-digestible, and dissolves in many organic solvents at high DS. The EC solubility depends on the DS – water-soluble at lower DS (0.8-1.3) and soluble in organic solvents at higher DS (2.2-2.6), but becomes insoluble in water.[81] Oil-soluble EC is widely used to prepare oleogels in the food or biomedical industry *via* the anti-solvent method or Pickering emulsion technique.[82] EC is too brittle to prepare the coating or use it as a binder in microcapsules and microspheres and often requires plasticizers to improve processability, flexibility, and thermal stability.[83,84]

Hydroxyethyl cellulose (HEC) nonionic water-soluble cellulose derivative obtained by reaction of alkali cellulose with ethylene oxide. It is readily soluble in water at any temperature and can be used as a thickener in food, a sizing agent in papermaking and the textile industry.[85] HEC is odorless, tasteless, non-toxic, has good water-retaining properties, and is film-forming. Because of its biocompatibility, HEC can be used to prepare hydrogel membranes for wound healing, which can contain antimicrobial agents.[86] The non-adhesiveness of HEC hydrogels ensures easy removal upon wound healing while reducing the toxicity of antimicrobial agents for human cells by suppressing the abnormal immune response. In another study, adding HEC into Portland cement increased its strength, hardness, and fracture toughness. Moreover, reduced porosity provided higher corrosion resistance due to decreased water absorption.[87]

Hydroxypropyl cellulose (HPC) is a cellulose derivative synthesized from alkali cellulose and propylene oxide and is soluble in water and alcohols, such as methanol, ethanol, and isopropanol. It dissolves in cold water but forms a thermoreversible gel above 45ºC, similar to MC and HPMC.[88] Also, phase separation occurs at constant temperatures for solutions over 40 % concentration.[89] HPC forms cholesteric liquid crystals (CLCs) in water solution at concentrations over 55 % and

various organic solvents.[90,91] The formation of the cholesteric phase (arranged in a helical structure) is explained by the chirality of anhydroglucose repeating unit. HPC has found the most widespread use in biomedical applications as a lubricant for artificial tears for people with insufficient tear production.

Xu *et al.* prepared transparent cellulose-based films by mixing microcrystalline cellulose with $ZnCl_2$ and $CaCl_2$.[92] Zn^{2+} ions are assumed to disrupt hydrogen bonds within cellulose fibrils, while Ca^{2+} ions provide cross-linking upon film formation. The obtained films possess a tensile strength of up to 88.2 kN/m^2 and may find potential use in pharmaceutical, biomedical, and packaging applications.

Similar to starch-based materials, cellulose can be combined with bio- and petroleum-based plastics and organic and inorganic fillers to prepare cellulose-based composites. The authors refer the readers of this review to the recent works on cellulose composites.[93-95]

3. Overview of Protein-Based Bioplastics

It wasn't until the mid-1900s that large-scale extraction of vegetable oils and proteins was first introduced.[96] However, since the introduction of plant oils and proteins, their uses have become essential to everyday life.[96,97] Soybean, canola, and corn are some of the most popular crops that now undergo large-scale extractions to produce their respective oils and protein isolates.[98-100] While uses of each vegetable oil have been thoroughly explored and alternative applications identified, less work has been done to explore the material applications of these plant proteins.[101-103]

3.1. Soybean Protein and Zein in Food Packaging Materials

Although there has been commercial and public interest in alternative uses of soy protein isolates, further investigation is required to develop applicable industrial materials.[104,105] Soy protein use as an additive has been found beneficial in numerous applications such as edible food technology, adhesives, and wood composites.[106,107] Soy protein-based materials are more limited to film applications such as food packaging. Indeed, previous work highlights the promising film-forming ability of soy protein isolate.[108,109] Films prepared using soy protein demonstrate smooth, transparent films with high flexibility. However, these films are weak and do not perform well as water barriers due to the hydrophilic exterior of soy protein. To mitigate some of these performance concerns, researchers have explored using various modifiers, including nano-additives such as nanocellulose,[110] nanochitosan,[111] or nanoSiOx.[112] While such additives enhance film performance, they do not address the need for cost-effective alternatives to petroleum-based plastics.

