Journal of Packaging Technology and Research
https://doi.org/10.1007/s41783-022-00140-9

REVIEW PAPER

Development and Characterization of Biodegradable Polymers for Fish Packaging Applications

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Received: 7 October 2021 / Accepted: 23 August 2022

Abstract

The most commonly used packaging materials are synthetic plastics. This petroleum-based synthetic plastics generate massive wastes to the environment and treating human life. During incineration, synthetic plastics release harmful gasses into the atmosphere. Research is under way to extract, develop, characterize, and apply biodegradable materials to minimize the impact of synthetic plastic. Biodegradable packaging is the best option to replace petroleum-based synthetic plastics. The objective of this review is to compile extraction, development, characterization and application of biodegradable films as packaging materials for fish products. Peer reviewed articles were downloaded from Springer Link, Science Direct, MDPI, and Taylor and Francis journals. Based on their origin biodegradable are classified into three namely biopolymers extracted from biomass (protein, carbohydrate and lipid), produced by classical chemical synthesis of bio-monomers and produced by microorganisms. Fabrication of biodegradable films involves the use of biopolymers, solvent (water or ethanol) and plasticizer (glycol, glycerin and sorbitol). Solvent or solution casting is the most widely used method to prepare biodegradable films. Instruments used to characterize biodegradable films are Fourier transform infrared spectroscopy, differential scanning calorimeter, X-ray diffraction, UV–Vis Spectrophotometry, Nuclear Magnetic Resonances and Thermal analysis. The advanced technology for fish products packaging are modified atmosphere packaging, active packaging, intelligent packaging and smart packaging. It is concluded that biodegradable film have great potential for fish packaging and do not harm the environment as it breakdown into organic matter.

Keywords Biodegradable polymers · Fish products · Packaging · Polysaccharide · Protein

Introduction

The major materials used for food packagings are low and high polyethylene, terephthalate, polystyrene, and polypropylene [1]. Annually, around 300 million tons of petroleum-based plastics are manufactured around the globe. However, they pose a serious global environmental problem by generating large volumes of non-biodegradable waste [2]. The current plastic waste disposable systems are land filling, incineration, composting and recycling [3]. Incineration of synthetic plastics releases harmful gasses such as NO, N_2O, NO_x, CO, SO_2, HF and HCl into the atmosphere [4]. The chemicals used during petroleum-based synthetic packaging materials are endocrine disrupters and release massive green house gasses upon burning. Some additives which are added during plastic fabrication can cause growth retardation, neurological damage, hormonal changes and cancer in children. This plastic materials enters into aquatic ecosystem it break down to microplastics in the marine environment, which aid as a vector for the assimilation of persistent organic pollutants in the oceans and are transported into the food chain via marine and wild life. Researches involving the production, characterization and application of biodegradable films have tremendously increased to minimize the ecological impact caused by synthetic packaging materials. Recently, great attention has being given to bio-based polymeric films for food packaging application which exhibits high potential to replace non-biodegradable materials [5, 6]. Biodegradable packaging

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Published online: 05 September 2022

materials are promising due to safe, sustainable, eco-friendly and extend the shelf life of food products [7]. As a result; the scientific community is paying more attention to the creation of films made from biodegradable polymers produced from renewable resources, such as proteins, polysaccharides, microbial polyesters, and polyurethanes. Therefore, realizing the top urgency of alternative packaging materials using natural based sources, this review is written to summarize extraction, development, characterization, and application of biodegradable packaging materials for fish products.

Methodology

Full length research and review papers were collected from Elsevier, Springer Link, Science direct, MDPI, Wiley online library, Taylor and Francis. The term 'extraction of biodegradable polymers', 'development of biopolymers film', 'characterization of biopolymers film' and 'application of biodegradable film', and 'advanced fish packaging technology' were used to search articles. A total of 270 articles were obtained, among which 100 articles were chosen as relevant on the basis of scope of study. The articles used for this review were classified as journal articles (96), and book chapters (4). Information was reviewed on types of biomass used to prepare biodegradable films, solvents and plasticizers used to make packaging materials.

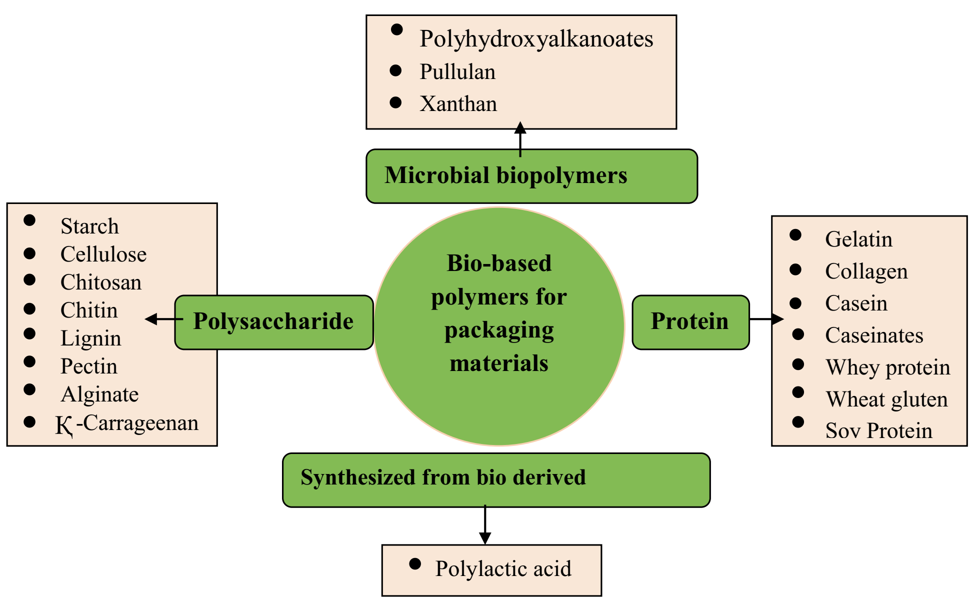
Extraction of Biodegradable Packaging Materials

Biodegradable polymers are naturally occurring polymers that are capable of undergoing decomposition by micro-organisms (bacteria, fungi and algae) into carbon dioxide, water, methane and biomass compounds under suitable conditions of temperature, moisture, and oxygen through the enzymatic action of microorganisms [8]. In order to develop natural biodegradable packaging materials, first the polymers must be extracted from natural biomass. Natural polymers include the polysaccharides, proteins, microbial biopolymers, and synthesized from bio-based derived monomers. The major types of bio-based polymers for packaging materials are illustrated (Fig. 1).

