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Bio-based smart materials for fish product packaging: a review

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ABSTRACT

Conventional packaging offers protection, containment, communication, and convenience to packaged food. The most commonly used packaging materials are petrochemical-based plastics which generate massive wastes that persist for a long time in the environment after their use. Bio-based materials are the best option to replace this synthetic plastic. This review presents the importance of packaging fish products using polysaccharides, proteins, polyhydroxyalkanoates, polylactic acids, pullulan, and xanthan gums loaded with different nanofillers and bioactive molecules. Bio-based smart materials easily decompose into carbon dioxide, methane, water, and inorganic compounds. Biopolymers can be produced from natural biomass, bio-monomers, and microorganisms. These biopolymers demonstrate excellent physiochemical, thermal, and mechanical properties when mixed or alone as fish packaging materials. Integration of nanofillers and bioactive molecules improves mechanical, gas barrier, antioxidant and antimicrobial properties of bio-based materials. Bioactive molecules like anthocyanins, betalains, curcumin, and clove oil are sensitive to pH, temperature, light, and time. Bioactive molecules can be loaded into bio-based packaging materials to monitor the real-time freshness of fish products during storage. It is concluded that bio-based smart materials have the potential for fish packaging, do not harm the environment, and easily interact with nanofillers and bioactive molecules.

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Introduction

Globally 179 million tons of fish were produced from captured fisheries and aquaculture in 2018.^[1] Fisheries play an important role in food security and create job opportunities for 59.5 million people across the world.^[2] With business-as-usual practices in Africa, the fish sector was estimated to create 20.7 million jobs by 2030 with an annual value of 3.3 billion USD.^[3] Fish is an excellent source of proteins, omega-3 fatty acids, calcium, phosphorus, iron, zinc, magnesium, vitamin A and iodine.^[4] However, fish products are highly perishable and need to be preserved in appropriate packaging for handling, distribution, and export.^[5] Plastic is the principal food-packaging material. Most (70%) of plastics wastes is generated from food packaging.^[6] Plastics are durable, cheap, strong, a good barrier to moisture, light, and highly processable into different shapes. However, the utilization of synthetic plastics leads to dispute as it resulted in depletion of petroleum reserves, global warming, and environmental

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pollution.^[7] The usage of petrochemical-based packaging materials is a risk to environmental safety and human health. Moreover, synthetic plastics are hardly recycled or have limited recyclability. Even though pyrolysis and hydrothermal processing convert plastic waste into fuel, its application is limited due to the emission of toxic gaseous during high-temperature combustion of plastics.^[8] In recent years, enzymatic and microbial biodegradation of synthetic plastics has started but the possibility to apply biological treatment is limited due to the difficulty in adhering and colonizing microbes on the surface of plastics. Generally, recycling, landfilling, and incineration are ineffective for the management of plastic wastes.^[9] The prevailing environmental pollution has pushed to explore sustainable packaging.^[10] An alternative option to this synthetic plastic is biodegradable polymers. It is a fact that these plastic food-packaging materials are disposable and non-biodegradable. Consequently, large amounts of plastic are accumulating in the terrestrial and aquatic ecosystems, hence various sectors like fishery, agriculture, hydroelectric power generation, marine transportation, health, and tourism are in danger.^[11] With the ever-increasing concern of dumped petrochemical-based packaging wastes in the environment, green and sustainable materials are highly demanded to reduce the harmful effects of plastics on the environment.^[12] Contrary to synthetic plastics, bio-based polymers decompose, degrade, return to the soil and decrease the volume of garbage. Bio-based materials are derived from renewable and sustainable biomass.^[12] Bio-based materials turn the bioplastics sector from a wasteful linear economy to a circular economy.^[9] Biopolymers derived from bio-based materials are categorized into natural, semi-synthetic, and microbial polymers.^[13] Polysaccharides, protein, lipids, polylactic acid, polyhydroxyalkanoate, pullulan, and xanthan gum are well-known bio-based materials due to their ability to form three-dimensional polymer networks. Polysaccharides are further categorized as starch, cellulose, lignin, κ -carrageenan, alginate, pectin, and chitosan. Protein is grouped into gelatin, casein, caseinates, whey protein, soy protein isolate, wheat gluten, and corn zein.^[14] Polymers produced by microorganisms are further classified as polyhydroxyalkanoate, pullulan, xanthan gum.^[13] Polylactic acid is synthesized from lactic acid that is produced by microbes through lactide formation, then condensation polymerization and ring open polymerization.^[15] Several studies indicated there is a great possibility of bio-based materials for developing eco-friendly fish packaging alternatives.^[16,17] This article aims to review the application of different bio-based polymers embedded with bioactive molecules for fish packaging materials.