Likewise, zein from corn has received much focus as a material resource due to its fewer competitive applications and potential for greater strength.[113] Although less widely used as compared to soybean, the applications of corn oil and corn protein have seen extensive investigation.[100,114,115] Such work has again highlighted zein from corn as a viable resource for food packaging preparation. Although zein protein is a more hydrophobic material and its physicochemical properties contribute to bioplastic products that demonstrate promising strength and improved barrier performance,[113] there are additional challenges that must be addressed. Previous studies have illustrated the poor flexibility of films prepared from corn protein, which prevents further advancement of zein-based industrial materials.[116,117]

Obstacles associated with protein-based plastics include limited mechanical properties, poor water barrier performance, time required for film preparation and characterization, and cost-effective resource availability.[23,25] Although extensive work has been done studying proteins as a material for bioplastic products, this work is time and resource-consuming.

3.2. Composite Polymeric Materials from Soy Protein and Zein

After discussing the challenges that come from using either soy protein or zein as a base material for food packaging films, it can be noted that there is potential to provide synergistic performance by eliminating each protein's specific disadvantages if applied simultaneously with a counterpart in preparing the bioplastic materials. Indeed, soy protein films are soft, flexible materials with poor water resistance,[108,109] while zein films are brittle films with greater moisture resistance.[116,117] Thus, there is a desire to combine the two proteins and prepare a bioplastic film from both materials. In doing so, biobased food packaging films may be achieved that have optimum mechanical and barrier properties. The previous research has explored the possibility of combining multiple plant proteins to prepare enhanced bioplastic films,[118,119] however, several challenges must be addressed.

The primary obstacle faced when investigating soy-zein composite materials is the concern of solubility. As most film formation studies utilize a solution-casting approach, the proteins must be soluble (or dispersed) in the same solvent system. However, in the case of soy protein and zein, the two proteins share no common solvents.[120] While soy protein is readily soluble in water,

zein will coagulate when introduced to a 50 % water solution.[121] Although Evans and Manley identified more than 50 solvents that can be used to dissolve corn protein, none of the available identified materials allowed for adequate dissolution of soy protein as well. Despite significant efforts, a common solvent (mixture) has yet to be identified, thus further highlighting the challenge of combining both proteins. For this reason, prior studies have instead investigated "laminated" films, wherein two independent layers are prepared using proteins separately to achieve greater barrier performance.[119,122] However, this approach requires thermal compression to laminate the films. Additionally, studies of this nature are unable to perform surface hydrophobicity measurements. This may be because such laminated materials demonstrate different levels of hydrophobicity on each side of the films, and thus, no number can be reported that is characteristic of the entire material. Thus, preparing a uniform film from multiple plant proteins requires significant changes in processing techniques and limits the methods of characterizations that can be performed and considered valid. In addition to the solubility and processing challenges of combining plant proteins in bioplastic production, cost, time, and resources challenges remain. For this reason, there is a need to effectively optimize the compatibility between soy and zein proteins, allowing for the advancement of bioplastic materials with enhanced mechanical and barrier properties. Patnode *et al*. demonstrated that computational methods, such as protein-ligand docking and quantitative structure-property relationship, are valuable tools for the design of experiments and for developing composite materials with soy and zein proteins.[123] As such, biopolymer composite films were prepared with various plasticizers (glycerol, sorbitol) and microfibrillated cellulose as reinforcing filler. Obtained materials possess high flexibility and Young's modulus of 99-400 MPa.[124]

The demand for biobased and biodegradable plastics is continually growing. Indeed, our society increasingly calls producers to action, asking for more environmentally friendly alternatives to current eco-damaging materials. Specifically, the desire to utilize plant proteins for food packaging products has gained significant interest as proteins such as soy and zein demonstrate good film-forming abilities and promising performance. Despite their favorable characteristics, research faces multiple challenges when preparing food packaging alternatives from plant proteins. One primary obstacle is the cost-performance analysis between bio-based products and their field performance. To improve industrial acceptance of protein-based packaging, the cost of production must be reduced and the properties of the material enhanced.

4. Conclusions

Despite their benefits, such as durability, flexibility, and cost-effectiveness, petroleum-based plastics pose significant environmental threats due to their non-biodegradable nature and fossil fuel depletion. The majority of plastics disposed of end up in landfills or oceans, polluting global soil and water resources and causing adverse impacts on ecosystems and human health.