Extraction of Polysaccharides

Polysaccharides are extracted from plants or algae. Polysaccharides like starch, cellulose, chitosan, chitin, lignin, alginate, carrageenan, pectin, and glucan have significant aspects for fish packaging materials. The major sources of polysaccharides and their optimum extraction methods from bio-based matrices are described (Table 1). They have the properties of fillers in biopolymer matrices, biocompatibility, biodegradability, non-toxic, antimicrobial, high thermal and chemical stability. This section discusses the best methods to extract polysaccharides and characterize as a biopolymers to develop materials for fish packaging applications.

Fig. 1 The major types of natural biodegradable polymers



Extraction of Cellulose

Cellulose ($C_6H_{10}O_5$) n is the most abundant natural polymer on Earth, forming a key constituent of the cell walls of terrestrial plants. It is a linear homo polysaccharide of repeating units of β -1, 4 linked D-glucose. Extraction of cellulose from fruits and vegetables involves removal of phenolic compounds, sugars, and water-soluble polysaccharides, then treating with HCl to remove pectin polysaccharides. Next, the hemicelluloses are removed by treating the residues with alkali followed by bleaching with sodium hypochlorite solution, eventually cellulose is recovered as precipitate [9]. Cellulose the most abundant polymers and cellulose derivatives (e.g., methylcellulose, hydroxypropylcellulose) have good film making properties. Cellophane was the first cellulose film developed in 1908. Cellulose based films are very efficient oxygen barriers and its water vapor barrier properties can be improved by the addition of hydrophobic polymers [10].

Extraction of Starch

Starch is a reserve material of photosynthetic tissues from plants storage organs and seeds. The two main constituent of starch is amylose and amylopectin. Amylose is a linear molecule of 1–4 linked a D-glucopryanosyl unit where as Amylopectin is branched molecule made of a-D-glucopryanosyl linked together mainly 1–4 linked but 1–6 linkages at the branch points. Starch can be obtained from wheat, rice, seeds, corn, potato, sweet potato and cassava. It is plasticized through destructuration in presence of specific amounts of water or plasticizers (glycerol, sorbitol) and heat and then it is extruded [11].

Extraction of Chitosan or Chitin

Chitosan or chitin is the second abundant polysaccharide resource after cellulose found in nature. It differs from cellulose only by -OH groups. Chitosan is the deacetylated derivative of chitin [12]. Chitin (C_8 $H_{13}O_5N$)n is a copolymer comprised of D-Glucosamine along with N-acetyl-D-Glucosamine and present in exoskeleton of crustaceans and in the cell walls of yeasts and fungi [16]. The two important steps in chemical methods are first, deproteinization by alkali treatment followed by demineralization by acidic treatments at high temperature finally bleaching with reagents to attain colorless products. The most preferred chemicals for this process are NaOH and HCl [13].

Recovery of Lignin

Lignin ($C_{31}H_{34}O_{11}$)n is a complex polymer of aromatic alcohols, and forms another important component of cell walls in plants, providing strength and restricting the entry of water. Lignin is a derivative of lignocelluloses. As compared to cellulose and hemicelluloses, it is more resistant to biological stresses. Lignin is extracted with 40% NaOH (aqueous solution), 20.4% distilled water and ethanol solution [8].

Pectin Extraction

Pectin is acidic, hygroscopic water-soluble polysaccharide occurring in different natural by-products. The major sources of pectin are lime peel or lemon. Pectin has linear chain structure containing the region made up of α-1,4-linked D-galactopyranosyluronic acid units. Pectin polysaccharide which is derived from the plant cell wall and it is obtained commercially by aqueous extraction of pectin material like citrus fruits and apples [13].

Alginate Extraction

Alginate is a hydrophilic colloidal carbohydrate, is the most polysaccharide extracted from brown seaweed. It is produced using dilute alkaline solution method. Alginate is widely used as stabilizing, suspending, film forming, gel-producing and thickening. Alginate is made of $1 \rightarrow 4$ linked β-D mannuronic acid and α-L-Guluronic acid residues. It is derived from brown seaweed like *Laminaria hyperborean*, *Laminaria digitata*, *Macrocystis pyrifera* and *Ascophllum nodosum* [13, 14].

Carrageenan Extraction

Carrageenan is polysaccharides extracted from red seaweed Rhodophycae family. It is anionic linear sulfated polysaccharides composed of d-galactopyranose residues [14]. Carrageenan is a partially sulfonated galactans extracted from red sea weed using dilute alkaline solution. After extraction, the dilute extracts are filtered, concentrated and precipitated with isopropanol to produce coagulum. Then the coagulum is pressed to remove solvent and washed. Finally, it is dried and milled.

Extraction of Glucans

Glucans are polysaccharides composed of 1, 2 bonded glucose monomers. It is carbohydrate (sugars) that are occurring in the cell wall of bacteria, yeast and fungus, algae, lichens, and cereals like oat, and barley [15]. β-glucans are