Polysaccharides-based biopolymers for fish product packaging

The most widely used polysaccharide biopolymers in fish packaging are cellulose, chitin/chitosan, starch, lignin, pectin, alginate, and κ -Carrageenan.^[18] In addition to food packaging, bio-based materials can be applied to the filtration of microbes, military security, and environmental management. Today numerous polysaccharides-based packaging materials have been developed for fish packaging materials (Table 1). Polysaccharides are easily modified to improve their physicochemical properties through heat gelatinization, pH changes, cross-linking, and hydrolysis.^[19] Gases barrier, mechanical, thermal, and chemical properties of biomaterials are important criteria to select polysaccharides as packaging materials.^[20] The water vapor, gas barrier, structural and mechanical properties of biopolymers can be improved by adding bioactive, nanofillers, cross-linkers, or plasticizers.^[21] Polysaccharide-based packaging material has several advantages as compared to other biopolymers. It easily forms covalent or non-covalent interactions networks with other polymers. They are compatible with additives, which improves the functionality of polysaccharide-based film.^[22]

Starch-based biopolymer is a widely accepted packaging film due to its availability in wide agricultural sources, ease of extraction, does not affect the sensory properties of food and can be edible without causing health risks to a human.^[10] Bio-based materials extruded from starch conserves frozen pack fish fillets for 360 days better as compared to low-density

Table 1. Polysaccharides-based biopolymers for fish product packaging

Polysaccharides	Bioactive molecules	Applications	References
Rice starch	Oregano essential oil (OEO)	<ul style="list-style-type: none"> Fish fillet packed in starch/OEO showed less oxidation and microbial growth 	[19,23]
Cassava starch	Citrus lemon peel extract (CLPE)	<ul style="list-style-type: none"> Fish packaged in a film containing 20% CLPE demonstrated low peroxide and Total Volatile Basic Nitrogen (TVB-N) values 	[24]
Starch/Pectin	Feijoa extracts	<ul style="list-style-type: none"> Antimicrobial against <i>E. coli</i>, <i>Salmonella</i> and <i>Shigella</i>. Increase the shelf life of perishable food 	[25]
Chitosan nanoparticles/collagen protein	Anthocyanidin and Cinnamon-perilla essential oil	<ul style="list-style-type: none"> Fillets of sea bream fillets wrapped in the composite film extended shelf life to 6–8 days 	[26]
Polycaprolactone Chitosan Rutin (PCL-CS-R)		<ul style="list-style-type: none"> PCL-CS-R films decreased the microbial population of <i>E. coli</i>, <i>S. aureus</i>, and <i>L. monocytogenes</i> by 18.39%, 19.27%, and 17.45% as compared to polycaprolactone film alone in rainbow trout packaging 	[27]
Cellulose nanofiber /Carboxymethyl cellulose	Shikonin	<ul style="list-style-type: none"> Discoloration of shikonin at weak acid and weak alkali to monitor fish freshness 	[28]
Pectin/Sodium alginate/Xanthan gum	Raspberry pomace extracts	<ul style="list-style-type: none"> Changes its color (pH-sensitive) used to monitor protein-rich foods 	[29]
Pectin	Oregano essential oil, Ginger essential oil	<ul style="list-style-type: none"> Significant protective effect on protein oxidation, prevention of endogenous enzyme activity Enhanced shelf life of yellow croaker for 7 days during ice storage. 	[30]
Pectin/Chitosan	Tarragon essential oil	<ul style="list-style-type: none"> Decreased bacterial deterioration of seafood during chilling 	[31]
Chitosan	10% purple tomato anthocyanins	<ul style="list-style-type: none"> Used to indicate the freshness of milk and fish/ monitoring food freshness/spoilage (intelligent film) 	[32]
Chitosan/ Methyl cellulose	Saffron petal anthocyanins	<ul style="list-style-type: none"> Antimicrobial against <i>E. coli</i> and <i>S. aureus</i>, antioxidant activity, and monitor lamb meat freshness during storage (smart packaging) 	[33]
Chitosan/ Methyl cellulose	Anthocyanin	<ul style="list-style-type: none"> A potent antioxidant, antibacterial against (<i>E. coli</i>, <i>P. aeruginosa</i> and <i>S. aureus</i>) and monitors fish fillet freshness at room temperature (smart packaging) 	[34]
Bacterial cellulose	Pelargonidin dye	<ul style="list-style-type: none"> Monitors changes in total volatile base nitrogen (TVB-N) amounts of tilapia fillets stored at 4°C and 25°C 	[35]
Sodium alginate	Purple onion peel extract (POPE)	<ul style="list-style-type: none"> POPE increased antioxidant activity, the thickness of the alginate-based films, extend the shelf life of foods with high water content and susceptible to lipid oxidation 	[36]
Sodium caseinate	1% TiO ₂ , Rosemary Essential oil (REO)	<ul style="list-style-type: none"> Exhibited antimicrobial by reducing the <i>psychrotrophic bacteria count</i> at the levels of 1% TiO₂ and 2% REO. Reduced lipid oxidation, and lipolysis of lamb meat during refrigerated storage for 15 days 	[37]
K ⁻ Carrageenan	CuO, TiO ₂	<ul style="list-style-type: none"> Good active food packaging films retained high-quality bananas (weight loss, firmness, and surface color) 	[38]
K ⁻ Carrageenan	Red cabbage extract (<i>Brassica oleracea</i>)	<ul style="list-style-type: none"> Helped to monitor freshness and quality of rainbow trout fillets (<i>Oncorhynchus mykiss</i>) because of color change when quality deteriorates 	[39]

