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Introduction to Natural Materials for Food Packaging

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1.1 Introduction

Food packaging material should also maintain the lifetime of the food by dodging adverse conditions such as spoilage microorganisms, mechanical vibration, shocks, gases, moisture, chemical contamination, bad odor, and exposure to oxygen. Fresh and healthy foods are the everlasting demands of the consumers in the global market after the inception of packaged foods [1, 2]. Bio-based polymers have been the first-choice materials for food packaging applications which not only promote sustainable material development but also overcome environmental concerns causing very less ecological threats. To enhance the shelf life of the food material as well as to uphold its quality as it is from the date of manufacture, plastic-based food packaging materials are used [3]. Muizniece-Brasava et al. [4] stated in their study that as per the recent statistics, an 8% annual increase in production of packaging materials from petroleum-based materials has been seen, but on the other hand, only 5% of those materials were potentially recycled. This resulted in almost million ton plastic packaging materials in landfills each year due to accumulation of non-recycled materials, thus affecting the environment which is the current-day problem on planet earth.

One concern about plastic pollution has motivated the growth of degradable, natural, and green product materials [5, 6]. Combining a biodegradable polymer consequent from renewable sources with a natural fiber filler to create a bio-composite represents a self-sustaining as well as a technically feasible alternative to so-called “commodity” plastic products in the food packaging sector. Three critical factors must be considered. To start, we must reduce our reliance on petroleum-based

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materials while increasing our use of renewable sources to make plastics, thereby reducing the amount of old carbon put into the atmosphere. Second, the use of biopolymers enables the package to be treated similar to an organic biodegradable residue following its use, thereby helping to decrease polymeric trash bound for landfills and incinerators. Eventually, use of natural fabrics as fillers enables the valorization of agricultural residues, thereby reducing the food production cycle's overall impact. Around each other, use of biocomposites composed of biodegradable polymers based on renewable sources and fillers resulting from agricultural fiber garbage as well as other by-products enables more justifiable products by promoting a cradle-to-cradle approach and the life cycle assessment (LCA) [7, 8]. Figure 1.1 shows various materials used in recent days for food packaging.

Green packaging materials made of biodegradable composite are gaining increasing interest in a range of disciplines owing to their distinctive characteristics in comparison to conventional petrochemical-based plastics [9]. Furthermore, they are fully biodegradable and degrade completely including organic material, H_2O, and carbon dioxide. These characteristics may enable their use in diverse applications, including smart nano-food wrapping [10–12], biomembranes for water purification, recycling of waste, as well as drug delivery. Thus, the primary function of packaging material is to enhance the quality and safety of food while extending its life span [13]. Due to their ability to prevent the transmission of humidity, oxidant, and flavors among foodstuffs and their surrounding environment, edible films may be a worthwhile alternative to plastics in a variety of applications [14]. As a result, use of edible coatings for preserving the quality of various foods has grown rapidly [15]. Recently, a variety of biodegradable food packaging materials, including sipping beverages,

Figure 1.1 Various food packaging materials. Source: Sanyang et al. [8]/with permission from Springer Nature.

sheets, silverware, overwrap, as well as lamination films, have been manufactured and distributed through grocery stores [16–18].

Owing to its least cost along with easy accessibility for industries, carbohydrate, a normally sustainable energy fructose polymer, is the most frequently utilized fresh material to produce biodegradable plastics [19]. Other studies have been conducted to determine its great potential in aqua-soluble pouches for storing detergents and insecticides, as well as to determine its utility in washable lining, satchels, and other medical equipment. Starch is composed of two molecules: amylose (a sequential chemical compound with very few branch offices) and amylopectin (a branched chain molecule). When starch is processed, the existence of amylose in significant amounts provides strength to the films. Tensile stress in layers is observed to reduce when amylopectin is a predominant component of the starch. Maize or corn flour is the primary source of starch, accounting for approximately 80% of the global market. Rice starches exhibit a range of characteristics depending on the paddy variety [20, 21], resulting in biodegradable films with a range of characteristics. Rice starches are being used in place of synthetic films to generate biologically decomposable films owing to their low cost, abundant availability in nature, and acceptable mechanical characteristics. However, these rice starch films lack adequate barrier properties against nonpolar compounds, restricting their application. This led to development of rice-based starch films with improved characteristics [22, 23].