Table 1 Sources of different polysaccharides and their extraction methods

Types of polysaccharides	Sources of polymers	Extraction methods	References
Starch	Pruned olive tree	16% NaOH, 170 °C for 60 min for pulping then bleaching followed by TEMPO-mediated oxidation and high-pressure homogenization treatment	[16]
Starch	Rice	Rice (1 kg) was soaked in sodium hydroxide (0.3%, 10 L) at 25 °C for 24 h. The slurry was suspended in deionized water, the pH was adjusted to 7.0 with HCl (0.5 M) and passed through nylon screen (53 mm).	[17]
Starch	Rice	Rice flour was soaked in 0.1% NaOH (1:2 m/v) for 18 h, filtered, centrifuged, for 10 min	[18]
Starch	Potato	Potato slurries was frozen at – 4 °C, freeze dried, and sieved using 150 µm	[19]
Cellulose	Kudzu	Kudzu fibers were depolymerized, bleached, acid hydrolyzed, and mechanically dispersed	[20]
Nanocellulose	Water hyacinth fiber	Digester sonication	[21]
Chitosan/chitin	Yellow lobster waste	Nanocrytals of alpha chitin were extracted by microwave assisted extraction technique for ten minutes using1M HCl and 124.75 W	[22]
Chitin/chitosan	Seafood wastes	Demineralization, deproteination, and decoloration	[23]
Chitin/chitosan	Shells from different sources	The shells are washed, dried, ground into powder then deproteination using NaOH (0.125 to 5 M), demineralization using (hydrochloric acid, sulphuric acid, nitric acid, acetic acid), and decoloration	[24]
Chitin/Chitosan	Nile tilapia fish scales	Scale was washed at 28 °C, dried at 45 °C for 18 h. Then demineralization (2% HCl), deproteinization (4% NaOH), depigmentation and deodorizing (1% NaClO), and deacetylation (4% NaOH at 45 °C for 18 h)	[25]
Lignin	Wheat straw Flax fiber Pine straw Alfalfa fiber	Isolation of lignin involves treating with peroxyformic acid/perofxyacetic acid, formaic acid/acetic acid, and bleaching	[26]
Pectin	Wild plum pomace	Pectin from wild plum pomace was extracted using HCl (13.26%) at 90 °C for 90 min	[27]
Pectin	Berries agro-industrial wastes	Berry was disinfected with sodium hypochlorite (12 °C) for 15 min and dried then pectin based films was developed to monitor shelf-life of Salmo salar	[28]
Alginate	Seaweed	Dried seaweed (20 g), suspended in 500 mL HCl (0.2 M), shaken for 12 h in ion exchange, and centrifuged. Finally alginic acid was suspended in $NaHCO_3$ 900 mL of 0.1 M	[29]
Ꝃ-Carrageenan	Red algae	A sample (10 g) of Gigrtina skottsbergii (red algae) was soaked in 800 ml distilled water for 1 h. The solution was heated at 74 °C for 4 h.	[30]
Carrageenan	Jaboticaba peels powder	Jaboticaba peels powder (0.5 g) were homogenized with 25 mL distilled water and extracted using closed multi-mode mini wave microwave	[31]
Glucans	Plants	The two best method to extracted glucans from plants are alkali and hot water extraction methods	[32]
β-glucans	Hull-less barley bran	Microwave-assisted ultrasonic extraction	[33]

used as texturing agents in food industry. The other function of β-glucan is microencapsulation of sensitive natural pigments. The prominent polysaccharides for fish packaging and their extraction methods are shown (Table 1).

Extraction of Proteins

Proteins from animals or plants sources have significant aspect in the development of packaging materials. Proteins have unique characteristics to be used in packaging applications. The peculiar properties of proteins are environmentally friendly, surface hydrophobocity, low production cost, water uptake capacity, barrier, thermal and mechanical properties, solubility and etc. The extraction method of proteins from both plants and animals is described (Table 2).

Extraction of Gelatin

Gelatin is an animal protein, obtained by hydrolysis of collagen, which shows the capacity of forming thermoreversible physical gels, is present in bones, skins and connective tissues of land and marine animals [34]. Enzymatic and acid hydrolyzes are most widely used method for extraction of

Table 2 Types of protein polymers and their extraction method

Types of proteins	Sources of polymers	Extraction methods	References
Gelatin	Fish skin collagen	Microwave rapid freezing–thawing coupling	[38]
Gelatin	Fish skin	Fish skin was pretreated with NaOH (0.05 M) and HCl (0.62 M) at 45 °C and pressure bar for 3 h	[39]
Gelatin	Fish skin, pig bone, pig skin, bovine bone and hide	Advanced gelatin extraction techniques (enzymatic, ultrasound assisted, and high-pressure treatment)	[40]
Casein/caseinates	Skimmed milk	Electrodialysis with bipolar membrane coupled to an ultrafiltration module	[41]
Whey proteins	Cheese whey	Magnetic-solid phase extraction through coating with Cu^{2+} and/or citric acid, finally 53% of proteins eluted with sodium chloride from Cu^{2+}/citric acid/Fe_3O_4 and only 24% from citric acid- Fe_3O_4	[42]
Whey protein	Bovine milk	Ion exchange and adsorption techniques	[43]
Whey protein isolate	Cheese whey	Capturing lactoferrin using sulfanilic acid modified chitosan mini spheres then recovering whey protein using glycidyl trimethylammonium modified chitosan mini-spheres	[44]
Soy protein isolate	Soybean protein	Extraction of soybean protein using by-products from NaCl electrolysis	[45]
Soy protein isolate	Soybean residue (okara)	The protein was extracted by vacuum filtration and precipitated at pH 4.6 with 2 N HCl and refrigerated at 4 °C	[46]
Corn zein	Corn silk	Solubility method using NaOH and Urea Filtration method using NaOH and H_2SO_4	[47]
Corn zein	Corn	Wet-milling of corn through steeping corn in water containing SO_2 or sodium metabisulfite, lactic acid to soften corn kernel at controlled pH and temperature	[48]

collagen from waste parts of skin and fins of fishes, egg and chicken; whereas bone collegen could be extracted by decalcifying with 0.5 M ethylenediaminetetraacetic (EDTA) acid. Sodium chloride, acetic acid and pepsin are used to hydrolyze non-collagenous protein at cross-linking sites [35]. Gelatin is an appropriate polymer for the fabrication of biodegradable packaging due to its ability to form films, carries functional agent, low melting and gelling points, good capacity of oxygen barrier, abundancy and biodegradability [35]. It protects food from light, oxygen, dehydration/moisture loss when used as external cover. Gelatin is first polymeric materials proposed as vehicle for transporting of bioactive compounds. Skin and bones of pigs and cattle are the most abundant sources of gelatin. As an alternative to mammalian gelatin, fish gelatin has gained tremendous attention due to vegetarianism concerns as well as the potential of halal and kosher markets. Due to the presence of -NH_2, -COOH and -OH reactive groups, gelatin compatibly mixed with other polymers for development of bio-composite packaging materials. The induction of intermolecular and intramolecular chemical bonding through addition of chemical or enzymatic cross linkers can improve poor mechanical properties of gelatin based biopolymer films [36].

Extraction of Casein and Caseinates

Casein is the principal protein fraction in cow milk, which accounts for 80% of its total protein content. Casein appears as micelles with 50–300 nm micelles and is relatively heat stable. The Na-caseinate (NaCAS) can be dissolved in water and forms easy to films [37].

Recovery of Whey Proteins

Whey protein is the soluble part of the milk and constitutes about 20% of the total amount of the proteins. Due to the cross linking and intermolecular hydrogen bonds, whey proteins provide excellent oxygen barrier properties and at low RH, the oxygen permeability is in the range of Ethylene vinyl alcohol polymers and is therefore useful to improve the oxygen barrier properties of food packaging [37].