polyethylene packaging.^[40] The application of cellulose alone as packaging materials is challenging due to its hydrophilic and crystalline nature. However, the derivatives of cellulose like carboxymethyl cellulose, hydroxypropyl methylcellulose, or methylcellulose hydroxypropyl all have good film-making properties.^[41] Packaging films made of CMC- tea waste and furcellaran inhibit the growth of microorganisms and accumulation of biogenic amine, hence extending the shelf-life of salmon fillets.^[42] Cellulose has intelligent properties, i.e. can visually indicate the condition of packaged food when loaded with other bioactive molecules. Cellulose when embedded with cyanidin-3-glucoside can act as intelligent packaging that can indicate the freshness of tilapia fillets stored at 4°C and 25°C.^[43] Chitosan and other polymers increase the shelf life of fish and its products under chilling conditions. Accordingly, the film made from chitosan-alginate has increased the shelf life of catfish by 15 days during refrigeration.^[44] The

chitosan and chitosan-alginate coatings are recommended for refrigerated storage of catfish fillets. Similarly, chitosan when combined with the Lactoperoxidase system increases the shelf life of fish burgers by 5 days^[45] K- Carrageenan, alginate, and agar are widely used to develop coatings and films due to their gel-forming abilities.^[46] Oregano essential oil (0.4%) added into pectin increases the shelf-life of yellow croaker from 20 to 27 days.^[47]

Protein-based biopolymers for fish product packaging

Protein-based polymers are helpful because they possess valuable characteristics for the production of food packaging materials. Proteins have been intensively studied as bio-based packaging materials due to their film-forming ability, UV-barrier properties, mechanical properties, transparency, and barrier properties against oxygen and carbon dioxide diffusion.^[22] Gelatin, collagen, fish proteins, corn zein, wheat gluten, whey, and soy protein are widely accepted for film formation due to their nature to form intermolecular bonds.^[48] Different proteins and other biopolymers-based fish packaging materials incorporated with different bioactive molecules are presented in Table 2.

Table 2. Proteins-based biopolymers for fish product packaging.