Among the various widely viable bioplastics, poly(3-hydroxybutyrate) (PHB) is a particularly interesting member of the hydroxyl alkanoates family for packaging applications. PHB is a plastic material that can be transformed industrially using standard polymer transformation equipment. Additionally, it has a good mechanical result in terms of strength and stiffness that is comparable to or greater than that of some commodities (for example, PP), as well as barrier characteristics (comparable to PET). PHB worsens in composting environments and other surroundings such as saltwater [3]. While PHB is an interesting choice for self-sustaining packaging applications, it does have some drawbacks that limit its widespread use in the fresh produce food packaging sector. PHB has a high intrinsic fragility, which upsurges over time as a result of a second crystal growth and physical aging. Additionally, owing to the high crystallinity, PHB has a narrow handling window, which makes it unsuitable for some prevalent food applications, such as blow molding [24, 25].

Edible coatings are very thin films of material (generally or less than 0.3 mm in thickness) which are used to cover food goods to substitute or strengthen the natural layers. They can be devoured as a product or after further expulsion. As a result, the ingredients used in the composition should adhere to applicable food policies and guidelines. Furthermore, the adhesives and films should not have a detrimental effect on the food product's organoleptic properties. Edible packaging may take the form of a surface-level coating on the meals or constant layers among compartments/ingredients of heterogeneous products (for example, grilled cheese, pastry shop fillers, and toppings) [26, 27]. Additionally, the coating can be given to individual pieces of a larger product that are not being independently wrapped for practical reasons, including nuts, kiwis, fruit, veggies, fresh-cut watermelon, and fruits.

Edible films as well as layering can be used to counter a variety of barriers associated with food marketing. These features can be specified as restraining the migration of moisture, solute, oil, and gas, enhancing structural stability, retentive volatile flavor compounds, and transporting dietary supplements. Additionally, they enhanced the attractive look by reducing physical damage, trying to conceal scar tissue, and enhancing surface glow. For example, citrus fruits have been encased with hot-melt paraffin wax to retard moisture absorption, eatable connective tissue canisters have been used to offer structural integrity to sausages, and fruits have been encased with sealant to shape the way glow and inhibit actual injury [28, 29].

1.2 Natural Biodegradable Polymers

Biodegradable polymers are considered to be the most likely solution for dodging various environmental hiccups like litter, landfills, and waste pollution which originate due to the use of nonbiodegradable polymers. But owing to the cost of processing and the limited range of selection, the utilization of biodegradable plastics is less than expected in various end applications these days [30]. Hence, it was stated in various researches that blend of one renewable and biodegradable polymer with another during the process of preparation reduces the cost and widens the prospects of industrial application. Materials including chitosan, proteins, starch, lignin, and cellulose are some of the prominently known biodegradable elements derived from polysaccharides and natural oil bioresources. Few other materials like PLA, PBH, and PCL are derived from partially biodegradable raw materials and they are also categorized under biodegradable polymers.

Natural oils, which can be obtained from both animal as well as plant sources, are one of the abovementioned potential substitutes for chemical raw materials. Triglycerides are widely used in agricultural chemicals, inks, as well as coatings, according to numerous studies. Many of the above implementations made use of brand-new triglyceride oil polymerization and monomerization methods. Nanocellulose, on the other hand, is a comparatively newly developed type of nanomaterial with superior physical as well as chemical properties that are widely used. The nanocellulose in these materials has the prospect to change the top layer chemistry of the embedded material, making them more flexible, stronger, as well as lighter than conventional nanomaterial [18, 19].

1.2.1 Starch-Based Natural Materials

Starch has been the most extensively as well as frequently utilized biopolymer derived solely from renewable as well as natural resources. Due to starch's low cost, complete biodegradability, and ease of availability, starch-based polymers are mostly in high demand these days. Any biodegradable polymer can be incorporated into thermoplastic starch (TPS) in an attempt to lessen the manufacturing costs of biopolymers. Aside from starch, polysaccharides obtained from plants are the most abundant as well as the renewable class of polysaccharides. Amylopectin and amylose are the two major glucose polymers in starch. Amylopectin seems to be a polymeric chain of D-glucose atomic chains linked together by −1, 4 branched bonds, while amylose is a short −1, 4 connected D-glucose chain made up of atoms with −1, 6 branched bonds. The hydrophilicity, as well as brittleness of starch, make it difficult to use, despite the fact that it is completely biodegradable, low cost, as well as able to generate film-forming components with low oxygen permeability as well as the capacity to be managed easily. As a result, starch-based polymers cannot be used in common applications like food packaging and plastic bag substitutes. Different researchers have used plasticizers like sorbitol, glycerol, and glycol underneath the activity of shear stress as well as heat throughout the extrusion process to transform starch into TPS to resolve the shortcomings mentioned above as well as enhance processing potential as well as flexibility [20–22].