Extraction of Wheat Gluten

Wheat gluten is known to form elastic networks for a wide range of bakery and other food products since centuries. The manufacturing of gluten films takes place by solvent coating, aided by mechanical mixing, heating as well as adjustments to acid and alkaline conditions [37]. Biopolymer films fabricated from wheat gluten are useful in food packaging given they are strong, heat sealable, flexible and relatively transparent. Like any other protein films and, wheat gluten films are excellent barriers to oxygen and carbon dioxide at low relative humidity and selectively permeable to gasses.

Extraction of Soy Protein Isolate

The soy protein isolates are usually a by-product from the soybean oil industry. Soy bean films show good-film-forming properties. SPI is useful for producing edible films and coatings through aqueous casting method. SPI film can be possibly produced through dry processes extrusion in the presence of the plasticizer like glycerol [37]. Soy protein isolate (SPI) films are good oxygen and lipid barriers but they have poor water barrier properties due to their hydrophilic nature. Glycerol is the most widely used plasticizer for SPI films because of its small size and hydrophilic nature [10].

Extraction of Corn Zein

Corn zein is an alcohol soluble (prolamin) fraction of maize proteins, which represent 50% of maize endosperm proteins. The manufacturing of zein protein isolates (ZPI) is more sophisticated than proteins from other plants and therefore zein isolates are quite expensive. Corn zein films provide lower water vapor permeability, better water resistance and good moisture barrier properties due to the presence of hydrophobic amino acids, high non-polar amino acids, low ratio of basic and acidic amino acids. However, Zein films are brittle, hence, require modification using cross-linking to improve its flexibility [10]. The major source of protein and their extraction method are discussed (Table 2).

Recovery of Biopolymers From Bio-Derived Monomers

Polylactic Acid

Polylactic acid (PLA) is one of the most commercially available and exploited bioplastics which are synthesized from bio-based monomers. It is obtained by conversion of corn, or other carbohydrate sources, into dextrose, followed by fermentation into lactic acid. Polylactic acid is produced by chemical conversion of carbohydrate sources into dextrose which is then fermented into lactic acid and followed by polycondensation. Polylactic acid can be obtained through polycondensation of lactic acid monomers or through ring-opening polymerization (breaking the cyclic lactide chain) of lactide with various metal catalysts in solution or as suspension [11, 12].

Recovery of Biopolymers Microbial Polymers

Polyhydroxyalkanoates

Polyhdroxyalkanoates (PHA) are thermoplastic polyesters produced by simple fermentation process. Some bacteria accumulate polyhdroxyalkanoates (PHAs) as a source of energy or carbon reserve. The culture media required by microorganisms are acetic and glucose. It is the parts of living organisms structure, hydrophobic and insoluble in water [12].

Pullulan

Pullulan-based biopolymers are reported as biodegradable, resistant to oil and grease, heat sealable, and therefore highly used as inner packing materials for instant coffee and noodles. *Pullularia pullulans* or *Aureobasidium pullulns* are fungus which produces pullulans from substrates containing sugars. Films based on pullulans are biodegradable, edible, non toxic, odorless, good barrier to oxygen, homogeneous, transparent, printable, heat sealable, oil resistant, flexible and tasteless in nature [11].

Xanthan Gum

Xanthan gum, made up of glucose, mannose, acetate pyruvate, and glucuronic acid, is used for edible coatings of meat and fruits as it has interesting characteristics that can prevent moisture migration during frying and extend the shelf life of food. Xanthan is non toxic, highly viscous and water soluble in nature. It is produced by aerobic fermentation of sucrose or glucose as major carbon source by *Xanthomonas campestris* [11]. Xanthan gum can be produced using X. campestris strains in different composition, viscosity and yields using cheese whey and citrus waste [8].

Development of Biodegradable Packaging Films

Polymer extraction is followed by melting the biopolymer mix then solution casting, melt mix, electro spinning, thermo pressing and casting, injection, screw extrusion, blow film extrusion, extrusion, injection molding, molding, rotational molding, compression molding, blow molding and foaming methods can be employed based upon the films to be made [49]. During fabrication of films, biopolymers solvents and plasticizers are required. Plasticizers such as glycerol, sorbitol and glycol are essential to make the films more flexible, softer and avoid pores and cracks in the polymeric matrix [50]. Plasticizers are required to break polymer to polymer interactions thus lowering the glass transition temperature (T_g) and reducing film rigidity and brittleness. Fabrication of biopolymers is a multi-step process that requires understating the nature of biopolymers and proper skill during processing. There are different methods for fabrication of biodegradable films from biodegradable polymer. Solvent casting (Solution casting) involves mixing of biodegradable polymer with organic solvent, drying of the films for 6 h to 2 days in a fume hood until the organic solvent is fully evaporated. Finally, the remaining solid film can be peeled off the container manually [51].

Development of Packaging Films Using Solvent Casting

Solvent casting or solution casting is the most frequently used methods for the preparation of biopolymer-based biodegradable film [52]. It involves mixing of biodegradable polymer with organic solvent. Then, the mixture is left for 6 h to 2 days in a fume hood until the organic solvent is fully evaporated. Finally, the remaining solid film can be peeled off the container manually [53]. As compared to polysaccharide solution, foam is formed in protein solution; hence, removal of bubbles (degassing) using vacuum pump before casting is an important step [52].

The development of biodegradable packaging materials must contain a film forming agent (polymer) which is capable of forming a suitable continuous, adhesive and cohesive matrix. The solvents like water or ethanol and plasticizer (sorbitol, glycerol and water) is also the required components. Even though these polymers are hydrophilic and crystalline in nature, they have excellent gas barrier properties which make them acceptable for their utilization in food packaging. The limitation of using biopolymers as food packaging materials as compared to non-degradable plastic materials include poor mechanical properties (e.g. low tensile strength), rigidity, brittle character, high hygroscopic capacity and barrier properties (e.g. high water permeability). To overcome these drawbacks, biopolymers can is blended with other polymers and plasticizers which resulted in good film-forming ability, plasticizers, and additives have met the demanding mechanical properties of packaging [54]. Plasticizers such as glycol, glycerin and sorbitol are added as additives in the formulation to modify mechanical properties and provide significant changes in flexibility, avoid pores, and extensibility of the polymeric matrix [54]. Plasticizer like sorbate is considered as GRAS hence, the addition of sorbate to biopolymers minimizes microbial contamination. Development of packaging films using solution casting method is depicted (Fig. 2).