Proteins	Bioactive molecules loaded	Applications	References
Gelatin	Eugenol nanofiber and Polylactic acid	<ul style="list-style-type: none"> Excellent antimicrobial and antioxidant biodegradable films 	[49]
Gelatin/ Kappa-Carrageenan	Anthocyanin and TiO ₂ nanoparticles	<ul style="list-style-type: none"> Changes its color based on changes in pH, act as antimicrobial, antioxidant and monitors fish freshness during storage 	[50]
Fish gelatin	Haskap berries (<i>Lonicera caerulea</i> L.) extract (anthocyanins and phenolic acids as major extract)	<ul style="list-style-type: none"> Fish gelatin containing 1% of haskap berries extract showed antioxidants and color change as a function of the pH of intelligent packaging to monitor the freshness of shrimp 	[51]
Gelatin/ Oxidized chitin nanocrystals	Black rice bran anthocyanins	<ul style="list-style-type: none"> Antioxidant activity and have great potential in monitoring freshness of shrimp and hairtail (smart packaging), high protein foods 	[52]
Gelatin/Chitosan (carboxymethyl chitosan)	0.5 mg/mL squid maillard peptides	<ul style="list-style-type: none"> Retarded lipid oxidation and microbial growth of bluefin tuna 	[53]
Gelatin/Chitosan	0.4% TiO ₂ - Ag	<ul style="list-style-type: none"> Improves antibacterial and UV barrier capability 	[54]
Gelatin/Chitosan	ε-Polylysine	<ul style="list-style-type: none"> Effective controlled six foodborne pathogens (<i>E. coli</i>, <i>K. pneumoniae</i>, <i>S. enteritidis</i>, <i>P. aeruginosa</i>, <i>S. aureus</i>, and <i>L. monocytogens</i>) 	[55]
Gelatin	Curcumin, betanin and anthocyanin	<ul style="list-style-type: none"> Demonstrated superior color retention as compared to curcumin and anthocyanins color change with pH change application for intelligent packaging to indicate freshness and quality of foodstuffs such as seafood products 	[56]
Soy Protein Isolate/Gelatin	Essential oils of <i>Zataria multiflora</i> and <i>Cinnamon zeylanicum</i>	<ul style="list-style-type: none"> The population of <i>S.aureus</i>, <i>B.cereus</i>, <i>S.typhimurium</i>, <i>L.monocytogens</i>, and <i>E.coli</i> was diminished via the inclusion of 20% <i>Zataria multiflora</i> active food packaging, extended shelf life 	[57]
Gelatin	Dialdehyde kappa-carrageenan (DAK-car) Thymol-loaded zein nanoparticles (ZNP)	<ul style="list-style-type: none"> Films containing a 1:2 ratio of gelatin films (GEL)/DAK- car and thymol-loaded zein 0.25 and 0.5 mg/mL of thymol have antioxidant and antimicrobial activity in active food packaging 	[58]
Whey protein isolate (WPI)	Emulsions and nanoemulsions of <i>Citrus sinensis</i> essential oil peel extract (CSEO)	<ul style="list-style-type: none"> The film preserves food against oxidation and microbial spoilage improving shelf life and quality of foodstuffs 	[59]
Gelatin	Oxidized guar gum green tea extracts (0.5 and 1.0% w/v).	<ul style="list-style-type: none"> Antimicrobial against <i>S.aureus</i>, higher antioxidant activity due to the presence of green tea extract 	[60]

Gelatin is one of the first biopolymer materials proposed to carry bioactive compounds.^[18] The fillets of common carp when coated with gelatin have improved the qualitative and sensory characteristics of fillets as compared to polyethylene plastics during freeze storage.^[61] Gelatin when combined with other biopolymers has some synergistic effects to control some foodborne bacterial pathogens. Gelatin-chitosan films enriched with rosemary essential oils have a great potential for controlling *Salmonella enteric*, *Campylobacter jejuni*, *Pseudomonas aeruginosa*, and *Escherichia coli*.^[62] Soy protein isolates incorporated with nanoparticles (1% CuO and TiO₂) decrease water vapor permeability of films, thus, can be applied as an active packaging system for different foodstuffs.^[63] Coating of packaging materials with whey protein isolates exhibits high oxygen barrier properties and delayed peroxide formation of frozen shrimp fried rice during six months of frozen storage.^[64] The addition of small amounts of chitosan in whey protein isolate creates a stronger network structure through complex coacervation, higher tensile strength, and lower water vapor permeability rate as compared to mono-component chitosan and whey protein isolate.^[65] Due to its hydrophobic nature, Zein protein has low water vapor permeability and solubility.^[14]

Polylactic acid and polyhydroxyalkanoates-based biopolymers for fish product packaging

Poly(lactic acid) (PLA) is synthesized from lactic acid monomers that can be widely used for fish packaging during chilling storage.^[66] Poly(lactic acid), polyhydroxyalkanoates, pullulan, and xanthan gum are the most widely used microbial polymers for fish packaging (Table 3). The properties that make PHAs ideal for food packaging materials are stability in the air, nontoxicity, hydrophobicity, and pure enantiomer.^[67]

Integration of nanofillers into biopolymers

Nanofillers are nano-sized materials that maintain the microbiological safety of food, improve the mechanical, thermal, oxygen, and moisture barrier properties of bio-based packaging of meat, fruits, and seafood in active packaging.^[78] Montmorillonite, nanofibers, nanowhiskers, silver, copper, zinc oxide, and Titanium dioxide are widely used nanofiller in active packaging Table 4.