1.2.2 Poly-Lactic Acid-Based Natural Materials

PLA, a biodegradable polymer, is one of the most frequently used biodegradable polymers, along with starch, a significant variant of the aliphatic polyester lactic acid, which is a byproduct from the fermentation of plants like sugar beets as well as corn. This biodegradable polymer, like starch, is cheap as well as plentiful, so it has received a lot of attention from researchers and manufacturers. Additional advantages include its biocompatibility, commercial availability, complete biodegradability, ease of processing as well as high transparency. These are the primary reasons for its widespread use [23]. Lactic acid is typically produced during the petrochemicals or bacteria fermentation. Plastic film (PLA) is created by condensation polymerization of lactic acid or by opening the ring polymerization reaction of lactide monomer enshrined in lactic acid (L or D-lactic acid). Polycondensation of lactic acid is used to produce PLA with a lower molecular weight if necessary. Ring-opening polymerization reaction and also azeotropic polymerization condensation of lactic acid, on the other hand, produce PLA with higher molecular weight and considerate mechanical properties [24].

Several scientists have attempted to make PLA-based biomaterials by reinforcing the PLA matrix along with nanocellulosic substances over the previous decade. The mechanical strength, as well as stiffness of the biomaterials depending on PLA, was increased when the original nanocellulose has been reinforced in the PLA matrix to receive biomaterials depending on PLA [25]. A variety of chemical and physical surface modification techniques, including polymer grafting or derivatization and macromolecule or surfactant coating, have been utilized to improve the compatibility as well as a scattering of nanocellulose fillers within a hydrophobic and non-polar PLA matrix, thereby improving the interfacial characteristics between the filler as well as matrix and the effectiveness of nanocellulose-embedded PLA composites [26–29].

1.2.3 Poly-Caprolactone (PCL)-Based Natural Materials

This polymer is a thermoplastic polymer that has better biodegradability, lower viscosity, better thermal computing ability, as well as the least melting point in the range of 55–60 °C [30]. This is because the interfacial bond formed by the straightforward mixing of chemically inconsistent nanocellulose and PCL fibers in reinforced PCL biomaterials may have led to the observation that nanocellulose could only be reinforced in small percentages with PCL matrix. This incompatibility, which results in poor interfacial properties, has been found to be remedied by surface-modifying nanocellulose fibers, which improve the reinforcement's compatibility with other components of biomaterials. Adding polymer chain surface transplanting straightforwardly to the nanocellulose fiber surface could improve the nanocellulose fibers' bioavailability in a PCL polymer matrix significantly. As a result, modified nanocellulose is an excellent choice for reinforcing PCL-based biocomposite materials [31–33].

1.2.4 Poly-Hydroxy Alkanoate-Based Natural Materials

Biodegradable polyester PHA is drawn from various hydroxy alkanoates via microbial fermentation as well as could be used in diverse applications, such as agricultural, medical as well as packaging industries. PHA begins with hydroxyalkanoate monomers, which are polyester family members. For example, these materials have low melting points, high crystallinity of thermoplastic elastomer molecules with thermoplastic polymers, excellent biocompatibility, as well as superior resistance to UV light. All of these properties of PHA have been governed by the monomer configuration. For food packaging with a short shelf life, PHA, as well as PHB, is the most likely candidate. Both P3HB-*co*-3HV polymers and P3HB homogeneous polymers are naturally occurring forms of polyhydroxybutyrate (PHB). Because they are polymeric granules, PHAs in bacteria serve as an energy storage medium, much like starch and fat do in plants as well as animals, respectively [34–36].

Compared to other non-polymers like polyethylene, the mechanical properties of PHB with 70% crystallinity were superior. The lamellar structure of PHB is the reason for its water permeability, barrier properties, as well as excellent aromatic behavior, which all contribute to its use in food packaging. As a result of these experimental studies, many researchers have attempted to use PHA/PHB in diverse applications despite its lower mechanical as well as barrier characteristics than PHA. When PHB was mixed with PLA as well as catechins have been added through the melt handling, the mechanical characteristics of the mixtures were evaluated. According to the findings, adding PHB–catechin combinations to plasticized PLA enhanced its mechanical characteristics, making it an excellent candidate for use in the containers of fatty foods [37].