Characterization of Biodegradable Packaging Materials

Biopolymer film is characterized for the purpose to develop parameters for processing and to determine final performance characteristics. The physical and mechanical properties of film to be characterized are film thickness, moisture content, water solubility, smelling capacity, color, light transmission, young's modulus, tensile Strength, percentage elongation at break, puncture strength, water vapor transmission rate, oxygen permeability, gas barrier properties, antibacterial activity and antioxidant capacity [52, 55]. Once the films are fabricated it will be characterized against the morphological, thermal, structural, mechanical and barrier properties. Some of the analytical techniques used for the characterization of biopolymers are Scanning electron microscopy (SEM), Fourier transform infrared spectroscopy (FTIR), Differential Scanning Calorimeter (DSC) analysis, X-Ray Diffraction (XRD), thermogravimetry analysis UV–vis Spectrophotometery and Nuclear Magnetic Resonance (NMR).

Fourier Transform Infrared Spectroscopy (FTIR)

Fourier transform infrared (FTIR) spectroscopy explores the mechanical and physical properties of biopolymers. It clearly depicts the intermolecular interaction of functional groups present in biopolymers (Proteins and carbohydrate). The spectrum confirms the presence of alkanes, alkynes, alkyl halides, alcohols, phenols, carboxylic acids, aldehydes, aromatics, nitro compounds and amine in polymers [56]. Fourier transform infrared spectroscopy technique is used to characterize biopolymers films. The most characterized attributes are interaction between biopolymers in blending films, cross-linking processes in biopolymer films and interaction between the bioactive substances with the biopolymer matrix [57].

X-ray Diffraction Analysis

Crystallinity of biopolymers can be successfully studied by X-ray diffraction (XRD) techniques [34]. X-ray Diffraction is most commonly used technique to identify the crystalline materials/phases inserted in biopolymer film. These materials have specific X-ray diffraction patterns, hence, it used to identify the phases and components present in biopolymer film [56].

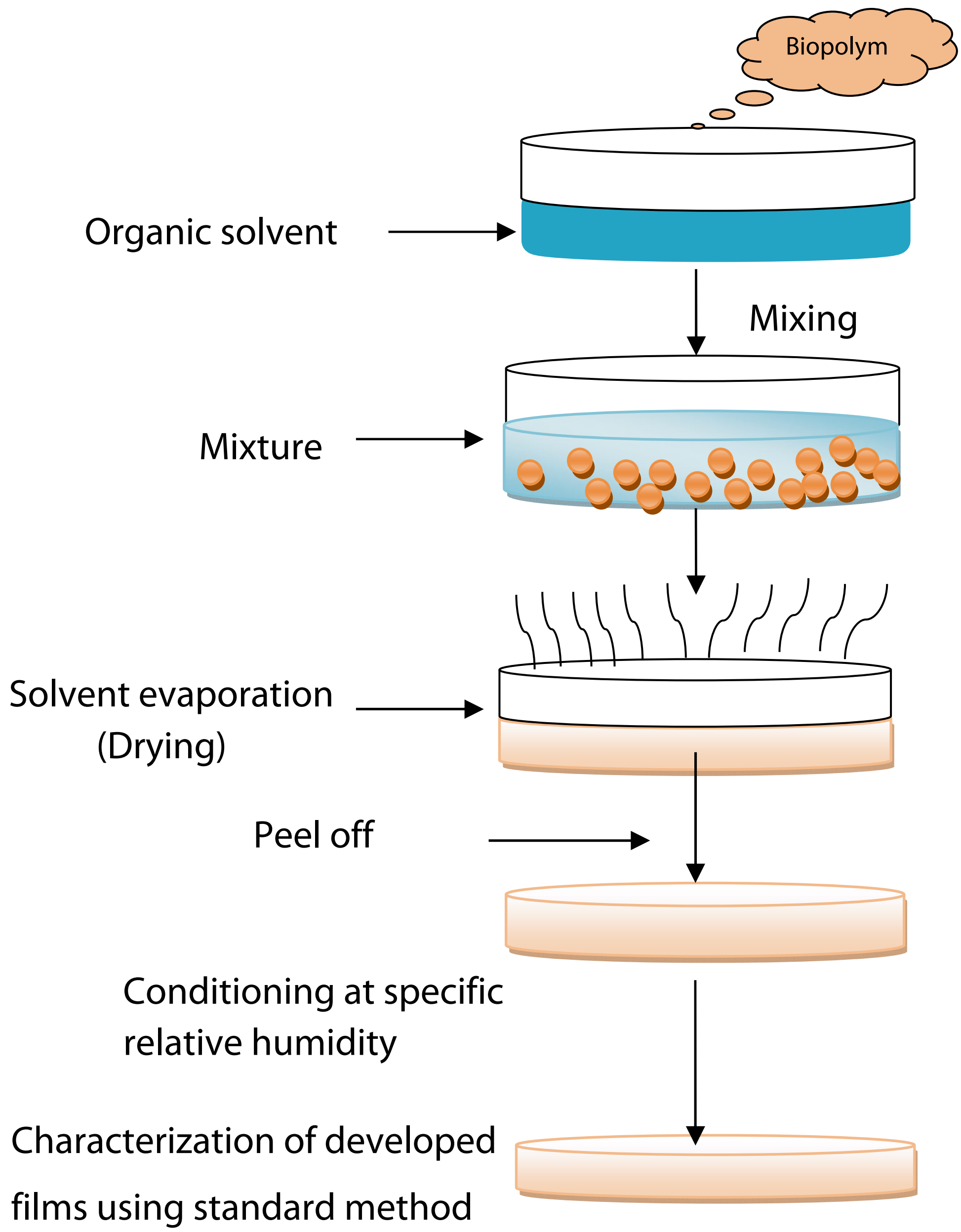
Scanning Electron Microscopy (SEM)

Scanning electron microscopy is sophisticated technique, produce high image quality and magnification, enables nanometer detail visualization and elemental mapping. Scanning Electron Microscopy works as follows. An atom sample interacts with electron beam. These interactions produce different signals which are captured by detectors and generate the sample image [56].

VIS–NIR Spectroscopy

Visible and near-infrared (VIS–NIR) spectroscopy is a well-established analytical instrument to measure the biological constituents of polymers [34]. VIS–NIR spectroscopy is an analytical tool for qualitative analysis of biological sample

Fig. 2 Development of biodegradable films using Solvent (solution) casting techniques



characteristics. It can be used to explore the composition of biodegradable materials and correlation of their mechanical properties.