Integration of bioactive molecules into biopolymers

The incorporation of bioactive molecules improves the mechanical, barrier, antioxidant, and antimicrobial properties of polymers.^[79] Bioactive molecules change their color based on changes in pH, temperature, light, time, and ammonia that making which acts as an indicator in monitoring the spoilage of packaged fish products.^[8] Fish spoilage is usually monitored by identifying adenosine triphosphate decomposition, microbial plate counts, total volatile base nitrogen, lipid oxidation, and sensory properties. Monitoring the real-time freshness of fish using chemical, physical, biological, and sensory methods requires professional staff, tedious, complicated, time-consuming, and destructive. The emerging techniques use halochromic (pH-sensitive) bioactive molecules that enable consumers and non-specialists to understand the freshness conditions of fish using their naked eyes.^[80]

Some of the pH-sensitive bioactive molecules (Table 5) are anthocyanins, betalains, carotenoids, carotenoids clove oil, curcumin, turmeric, rosemary, oregano, thyme, green tea, sage, basil, ginger, coriander, garlic, nutmeg, mace, savory and fennel.^[81] The most intensively studied bioactive molecules as colorimetric indicators are anthocyanins,^[82] curcumin,^[83] and carotenoids.^[84] Anthocyanins incorporated into biopolymer films have been used as smart indicators to monitor fish spoilage in Grass carp,^[85] rainbow trout,^[86] Hair-tail, Spade nose shark,^[84] Pangasius,^[87] and Atlantic mackerel.^[88] The other widely used bioactive molecule is betalains. They are secondary nitrogen-containing metabolites of plants, water-soluble is responsible for yellow and orange to red-purple and



Table 3. The use of polylactic acids and polyhydroxyalkanoates-based biopolymers for fish product packaging.

Polymers	Bioactive molecules loaded	Role of the packaging films	References
Polylactic acid	Cinnamaldehyde/ Tea polyphenol	<ul style="list-style-type: none"> Damaged the cell wall membrane leading to change in cell membrane permeability, protein synthesis, and expression of <i>Shewanella putrefaciens</i> fresh-keeping material with antibacterial properties 	[68]
Polylactic acid (PLA)	Nanocellulose/ Chitosan	<ul style="list-style-type: none"> Poly(lactic acid) coated with nanocellulose/chitosan by showing the color change with spoilage progress has the potential to monitor spoilage in beef and other meat products refrigerated at 4°C 	[69]
Polylactic acid	ZnO	<ul style="list-style-type: none"> Poly (lactic acid) (PLA)/ZnO nanocomposite films fabricated using ultra-high pressure has a great potential for food packaging with reduced permeability toward oxygen and water vapor 	[70]
Polylactic acid	Nanocellulose	<ul style="list-style-type: none"> An Indian anchovy (<i>Stolephorus indicus</i>) fish packed in PLA/nanocellulose demonstrated increased shelf life as compared to neat PLA and LDPE during chilling storage for 20 days 	[71]
Poly(lactic acid (PLA) / Sawdust (SD) particles	Bacteriocin from <i>Weissella hellenica</i> BCC 7293 was diffusion coated onto PLA/SD	<ul style="list-style-type: none"> Retarded the growth of Gram-positive (<i>Listeria monocytogenes</i>, <i>Staphylococcus aureus</i>) and Gram-negative bacteria (<i>Pseudomonas aeruginosa</i>, <i>Aeromonas hydrophila</i>, <i>Escherichia coli</i>, and <i>Salmonella typhimurium</i>) antimicrobial packaging for pangasius fish fillets 	[72]
Poly (butylene adipate co-terephthalate)–PBAT	Oregano (<i>Origanum vulgare</i>) essential oil (OEO)	<ul style="list-style-type: none"> Revealed antibacterial activity (reduced growth of coliforms, <i>Staphylococcus aureus</i>, and psychrotrophic microorganisms) and prolonged storage quality of fish fillet for 10 days 	[73]
Poly(lactic acid (PLA)-polyhydroxybutyrate (PHB)	Cinnamaldehyde (5%)	<ul style="list-style-type: none"> PLA-PHB films are active packaging films that can substitute plastic for chilled salmon storage and reduced the total bacterial counts of salmon dices 	[74]
Poly(butylene adipate-co-terephthalate (PBAT)	Silver montmorillonite (Ag-MMT)	<ul style="list-style-type: none"> PBAT/Ag-MMT reduced water and oxygen permeabilities, antibacterial and extended shelf life of sea bream for 15 days 	[75]
Pullulan and sodium carboxymethylcellulose (PUL–CMC)	Gallic acid (GA)/ Epsilon-poly-L-lysine (PL) hydrochloride	<ul style="list-style-type: none"> Inhibit protein degradation, microbial growth and lipid oxidation of stored sea bass (<i>Lateolabrax maculatus</i>) fillet during storage at 4°C for 20 days (synergistic effect of GA and PL) 	[76]
Gellan gum (GM) heat-treated soy protein isolate (HSPi)	<i>Citriatariatea</i> (CT) extract	<ul style="list-style-type: none"> Gellan gum improves the release of CT anthocyanins and HSPi regulated its release rate. The films have antimicrobial, antioxidant, and change of color on spoilage progress and hence help to monitor shrimp spoilage 	[77]