1.2.5 Polyglycolide-Based Natural Materials

Ring-opening polymerization structure comprising a cyclic lactone as well as glycolide is used to make polyglycolide. The crystallinity index is around 50%, and as a result, it is insoluble in a variety of organic solvents. Its melting point is 222–226 °C, and its glass transition temperature is 37–42 °C. Polyglycolide is a strong material. Its biomedical implementations are limited, nevertheless, owing to its poor solubility as well as high acid-producing deterioration rate. The result is the development of caprolactone, trimethylene carbonate glycolide, or lactide, copolymers for healthcare devices [38, 39].

1.2.6 Polycarbonate-Based Natural Materials

It's a polymer with something like an elevated molecular weight that's easy to mold and bend. Two chemicals, glycolide and dioxanone, were combined to create copolymers. The copolymerization of propylene oxide as well as CO_2 results in polypropylene carbonate. For example, polycarbonate is easy to use and has a high degree of impact resistance. In the past, it has been combined with numerous different polymeric materials as a traditional method of use. The company sells a polyester carbonate called poly(oligo)(tetramethylene succinate)-*co*(tetramethylene carbonate). Carbonate addition to polyoligotetra-methylene succinate might well have induced crystal structure disorder, reducing its melting temperature, as well as attempting to make it more vulnerable to enzymatic as well as microbial attacks than polyolefins. This copolyester carbonate is more microbially degradable than either of its constituent elements [40, 41].

1.2.7 Soy-Based Bio-degradable Polymers

Polymer removal must have got to be the most pressing environmental issue for scientists. This has sparked a new wave of research that aims to use sustainable agricultural materials like starch or protein to create biopolymers. There are numerous advantages to using soybeans, including their low cost, wide range of applications, as well as appropriateness for the production of biodegradable plastics. PCL and polyethylene terephthalate (PBT) seem to be two other biopolymers commonly utilized within biodegradable materials for fiber reinforcement.

1.2.8 Polyurethanes

With so many applications in new methodologies like elastomers, adhesives, foams, fabrics, and coatings, that polyurethane has developed, it is no surprise that the material has become so widely used. There are numerous distinct physical and chemical properties to this particular polymer substance. The chemical composition of polyurethanes affects the biodegradation process. A suitable soft part can be used to halt or slow down the deterioration process. Polyether-based polyurethanes are completely biodegradable. If the polyol is polyester, polyurethanes are voluntarily biodegradable [42, 43].

1.2.9 Polyanhydrides

Polyanhydrides are being studied by a number of scientists, who discovered that the hydrolyzable locations in the recurring unit make them interesting biodegradable materials. There are few uses for aliphatic homo-polyanhydrides because of their elevated crystallinity index as well as rapid degradation. Polyanhydride degradation can be slowed by altering the polymer's hydrophobic and hydrophilic components [44]. The hydrophobicity of the polymer's diacid building blocks contributed to the polymer's slower degradation. They have been extensively studied in the field of biomaterials because of their hydrophobic aromatic comonomers. Polyanhydrides with diverse linkages, including ester, ether, along with urethane, are being made because of the huge assortment of diacid monomers available. For medical implementations, anhydride–amide copolymers were also established to improve the mechanical characteristics of polyanhydrides [45, 46].

1.3 Biodegradable Polymer Blends and Composites

1.3.1 Polylactic Acid and Polyethylene Blends

With compatibilizers, the impact opposition of the material has been even greater, while tensile properties including elongation, tensile strength, as well as Young's modulus had been relatively low in compatible as well as noncompatible substances, respectively, than in genuine PLA. PLA/PE blends have been made by Raghavan and Emekalam [47], and the degradation of the blends was studied in relation to the addition of starch. Filler materials were added to PLA/PE blends to increase Young's modulus as well as lower the stress as well as strain levels, according to a study of the mechanical characteristics of the blends. The mechanical but also thermal characteristics of PLA and polyethylene blends have been only tested by a small number of authors. The tensile properties of the blend lessened without a rise in thermal stability, as well as the blend's compatibility, was poor [48].