NMR Spectroscopy

NMR spectroscopy is a technique which determines the molecular dynamics of water adsorbed on surface and functional groups of biopolymer [34]. The vibrational modes of polymers influence the NIR spectrums which are caused by interaction of EMR absorbed at specific wavelengths. Such technique is used for identification of proteins and carbohydrates in biopolymers. NMR spectroscopy is a technique widely used to identify the structure of the compound as well as purity of materials. It provides information about the number of magnetically distinct atoms [56].

Thermal Analysis

Thermal analysis (TA) is an analytical technique which characterizes chemical and thermal stability, rheology and molecular relations times, phase transition temperature and kinetics. The two common thermal analysis applied to biopolymers are Thermogravimetry and Differential Scanning Calorimetry [34, 56].

Thermogravimetry (TGA)

Thermogravimetry is a type of thermal analysis that monitors the sample mass against time or temperature on a controlled environmental furnace. It determines thermal stability, chemical composition, water content and oxidative stability. It analyzes the increase or decrease in thermal degradation peaks and verify the incorporation of nano-particles and active compounds in biopolymer film [56].

Differential Scanning Calorimetry (DSC)

Differential Scanning Calorimetry is an important technique for analysis of polymer. It determines thermal events, oxidative stability, thermal degradation and water loss. It also determines glass transition temperature (T_g), which indicates the reversible transition in amorphous materials from a brittle state to viscous state. Transition temperature indicates the miscibility of biopolymer blends [56].

The physical, morphological, and thermal properties of biodegradable packaging materials improved during addition of some other biopolymers into other have illustrated (Table 3). The most frequently characterized properties of packaging materials are thermal, barrier, water vapor permeability, elongation at break, young modulus, interaction of functional groups of biopolymers, light transmission, films' surface morphology, mechanical properties, tensile strength and etc. Characterizations of biodegradable packaging films have significant aspect for exploring different combination of biopolymers. It enables to understand what properties of the films are significantly affected due to the additions biopolymers to the other. Determination of the packaging properties enable to decide the optimum ratio of biopolymers to be blended to get the best packaging films in all properties.

Application of Natural Based Polymers of Fish Packaging

Biopolymers have several application area such as food packaging, drug delivery, entertainment industry, medicine, electrical, electronics and computer industry, casings agricultural drugs, orthopedics, protein delivery, bioimaging and diagnosis, tissue engineering, Cosmetic preparation, waste water treatment, paper and textile, biomedical application and skin care [8]. Application of biodegradable materials for food packaging has gained interest as alternatives choice to conventional food packaging polymers due to their availability, broad and abundant source range, degradability, eco-friendly, compatibility with foodstuffs and food application [87]. Biodegradable films have been developed using gelatin, either alone or in combination with chitosan for application to fishery products. Packaging materials fabricated from biopolymers incorporated with bioactive substances are useful for monitoring fish freshness. Accordingly, carboxymethyl-cellulose/starch incorporated with anthocyanins was used to monitor fish freshness [88]. Corn biopolymer (Zein nanofibers) incorporated with alizarin as indicator dye was fabricated to monitor rainbow trout filets freshness in real time through color changes. These biopolymers containing zein and alizarin has excellent application for use as intelligent trout filet monitoring as demonstrate visual color changes from yellow to magenta during storage at 4 °C [89].

Methylcellulose/Chitosan nanofiber and barberry anthocyanin film has demonstrated real-time freshness monitoring of meat and seafood products based on change of color from reddish-pink to pale peach and eventually yellow when the packaged meat/seafood was subjected to different pH [90]. Corn starch and polyvinyl alcohol biopolymers and anthocyanin extracted from purple sweet potato and red cabbage were used to fabricate pH sensitive film. The film demonstrated visual color changes due to TVB-N variation which can be applied to monitor freshness of shrimp [91]. Freshness of rainbow trout filet refrigerated at 4 °C was monitored using pH sensitive biopolymers fabricated from Starch-Cellulose and Alizarin dye. The film has shown color changes from orange to reddish brown due to TVB-N amounts of fish filet, hence, this film can be used as indicator of spoilage in refrigerated rainbow trout filet [92].

Advanced Technology for Fish Products Packaging

Modified Atmosphere Packaging

Fish is the most perishable food items. It is susceptible to spoilage due to autolysis, lipid oxidation and microbial growth. Modified Atmosphere packaging (MAP) was developed in 1930s that was aimed to minimize fish spoilage. MAP is a packaging condition in which air is replaced with different gas mixtures to control microbial activity, which enhance the shelf life of fish products [93, 94].Gasses which are widely used in MAP are Oxygen, CO_2, and N_2. Depending upon whether the packed fish is lean or oily, the composition of the gas mixture used for MAP of fresh fish varies. For fatty or oily fish higher values of Carbon dioxide, reduced level of oxygen and 40%–60% nitrogen is an ideal gas composition to slow oxidative rancidity in fatty fish. For lean fish, 30% oxygen, 40% carbon dioxide and 30% of nitrogen is recommended [93, 94].

Active Packaging

Active packaging is defined as a type of packaging that changes the condition of the packaging and maintains these