Table 4. Nanofillers frequently embedded into polymers.

Nanofillers	Embedded polymers	Characteristics of nanofillers	Mechanisms of action of nanofillers	References
TiO ₂ /Ag	Rhinobatoscemiculus gelatin	Antioxidant properties	<ul style="list-style-type: none"> Embedding 4% TiO₂-Ag nanoparticles into gelatin film gave good metal chelating and DPPH scavenging activity 	[93]
TiO ₂ /ZnO	4A-Zeolite Gelatin	Antimicrobial properties	<ul style="list-style-type: none"> TiO₂ combined with ZnO disrupted the microbial cell membrane and emitted reactive oxygen species damaging phospholipids molecules in the cell membrane 	[94]
Se/ZnO	Gelatin/Cellulose nanofibers	Antioxidant and antimicrobial activity	<ul style="list-style-type: none"> Antimicrobial against <i>Listeria monocytogenes</i> > <i>Escherichia coli</i> > <i>Staphylococcus aureus</i> > <i>Pseudomonas fluorescens</i> through releasing ions and catalysis the formation of active oxygen species Antioxidant through donating electrons from oxygen atoms of nanoparticles to nitrogen atoms odd electron in DPPH 	[95]
TiO ₂	Poly(lactic acid)Modified with Lycopene	Antimicrobial and antioxidant activity	<ul style="list-style-type: none"> Due to lack of lipoprotein in the cell membranes, it can easily damage the cell wall and enter into the cell of gram-positive bacteria. Antimicrobial (<i>Escherichia coli</i> and <i>Staphylococcus aureus</i>) and antioxidant film with suitable color properties for active and intelligent packaging in food 	[96]
ZnO	Chisosan/Melissa officinalis essential oil	Antimicrobial and antioxidant	<ul style="list-style-type: none"> Due to the presence of a positive charge in chitosan polymer, citronellal and geraniol in Melissa essential oils increases the permeability of the cytoplasmic membrane to ATP which leads to cell death (antimicrobial biodegradable composite film for food packaging) 	[97]
TiO ₂	Gelatin/Grape fruit extracts	Antibacterial against <i>E. coli</i> , <i>L. monocytogenes</i> and antioxidant	<ul style="list-style-type: none"> Polyphenol and α-tocopherol cause cell death due to the accumulation of reactive oxygen species which leads to a change in the membrane permeability and alters metabolic mechanisms 	[98]
ZnO/SiO ₂	Chitosan	Antibacterial	<ul style="list-style-type: none"> UV-barrier, antimicrobial (<i>Escherichia coli</i> and <i>Listeria monocytogenes</i>) and antioxidant activity Mold growth of <i>Escherichia coli</i>, IRAQ 3 and <i>Staphylococcus aureus</i>, S33R has been prevented via the inclusion of 5% ZnO-SiO₂ 	[99]
TiO ₂	K-CarrageenanXanthan gumGillam gum	Antibacterial	<ul style="list-style-type: none"> Protection from UV light, partial antimicrobial activity against <i>Staphylococcus aureus</i> 	[100]
ZnO	Poly(lactic acid)	Antibacterial	<ul style="list-style-type: none"> Exhibited antibacterial activity against <i>Escherichia coli</i> and <i>Listeria monocytogenes</i> during storage of minced fish paste 	[101]
ZnO	Carboxymethyl celluloseGrape seed extract (5 wt%)	Antibacterial	<ul style="list-style-type: none"> Exhibited UV-light and water vapor barrier properties Zinc oxide nanoparticles increased water and mechanical properties Antibacterial against <i>L.monocytogenes</i> and <i>E.coli</i> It preserves high fat meat products Less water vapor barrier and antioxidant activities 	[102]

Table 5. The beneficial effect of incorporating bioactive compounds into various polymers.