1.3.2 PLA and Acrylobutadiene Styrene (ABS) Blends

As a result of ABS's mechanical properties including tensile strength, impact strength, as well as tensile modulus, new blends with unique attributes have become more commonplace. Synthetic polymer blends with SANGMA and ETPB were also produced via the inclusion of ethyl triphene phosphinium bromide as a catalyst. SANGMA had been an essential responsive alignment for PLA/ABS blends with ETPB as a catalyst, as demonstrated by a rise in rubber particle distribution and improved resistance to impact loads as well as strain with a negligible deficit of tensile modulus as well as strength especially in comparison with pure PLA composites [49].

1.3.3 PCL and Polyethylene Blends

Many researchers used an internal mixer to organize the PCL/PE blend as well as appraise phase inversion during compounding. The mixture is inconsistent in the range of compatibilities that was tested. Maleic anhydride was used to prepare PCL as well as low-density polyethylene (LDPE) blends, which were then compared to PCL and block polyethylene glycol (PEG) blends. When compared to PCL/LDPE blends, the latter's mechanical characteristics have been superior, but the former was much more compatible [50].

1.3.4 PCL and Polyvinyl Chloride Blends

Thermal stabilizer dibasic lead phthalate (DLP) has been shown to affect PCL phase dispersion in PCL/PVC polymer blends. PCL, as well as PVC polymer blend solution rheology, was then happened. The H bond here between two chains of PCL and PVC resulted in complete compatibility in the blend. Few other experiments also affirmed their excellent suitability; a thermal property evaluation showed that perhaps the mixture had single Tg, which was in connection with the mechanical characteristics of the generated mixture, which also had shown that the combination break elongation rises as the PCL material is continued to increase [51].

1.3.5 TPS and Polypropylene Blends

Glycerol-containing TPS/PP polymer mixtures prepared and analyzed by a single screw extruder machine are few as well as far between. Shear-thinning behavior in conventional production machines indicated that the blends could be processed. In addition, the lubricating effect of glycerol on the material as well as the capillary rheometer dies decreased the mixture's viscosity as the glycerol content rose. Young's modulus significantly increased while strain reduced as TPS as well as glycerol content in the blend enhanced. This was revealed by the mixture's mechanical characteristics. Experiments on plasticizing biodiesel glycerol as well as glycerol used in the production of TPS as well as PP blends were few and far between; when the TPS content was enhanced, the tensile strength was reduced. The study found that the clay-modified TPS, as well as PP blends, had better mechanical characteristics than unmodified blends because they contained biodegradable components and had good mechanical attributes [52, 53].

1.3.6 TPS/PE Blends

Research on TPS as well as PE blends has been conducted by a small number of researchers. As a result of this method, two extruders were linked together, producing TPS and blends of TPS with glycerol as plasticizers. The physical, mechanical, as well as thermal characteristics of the formulated mixtures are being analyzed. When compared to other polyethylene blends, this one's thermal stability suffered because of the incompatibility of the two components. The strain of such mixture has been found to be comparable to that of PE; however, its modulus was found to be lower. Only a few studies looked at the mechanical as well as thermal characteristics of TPS/LDPE blends when ethylene, as well as vinyl acetate blend copolymers, were utilized as plasticizers; glycerol has also been used. Increased ethylene, as well as vinyl acetate substance in the mixture, resulted in improved mechanical properties as well as thermal stability [50, 54].

1.3.7 Poly(Butylene Succinate) Blends

PBS is an environment-friendly, biodegradable polyester with excellent thermal and chemical resistance. A polymer, they're a member of the group (alkenedicarboxylate). Plant-based fibers and fillers were added to PBS to improve its properties as well as lower its manufacturing costs. Rice straw fiber composites with amino acids as coupling agents have been studied by a small number of scientists. A binding agent comprising amino clusters resulted in composites with exceptional mechanical characteristics. The mechanical properties of PBS-reinforced coir fiber composites were examined by other researchers who used a 5% NaOH alkali treatment. An increment in fiber volume fraction resulted in a rise in tensile properties, while the strain at break was reduced. Research into the crystallization of PBS/cotton stalk bast fiber composites has revealed that cotton stalk bast fibers serve as both nucleating agents as well as defensive measures to chain segment transport all through crystallization [55, 56].

Flame retardant microencapsulated ammonium polyphosphate was studied and compared to magnesium hydroxide as well as aluminum hydroxide in terms of the thermal properties of composites. Only a few experimenters used melt mixing to produce PBS/bamboo fiber composites. In aspects of flame retardant properties, ammonium polyphosphate beat out magnesium hydroxide as well as aluminum hydroxide. Few studies have examined how sisal fiber content affects the rheological properties of PBS supplemented with sisal fiber composites. Shear-thinning was observed in the composites, with viscosity decreasing as the shear rate increased. Furthermore, a non-Newtonian composite index (n) reduces as the fiber content increases, suggesting that perhaps the composite viscosity is stable over a wide range of shear speeds [57].