Table 3 Characterization of biodegradable packaging materials

Types of natural based polymers	Method of preparation	Plasticizers	Properties of the packaging materials improved	References
Cellulose Curcumin Chitosan	Solution casting method	Glycerin	WVPR and OP decreased by 80.47% and 76.96% EAB and antioxidant activity decreased from 34.5 to 23.8% and 0.17 to 0.15 respectively	[58]
Carboxylmethyl cellulose Fish gelatin	Solution casting method	1 to 3 ratio of sorbitol-glycerol	Addition of 50% CMC to fish gelatin increased tensile strength and young's modulus and decreased EAB	[59]
Carboxymethyl cellulose Polyvinyl alcohol	Solution casting method	0.1% v/v glycerol	Films containing rose petal extract improved tensile strength, however no change in elongation at break as compared ot neat films	[60]
Carboxymethyl cellulose Gelatin Agar	Solution casting method	2% glycerol	Films made from 2% glycerol and gelatin/CMC/agar had lowest water vapor permeability and higher biodegradability	[61]
Starch, polyvinyl alcohol Betacyanins	Solution casting method	1.2% glycerol and 20 mL of betacyanins	Films made from red pitayaa and prickly pear fruit extracts have uniform cross-sections, 9.37 and 9.26×10^{-11} g m^{-1} s^{-1} Pa^{-1} water vapor barrier and 8.26 and 7.87 MPa mechanical strength	[62]
Bacterial cellulose Carboxymethylcellulose olive and ginger oil	Solution casting method	30% glycerol	Water solubility has increased for both olive and ginger oil up to 37.5% and 41.08% respectively Good antibacterial against *S.aureus, P.aeuginosa and E.coli, C.albicans and Trichosporon* sp.	[63]
Polylactic acid Tapioca starch	Injection molding method	–	Ratio of Polylactic acid to tapioca starch composite at 70/30 wt% had the optimum stage of tensile strength	[64]
Polycaprolactone Polylactic acid Green tea extract	Solution casting method	–	The films containing 30% polylactic acid has reduced hydrophilicity, water solubility, water vapor permeability, and oxygen transition rate as 14.96%, 38.89%,8.75%, and 35.55% respectively	[65]
Chitosan Sardinella protein isolate	Solution casting method	–	The structural and thermal properties of improved as revealed by FTIR, SEM and TGA Composite film has lower mechanical properties than chitosan film	[66]
Nanocelluloses from eucalyptus, pine and cocoa shell	Solution casting method	–	Addition of nanocellulose from cocoa bean shell increased thermostability, solubility, and soil biodegradability	[67]
Persian gum Gelatin	Complex coarcervation approach	–	The composite film showed enhanced water vapor barrier properties and tensile strength however water solubility and elongation at break was reduced	[68]
Chitosan Polycaprolactone	Solution casting method	2% aqueous acetic acid solution	Addition of chitosan improved the tensile strength, elongation at break, tensile modulus, and water vapor permeability as 30 MPa, 8%, 450 MPa, and 4.7 /mm/m^2/day kPa respectively	[69]

Table 3 (continued)

Types of natural based polymers	Method of preparation	Plasticizers	Properties of the packaging materials improved	References
Starch Polyvinyl alcohol Sweet lime juice	Solution casting method	Glycerol	Film made from 1.5/1.5/0 sweet lime juice/polyvinyl alcohol/starch have better moisture content, solubility, water vapor permeability, transparency, tensile strength and elongation at break as 36.67%, 85.09%, 2.5410^{-6} g/pa h m, 1.98, 509.32 kPa, and 11.31% respectively	[70]
Polylactic acid Zein Eugenol oil	Solution casting method	20% polyethylene glycol	Addition of polylactic acid to zein increased elongation at break, mechanical and barrier properties, however, it reduced tensile strength, water vapor and gas permeability. Eugnol oil inhibited the growth of S.aureues and E.coli	[71]
Soy Protein isolate Carvacol	Casting	Glycerol	Addition of carvacrol increased water vapor permeability, transform protein structure from β-sheet to α-helical structure, and high antimicrobial activities against *L.grayi*	[72]
Polylactic acid Curcumin and Fenugreek essential oil	Casting	–	The incorporation of curcumin and fenugreek essential oil into polylactic acid improved UV-blocking, tensile strength, surface color, flexibility, thickness, and water contact angel. However, reduced water vapor permeability	[73]
Corn starch Orange peel powder	Casting	–	Corn starch containing 40% orange peel powder has highest 1.41 MPa and 11.12% tensile strength and percentage elongation at break	[74]
Rice starch Carrageenan	Phase inversion method	–	Films containing 1.5% and 2% Carrageenan and rice starch has improved the mechanical and water vapor permeability	[75]
Carboxymethyl cellulose Polyvinyl alcohol Fish gelatin	Casting method	–	Addition of 10% carboxymethyl cellulose or polyvinyl alcohol improved mechanical strength, water vapor barrier and water solubility properties of gelatin film	[76]
Polybutylen adipate-co-terphathalate Thermoplastic starch Zinc oxide	Blown extrusion method	–	Zinc oxide (5%) increased amorphous starch content The films increased the shelf life by three days under refrigeration	[77]
PBAT Cassava starch	Blown film extrusion	Glycerol	Increased addition of hydroxypropyl groups in cassava thermoplastic starch blended with polybutylene adipate terephatalate enhanced clarity by 3.2 times, higher clarity, improved elongation by 210%, and reduced water vapor permeability by 34%	[78]

Table 3 (continued)

Types of natural based polymers	Method of preparation	Plasticizers	Properties of the packaging materials improved	References
Starch Chicken gelatin	Casting method	25% w/w glycerol	Increased ratio of gelatin increased strain (2.9 to 285.1%), water vapor permeability, crystallization temperature (190 to 206 °C) but decreased tensile strength (14.6 ± 0.5 MPa to 1.5 ± 0.3 MPa) water solubility	[79]
Rice starch Oregano essential oil	Casting solution method	Glycerol	Film made from starch (3%), glycerol (30%) and Oregano essential oil (4.5%) has lower water permeability (3.7 g mm/kPa/m^2/day), solubility (24%), and high tensile strength (4.4 MPa)	[18]
Bacterial cellulose Fish gelatin Cinnamon essential oil	Casting solution method	–	Increased addition of cinnamon essential oil has increased surface hydrophobicity (120%) and elongation at break. The barrier performance against oxygen, carbondioxide and water vapor enabled the films to be used as modified atmospheric packaging	[80]
Cellulose nanocrystals Pearl millet starch	Solution casting process	30% glycerol	The films improved for crystallinity, tensile strength, young-modulus moisture barrier properties	[81]
Cellulose nanocrystal Gelatin Polyvinyl alcohol	Solution casting method	30% glycerol	The nanocomposite film had tensile strength of 7–14 MPa, elongation at break 45%-81%, its thermal stability increased to 385 °C, the storage modules of 3 GPa with glass transition temperature shift from 78 °C to 98 °C, water vapor permeability decrease from 2.35×10^{-6} g/(m•h•Pa) to 1.64×10^{-6} g/(m•h•Pa), and moisture uptake reduced from 22.5% to 19.05%	[82]
Cellulose nanofiber Savory essential oil	Solution casting method	7% glycerol	Film made from 3% cellulose nanofiber and 3% savory essential oil enhanced glass transition temperature, tensile strength and water barrier properties, and antimicrobial against five food borne pathogens	[83]
Micro-nanocellulose Nanocellulose Cassava starch Polyphenols	Casting method	–	Addition of micro-nanocellulose and nanocellulose to cassava starch reduced water vapor transmission rate, improved hydrophobicity and DPPH free radicals has increased	[84]
Sodium alginate Lignin	Solution casting method	1.26 g of glycerol per 100 g	Films made from alginate incorporated with lignin had lower water solubility (26%), tensile strength (0.146 MPa), glass transition temperatures, and cytotoxicity. However, the antimicrobial and light barrier to prevent ultraviolet-induced lipid oxidation of food	[85]