Bioactive molecules	Embedded polymers	Benefits of bioactive molecules	Mechanisms of action of bioactive molecules	References
<i>Echium amoenum</i> anthocyanins	Bacterial cellulose film	<ul style="list-style-type: none"> Color change spotted to identify freshness/ spoilage of shrimp for fresh (violet), use soon (gray), and spoiled (yellow) shrimp 	<ul style="list-style-type: none"> As pH increases anthocyanin's structure changes from flavylium to quinoidal, eventually changing its color from blue-purple to green and yellow 	[103]
Gallic acid Clove oil	Gelatin Chitosan	<ul style="list-style-type: none"> Gelatin-chitosan-gallic acid-clove oil prolonged the shelf life of salmon fillet for five days 	<ul style="list-style-type: none"> Better gas and oxygen barrier properties, inhibition of microbial growth, antioxidant effects of coating 	[104]
Oxidized chitin nanocrystals Curcumin	Chitosan	<ul style="list-style-type: none"> The color of the films was changed due to a change in pH which enabled identify spoilage of the hairtail and shrimp 	<ul style="list-style-type: none"> The mechanical, barrier against light and water vapor had been improved by curcumin – chitin nanocrystal incorporation in the film 	[105]
Betalains and phenolic compounds from amaranthus leaf extract	Gelatin	<ul style="list-style-type: none"> Enhanced the storage quality of fish/chicken from three days for control to 12 days for amaranthus leaf extract treated film 	<ul style="list-style-type: none"> Antioxidant, antimicrobial, and intelligent packaging (visible color change from red to yellow on spoilage) 	[84]
Betacyanin	Glucomanan	<ul style="list-style-type: none"> The film had changed its color from purple to yellow when TVB-N reached 39.74 mg/100 g 	<ul style="list-style-type: none"> Smart packaging during fish <i>Osphronemus gouramy</i> storage to monitor freshness 	[106]
Lyophilized alga <i>Fucus spiralis</i> powder	Gelatin	<ul style="list-style-type: none"> The formation of FFA and growth of proteolytic, aerobes, and psychrotrophic bacteria had been impaired, increasing antimicrobial and antioxidant effects with increased alga in the film 	<ul style="list-style-type: none"> The activity of oxidative phosphorylation and microbial enzyme had been inhibited 	[107]

Table 6. Essential properties of packaging improved due to immobilization of nanofillers/bioactive molecules into polymers.

Polymers/ bioactive molecules/ nanofillers	Properties of biocomposite packaging materials modified	Mechanisms of action of the packaging materials	References
Carboxymethyl-cellulose (CMC) Starch Purple sweet potato anthocyanins (PSPA)	<ul style="list-style-type: none"> New interaction between cellulose and PSPA appeared PSPA was uniformly dispersed in CMC and Starch The addition of PSPA increased tensile strength, decreased elongation at break and moisture content 	<ul style="list-style-type: none"> Changed its color from red to blue and is sensitive to ammonia Intelligent packaging to monitor fish freshness 	[85]
Starch Jackfruit seeds anthocyanins	<ul style="list-style-type: none"> Higher tensile strength, reduced resistance 	<ul style="list-style-type: none"> Fish freshness indicator Intelligent packaging 	[108]
Starch Roselle anthocyanins	<ul style="list-style-type: none"> Anthocyanins were immobilized into starch film, water content and tensile strength decreased, elongation at break increased 	<ul style="list-style-type: none"> The film was sensitive to ammonia and monitors fish freshness at 4°C Intelligent packaging 	[109]
Bacterial nanocellulose Black carrot anthocyanins	<ul style="list-style-type: none"> Immobilization of black carrot anthocyanins decreased the mean diameter of nanofibers 	<ul style="list-style-type: none"> Monitors the freshness of common carp and rainbow trout at 4°C. Consumers realize spoilage by the naked eye due to color changes Intelligent packaging 	[110]
Zein nanofibers Alizarin	<ul style="list-style-type: none"> Alizarin is incorporated in to the zein matrix through hydrogen bonding Dehydration temperature, protein unfolding and glass transition reduced 	<ul style="list-style-type: none"> Monitor the freshness of trout through changing its color in response to microbial and color changes 	[111]
Chitosan/ Xylan/ Hydroxyapatite/ Curcumin	<ul style="list-style-type: none"> Immobilization of curcumin into the polymers did not alter the surface of the film 	<ul style="list-style-type: none"> Monitor freshness in Indian oil sardines Smart packaging 	[112]