1.4 Properties of Natural Materials for Food Packaging

Any food packaging material is expected to possess various characteristics such as barrier, thermal, mechanical, and biodegradable properties. Selection of natural materials based on all the above properties is the prevalent area of materials research. These materials offer improved gas barriers, antioxidants, antimicrobial, and light-blocking effects along with the inherent characteristics of the bio-based polymers.

1.4.1 Barrier Properties

Several of the requirements as well as a critical factor for biodegradable materials being used in food containers as well as other areas, which include the biomedical field, is that they have a higher moisture boundary property. However, TPS films have an increasing water vapor permeability (WVP). Even though starch is naturally hydrophilic, even before merging to glycerol, the bulging of the network could indeed retain a considerable amount of liquid. This bulging compromises the matrix's integrity of the structure, resulting in inadequate barrier properties [58]. The degradable films' excessive moisture permeability results in exterior trashiness. The sophisticated association between both the polymer matrix as well as protective characteristics are determined by a number of variables, including the matrix's structure, polarity, crystallinity, molecular weight, as well as the type of reinforcement. Moisture transmission among both food as well as the surrounding atmosphere results in spoilage; thus, food should be as resistant to WVP as feasible. The ASTM D570-81 standard way of determining a material's waterproofing requires curing prior to immersing weighed (Wi) samples in a specified volume of deionized water for 24 hours at ambient temperature. The specimens should then be eliminated as well as the moisture removed prior to weighing (Wf) [59, 60].

Relative humidity (RH) has noticeable impact on aquatic uptake of TPS layers. Aquatic acceptance and mechanical characteristics of resources at various RH are crucial for simulating the nature of initial packaging layers that are utilized to stock healthier food items (both veg and non-veg), etc. Aquatic acceptance depends on the group of plasticizers which are utilized while handling, which is added in a reasonable investigation on water uptake for TPS created from glycerol and bio-based isosorbide as plasticizers with corn starch as medium (TPSG and TPSI, respectively). At 75% of RH 52, TPSG was observed to hold aquatic acceptance of 25.7%, and TPSI which holds 22.8%. At 50% RH, TPSG possessed an aquatic acceptance of 10.4%, while TPSI possessed an 8.8% water uptake. When the RH was lowered to 25%, the moisture absorption values decreased further to 5.5% and 4.5%, respectively. The oxygen permeability of these materials varied insignificantly up to RH 75%, at which point it increased exponentially. Chitosan and chitin were found to have a significant impact on WVP values when added to TPS. The water vapor pressure of control TPS film was determined to be 1.3360 g/s m Pa. On the other hand, WVP values of 0.8760 and 0.5960 g/s m Pa indicate that the addition of chitin to TPS enhanced difficult characteristics more than the addition of chitosan, owing to the higher concentration of acetyl groups in the chitin structure. The majority of the published research on starch-based wrapping focuses on reducing WVP through the use of various fillers. Nonetheless, there is considerable potential for developing an intelligent packaging film that incorporates dynamic nanofillers [61–63].

1.4.2 Biodegradation Properties

A bio-based degradable polymer is defined as a polymer that degrades initially as a result of microorganism metabolic activity. Polymeric materials degrade primarily as a result of the bioactivity of microbes such as microorganisms, plankton, and germs. Amylases and glucosidases are enzymes that can attack and degrade starch. Nature provides a specific team of enzymes capable of attacking specific types of polymers. In general, three distinct classes of enzymes deteriorate a lignocellulosic polymer into glucose units: endo-cellulases, exo-cellulases, and cellobiohydrolases [64, 65]. These three classes of biocatalysts are collectively referred to as cellulases; even so, each class is capable of attacking a particular format of the polymer. No enzyme is capable of degrading the polymer effectively on its own. Bacteria produce a set of enzymatic necessary for polymer degradation by utilizing the organic matter in their surroundings. Biodegradable polymers fall into two categories: (i) proteolytic enzymes biodegradability polymeric materials (e.g. biopolymer, carbohydrate, glucans, etc.) and (ii) photo- or thermo-oxidizable polymers. Abiotic oxidative and biodegradative reactions occur at a higher rate in the presence of concentrated humidity than in the absence of saturated humidity. When bio-based polymers are released into the environment following their use, they are entirely degraded by microorganisms found in soil, saltwater, rivers, streams, and sewage. They have no negative impacts on the environment and contribute to the reduction of the greenhouse effect [66].