Over 6.3 billion metric tons of plastic have accumulated in landfills since mass production began in the 1950s. Such tremendous amounts of plastic waste pose significant risks to ecosystems often accompanied by harmful chemicals leaching into groundwater. Over 150 million metric tons of plastics are now present in the marine environment. This accumulation has led to the formation of colossal garbage patches in the oceans, the most infamous being the Great Pacific Garbage Patch, which covers an area twice the size of Texas (1.6 million km^2) and continues to grow nowadays.

On the contrary, bioplastics offer a sustainable alternative. While produced from renewable resources such as cellulose, starch, and plant proteins, they help reduce the carbon footprint, provide unique properties, and are often biodegradable. However, their production faces challenges such as high costs and competition with the food industry for raw materials. Additionally, their mechanical properties usually need to be enhanced to match conventional plastics.

This review covered the production, properties, and potential application of bioplastics from starch, cellulose, and plant proteins. Thermoplastic starch (TPS) and its derivatives show great potential for packaging applications due to its easy processability, diversity of mechanical properties, excellent film-forming ability, and good gas barrier properties. However, starch-based materials have limitations, such as high hydrophilicity, which negatively affects their water barrier performance. Cellulose is the most abundant biopolymer in nature and provides a wide range of properties. Even though it inherently has good mechanical performance, it lacks film-forming properties and thus can be functionalized or used as a reinforcing filler in polymer matrix composites. Cellulose composites and cellulose derivatives are often used in food packaging, hydro- and oleo-gels preparation, drug delivery, water treatment, and lacquers. Soy protein and zein are promising for bioplastic applications, particularly in food packaging, but their mechanical properties and water resistance need further improvement. Moreover, additional studies on compatibilization and synergistic effects between different plant proteins are required.

Future research should focus on overcoming the challenges of bioplastic production, finding cost-effective

manufacturing processes, improving water barrier performance, and incorporating bioplastics into the circular economy by either recycling or composting the used materials. By addressing these issues, bioplastics can become a viable and sustainable alternative to petroleum-based plastics, reducing environmental pollution and synthetic resource depletion.

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БІОПЛАСТИК З ПРИРОДНИХ ВІДНОВЛЮВАНИХ ПОЛІМЕРНИХ РЕСУРСІВ: ОГЛЯД

Анотація. *Пластмаси на основі нафти міцні, гнучкі, дешеві та широкодоступні, тому мають усе більший попит серед населення Землі, яке постійно зростає. Однак оскільки звичайний пластик (особливо одноразові вироби та матеріали) не є біорозкладним, він після закінчення терміну експлуатації створює постійну загрозу довкіллю, зокрема здоров'ю тварин і людей. За оцінками, щорічно в навколишнє середовище потрапляє близько 20 мільйонів метричних тонн одноразового пластикового сміття. Незважаючи на нещодавні глобальні ініціативи, показники переробки залишаються низькими через нерозвинену інфраструктуру та відсутність глобальної стандартизації. Лише близько 9 % пластикових відходів було перероблено в усьому світі, переважно шляхом механічної переробки, і близько 12 % спалюється (четвертинна переробка). Близько 79 % річного обсягу виробництва пластику на основі нафти, виготовленого як у країнах, що розвиваються, так і в розвинених країнах, потрапляє на звалища та в океани. Будучи виготовленим з різноманітних природних відновлюваних полімерних ресурсів, біопластик як стійка альтернатива має кілька переваг перед своїми аналогами на основі викопної сировини. Зокрема, біопластики сприяють зниженню викидів вуглецю, можуть мати цінні й унікальні термомеханічні і фізичні властивості та продуктивність; вони є універсальними, енергоефективними та, що найважливіше, часто мають властиву біорозкладаність. Цей огляд присвячений біопластикам з окремих біополімерів рослинного походження – целюлози, крохмалю (та їхній похідних) і рослинних білків. Оцінено хімізм, переваги та проблеми, а також деякі шляхи застосування таких полімерних матеріалів.*

Ключові слова: *біопластик, відновлювані ресурс, стійкість, біорозкладаність, целюлоза, крохмаль, рослинні білки.*