(Continued)

Table 6. (Continued).

Polymers/ bioactive molecules/ nanofillers	Properties of biocomposite packaging materials modified	Mechanisms of action of the packaging materials	References
Bacterial cellulose Chitosan Curcumin	<ul style="list-style-type: none"> Improved contact angles, mechanical properties, water vapor transmission, lower moisture content, and oxygen transmission rate 	<ul style="list-style-type: none"> Promising active packaging materials 	[113]
Chitosan Rice berry phenolics extract	<ul style="list-style-type: none"> Chitosan- rice berry phenolics extract films exhibited increased thermal stability, mechanical resistance, hydrophobicity, barrier, and antioxidant properties 	<ul style="list-style-type: none"> The film changed its color from orange-red to yellow (naked eye detectable color changes) during shrimp spoilage monitoring 	[114]
ZnO Rosemary extract Anthocyanins Montmorillonite	<ul style="list-style-type: none"> Showed remarkable antibacterial, antioxidant, and air barrier activity 	<ul style="list-style-type: none"> Zn²⁺ ions damage the cell membrane and interact with intracellular contents of <i>E. coli</i> Scavenges free radicals by terminating oxidizing chain reactions 	[115]
Gelatin/Cellulose/ Arabic gum TiO ₂ Garlic extract	<ul style="list-style-type: none"> Enhanced the mechanical, antimicrobial, thermal, water vapor, and oxygen transmission rate 	<ul style="list-style-type: none"> Bionanocomposite film synergistically controlled the growth of microbes during refrigeration of tilapia fillets 	[116]

violet colors. Based on its principal sources this pigment can be categorized into two broad types: betacyanins and betaxanthines.^[89] Curcumin is also a known bioactive molecule found in the rhizome of turmeric (*Curcuma longa* L.) that can change its color when food loses its freshness.^[90]

Important properties of packaging materials

The most crucial properties of packaging materials are tensile strength, water vapor transmission rate, oxygen transmission rate, elongation at break, the thickness of the film, water-solubility, moisture content, mechanical properties, surface morphology, antioxidant and antibacterial properties.^[91] The instruments used to characterize the properties of packaging materials are Fourier transform infrared spectroscopy, Scanning electron microscopy, Differential Scanning Calorimeter, X-Ray Diffraction.^[92] The embedding of nanofillers, bioactive molecules, and essential oils into biopolymer improves the nature of packaging materials (Table 6).

Conclusion and future perspective

Valorization of wasted food into useful packaging materials is the top agenda to avert the effect of synthetic plastics on the environment. Polysaccharides like cellulose, starch, chitosan or chitin, lignin, pectin, alginate, and carrageenan can be extracted from natural biomass. Similarly, proteins like gelatin, casein and caseinates, whey proteins, wheat gluten, soy protein isolate, and corn zein are easily isolated from plants and animals' sources. Polyhydroxyalkanoates, pullulan, and xanthan gum can be produced by microorganisms. All these biopolymers notably demonstrate excellent physico-chemical, thermal, and mechanical properties when mixed or alone to be used as packaging materials. Some of the essential properties like morphological, thermal, gas barrier, water vapor permeability, antioxidant, and antibacterial of packaging films can be improved by immobilizing nanofillers or bioactive molecules. As a result, these biodegradable packaging materials can be applied as active, intelligent, or smart based on the types of natural pigment during fish packaging. These natural plant-based pigments are nontoxic and safe have antioxidant and antimicrobial properties, enhance shelf life, and enable monitoring of the freshness conditions of fish under storage conditions. Future research has to focus on the use of nanotechnology and smart sensors which allow communication information about the product to the consumers.

Disclosure statement

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