Few authors demonstrated complete dissolution of starch-based films containing glycerol, agar, and Sorbian mono-oleate in 30 days using an indoor soil composting method. The soil contained a diverse population of bacteria and fungi (*Staphylococcus* spp., *Salmonella* spp., *Streptococcus* spp., *Moraxella* sp., *Bacillus* sp., *Aspergillus* sp., and *Penicillin* sp.). Bacteria were counted at 30 3 106 to 43 3 106 CFU/g of samples, while fungi were counted at 18 3 103 to 23 3 103 CFU/g of soil. The number of bacteria associated with degradation was discovered to be 29.76 3 106, while the number of fungi was found to be 16.93 3 103. Microbes expanded in response to available growth and water resources. The glycerol content of the film affects microbial growth because it promotes swelling, which results in movement of chemical species of water and thus microbial growth. Microorganisms used starch as their sole carbon source, resulting in the destruction caused by the layers. Morphological observations indicate that the surface begins to erode after 10 days and completely after 20 days of biodegradation. The effect of variables on the biodegradation of starch films was investigated using a three-level Box–Behnken response design. The results indicated that the presence of water within the microstructure facilitates the entrance and improvement of microorganisms [67, 68].

1.4.3 Consequences of Storage Time

Molds, pultrusion, injection molding, and other processes are used to convert starches to thermoplastics. Liquid, polymeric, and other diluents are frequently added to aid in the decomposition of starch. Following processing, it has been demonstrated that TPS age and recrystallize into a variety of particle morphologies based on the making and storage circumstances. Aging is defined as the identified physical and/or chemical modifications in the characteristics of a PE as a feature of storage time when the polymer is kept at a constant temperature, under no stress, and unaffected by external parameters. Thus, aging of carbohydrates is a critical phenomenon that must be considered prior to application [69, 70]. A significant disadvantage of using starch is that TPS products deteriorate in reliability, appropriateness, and shelf life over time due to starch retrogradation. It is the process by which TPS's mechanical characteristics change as a result of recrystallization. The recrystallization process is triggered by macromolecules' proclivity for hydrogen bond formation during the evaporation of water and/or other cleaning agents. This process can be classified as amylose recrystallization or amylopectin irreversible crystallization. Retrogradation is also known as the lengthy recrystallization of amylopectin due to the slower rate of reversible recrystallization of amylose [71–73].

Aging has a complex influence on the development and consistency of TPS. Retrogradation occurs in TPS over time and is dependent on the type of plasticizer used (Figure 1.2). This degradation process affects the material's properties and applications. Crystallinity values vary according to storage duration, heat, humidity levels, and plasticizer composition. Additionally, methods for determining the retrogradation degree, such as X-ray diffraction analysis, are discussed. The effects of retrogradation on TPS properties such as tensile, elongation, and modulus are discussed. The rigidity of the product, as demonstrated by an increment in Young's modulus, was correlated with amylopectin reordering away from the starchy component, as demonstrated by an increase in B-type degree of structural order in a solid. These radical variations in the TPS suggest that as starch chains age, they become less mobile. According to the published literature, when various sets of