Table 3 (continued)

Types of natural based polymers	Method of preparation	Plasticizers	Properties of the packaging materials improved	References
Konjac glucomannan Pullulan Acai berry extract	Solution casting method	-	Addition of pullulan increased tensile strength (21.25 to 50.27 MPa), elongation at break (10.64 to 19.19%), whereas addition of acai berry extract improved flexibility, UV-shielding, thermostability, antioxidant, antibacterial, and decreased water vapor permeability (2.07 to 1.67 g●mm/m2●day) and tensile strength	[86]

conditions during the entire storage period by incorporating additives into packaging film or within packaging containers [93, 94]. It embeds components into the packaging that are able to release or absorb substances from or into the preserved food or the surrounding environment to maintain quality and prolong shelf life. Active packaging contains emitter (N_2, CO_2, ethanol, antimicrobials, and antioxidants) or scavenger (O_2, odor and ethylene) gasses during packaging, storage and distribution [94]. AP systems can be divided into active-releasing systems (emitters) adding compounds into the headspace or to the packaged food and active scavenging systems (absorbers) removing undesired compounds from the food or its environment [95]. Active packaging release or absorb substances into or from the packaged food or environment surrounding the food package. Active packaging employs moisture absorber, oxygen scavengers, carbon-dioxide emitters, antimicrobial packaging, flavor and odor absorbent packaging [96]. This innovative packaging system relies on the positive interaction between the product, packaging, and the environment, mediated by a component that enables the release or absorption of substances into/from the packaged food or the environment surrounding the food product [95].

Intelligent Packaging

Intelligent packaging is defined as 'a packaging system able to perform intelligent function such as sensing, detecting, recording, tracking and communicating. This packaging system can be used to facilitate monitoring of freshness, decision making and provide information whenever there is changes in food quality and warn of possible problems [97]. Intelligent packaging is defined as a type of packaging that monitor the situation of packed food or environment inside the packaging such as pH, temperature, moisture level, gaseous composition and spoilage metabolites to provide information about chemical, biochemical, physical, and microbiological quality to the consumers. It monitors the condition of packaged foods and provides information on the quality of the packaged food during transport and storage. It is an extension of traditional packaging responsible for communication with consumers based on their ability to detect and record changes occurring in food or the packaging environment [23].

In intelligent packaging, materials are deliberately added to interact with the packaging environment and monitor the state (temperature, storage time, shelf life, etc.) of the packaged food products. IP can use sensors or indicators that either monitor the environmental conditions influencing the quality attributes (time–temperature indicators (TTIs), gas leakage indicators), monitor quality attributes and indicator compounds of the product itself (biosensors, freshness sensors and indicators, microbial growth indicators), or use data carriers for a more effective communication between the product and the consumer (radio-frequency identification (RFID) tags, antitheft devices) [94, 97]. Intelligent packaging used for monitoring of fish freshness is divided into three categories: freshness sensor, freshness indicator and radio frequency identification tag. Radio frequency identification labels is composed of tags and card readers, a communication technology that uses wireless sensors and data reading to monitor the freshness of fish products [97]. Fish freshness quality attributes can be assessed using sensory, chemical, physical and microbial methods. Appearance, color or texture of fish products is measured using Quality Index method. Chemical attributes like ATP, *K*-value or Total volatile basic nitrogen/TVB-N, Biogenic amine or lipid oxidation can be measured using Kjeldahl method, steam distillation, Fourier Transform Infrared Spectroscopy, GC–MS or HPLC. Physical attributes of fish freshness like color, texture or conductivity are measured using physical property analyzer or color difference meter. Finally microbial characteristics of fish products can be asses using specific spoilage organisms (SSO) [97].

Smart Packaging

Smart packaging is an amalgamation of active and intelligent packaging. On one hand it monitors changes in a product or its environment (intelligent packaging) and on the other hand acts upon these changes (active packaging). Smart packaging uses a variety of sensors for monitoring food quality and safety by detecting and analyzing freshness, pathogens, leakages, carbon dioxide, oxygen, pH level, time or temperature. It is used to extend shelf life, monitor freshness, display information on quality, and improve product and customer safety. Smart packaging monitor the quality of the food in real time and inform food conditions to the consumers by emitting a signal (Colorimetric, optical, chemical, electric etc.) in response to any changes within the packaging conditions and food quality [98].

Currently researchers are exploring natural pigments derived from plants and wasted food as indictors in biodegradable packaging [99]. The frequently studied natural pigments are Curcumin, anthocyanins, shikonin, betalain and alizarin [100]. Among these natural pigments, red-violet anthocyanins are the most intensively studied pigments for their ability to change color due to pH changes in packaged environment. It can be extracted from many plant species including leaves, stems, rhizomes, roots, fruits, flowers and seeds [100].

Conclusion and Future Perspective

The research related to biopolymers as packaging materials is just in early infant stage, but yet promising and enhanced chemical branch and methods are continuously being innovated. Thus, it can be concluded that biodegradable polymers have great potential in food packaging applications and also do not harm the environment by breaking down into the organic matter. Biopolymers help in reducing the environmental impact of plastic production, processing and in a way lead toward green economy. The use of bio-based polymers is increasing at a rapid pace for packaging of food and other applications. Several studies have been conducted describing the development of food packaging materials from biomaterials, especially polysaccharides and proteins. Biodegradable packaging films can be used as active and intelligent packaging. Films incorporated with naturally derived pigments from plant sources can offer a simple, easy to fabricate, economical, non toxic, safe and cost effective solution for continuous, real time monitoring of fish products with an opportunity for commercialization. Plant bioactive molecules give antioxidant, antimicrobial properties and enhance the interaction of polymer matrix. The incorporation of plant extracts into films is a promising method to prevent or reduce the deterioration of food quality, thus contributing to preserving and extending the shelf life of food. Future researches have to be more focused on adding value to the packaging materials, i.e., use of nanotechnology, smart sensors, etc., which will not only maintain the integrity, but also communicate the information about the product to the consumers.

Acknowledgements The authors are grateful for continuous encouragement to staff members of Jimma University, College of Agriculture and Veterinary Medicine, Postharvest Management Department.

Funding No external funding was used.

Declarations

Conflict of interest No potential conflict of interest was reported by the author(s).

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