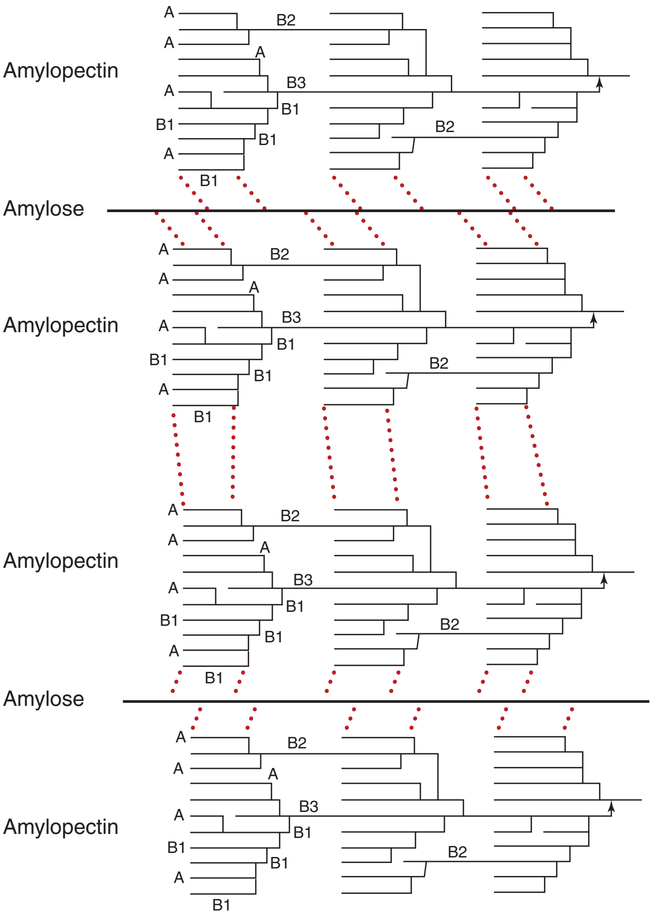

Figure 1.2 Retrogradation mechanism of starch solution. Source: Niranjana Prabhu and Prashantha [74]/with permission of John Wiley & Sons.

plasticizers and aging are used, starch-based materials exhibit big variations in material characteristics, and each plasticizer may be helpful for very particular purposes [74–78].

1.5 Environmental Impact of Food Packaging Materials

Implementation of inorganic food packaging films is a paramount factor that has to be considered to avoid the environmental hazards which it causes. Though the fabricated composites satisfy the regulatory limits for a food packaging material and have been proven to be an effective packaging material, their end environmental effect at the time of disposal is also to be taken into account [79]. Very few studies were carried out during the earlier stages on the detrimental environmental hazards caused by inorganic packaging materials over the environment. On contrary, during recent times, LCA has been used as a unified and systematic tool to determine the environmental effects of using an inorganic food packaging material through numerous experiments by considering various product lifecycle stages such as the raw materials used for the production of composite films, the process of film manufacturing, time of usage and method of disposal [80, 81].

From various studies, it could be stated that the environmental effects of inorganic polymer–metal packaging materials depend on factors such as degree of filler incorporation, method of manufacturing and synthesis of metal fillers, and the initial effect of food storage upon the environment. As migration capability and ionic or particulate mobility of the food packaging material plays a significant role in determining the environmental and toxic impacts, regulations regarding the migration evaluation of the food packaging material have to be implemented strictly to avoid adverse environmental effects [82, 83]. All these parameters are deemed to be important if the disposability of the food packaging material is below the par level. All these effects could readily be mitigated if natural materials are employed for food packaging and a few disadvantages like human toxicity, environmental burden, and disposal problems can be overcome by using those natural materials. It is also important to check the activity of the natural material employed for food packaging since it involves food consumption by human beings [84].

1.6 Conclusion

Evolution of natural, biodegradable materials in place of synthetic materials for food packaging applications has become the order of research today. Since plastic-based packaging materials accumulate to a greater extent through landfills and cause serious environmental pollution, manufacturing of biodegradable food packaging materials from natural and renewable materials is deemed to be necessary. All these materials have their importance since they were derived from natural and renewable sources. Research has to be oriented toward the commercial aspects of food packaging materials in such a way that they provide real-time information that helps for the commercialization of natural food packaging materials.

TPS, PLA, PHB, chitosan, and cellulose-based materials are used in various food packaging industries which in turn enhances the farmers economy who produces the raw materials. If the farmers themselves use all these natural-based materials in applications like compostable materials for agricultural and horticultural fields, bags, and mulching films, they will be beneficial to the farmers also. Usage of these materials poses various advantages like reduction in toxic gas release, decreased use of nonbiodegradable materials, and mitigation of environmental pollution. Cost is another important factor to be considered while designing an application based on natural materials for food packaging. Various researches have to be focused on reducing the cost of utilization of these natural material-based packaging materials since the cost is higher than the petroleum-based plastic materials during the current scenario. Such researches not only strive to reduce the cost of these natural materials but also enhance the applicability of natural materials for food packaging. As the development and application of natural materials need a multidisciplinary approach, efforts by various specialists from fields like microbiology, environmental science, chemistry, chemical engineering, polymer engineering, and other industrial giants who enforce the regulation on the use of natural materials have to meet at a concurrent point for the implementation and commercialization of natural biodegradable and eco-friendly food packaging materials.

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