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

Alemu Lema Abeltia,b, Tilahun A. Tekaa, Sirawdink Fikreyesus Forsidoa, Metekia Tamiru, Geremew Bultosad, Ashraf Alkhtib, and Emily Burton

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, polyhydroxalkanoates, polyactic 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.

Introduction

Globally 179 million tons of fish were produced from captured fisheries and aquaculture in 2018. Fisheries play an important role in food security and create job opportunities for 59.5 million people across the world. 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. Fish is an excellent source of proteins, omega-3 fatty acids, calcium, phosphorus, iron, zinc, magnesium, vitamin A and iodine. However, fish products are highly perishable and need to be preserved in appropriate packaging for handling, distribution, and export. Plastic is the principal food-packaging material. Most (70%) of plastics waste is generated from food packaging. 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 pollution.\textsuperscript{[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.\textsuperscript{[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.\textsuperscript{[9]} The prevailing environmental pollution has pushed to explore sustainable packaging.\textsuperscript{[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.\textsuperscript{[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.\textsuperscript{[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.\textsuperscript{[12]} Bio-based materials turn the bioplastics sector from a wasteful linear economy to a circular economy.\textsuperscript{[9]} Biopolymers derived from bio-based materials are categorized into natural, semi-synthetic, and microbial polymers.\textsuperscript{[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.\textsuperscript{[14]} Polymers produced by microorganisms are further classified as polyhydroxyalkanoate, pullulan, xanthan gum.\textsuperscript{[13]} Polylactic acid is synthesized from lactic acid that is produced by microbes through lactide formation, then condensation polymerization and ring open polymerization.\textsuperscript{[15]} Several studies indicated there is a great possibility of bio-based materials for developing eco-friendly fish packaging alternatives\textsuperscript{[16,17]} This article aims to review the application of different bio-based polymers embedded with bioactive molecules for fish packaging materials.

\textbf{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.\textsuperscript{[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.\textsuperscript{[19]} Gases barrier, mechanical, thermal, and chemical properties of biomaterials are important criteria to select polysaccharides as packaging materials.\textsuperscript{[20]} The water vapor, gas barrier, structural and mechanical properties of biopolymers can be improved by adding bioactive, nanofillers, cross-linkers, or plasticizers.\textsuperscript{[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.\textsuperscript{[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.\textsuperscript{[10]} Bio-based materials extruded from
Table 1. Polysaccharides-based biopolymers for fish product packaging

| Polysaccharides                     | Bioactive molecules                                      | Applications                                                                 | References |
|-------------------------------------|----------------------------------------------------------|------------------------------------------------------------------------------|------------|
| Rice starch                         | Oregano essential oil (OEO)                              | Fish fillet packed in starch/OEO showed less oxidation and microbial growth  | [19,23]    |
| Cassava starch                      | Citrus lemon peel extract (CLPE)                         | Fish packaged in a film containing 20% CLPE demonstrated low peroxide and Total Volatile Basic Nitrogen (TVB-N) values | [24]       |
| Starch/Pectin                       | Feijoa extracts                                          | • Fish fillet packed in starch/OEO showed less oxidation and microbial growth | [22]       |
| Chitosan nanoparticles/ collagen protein | Anthocyanidin and Cinnamon-perilla essential oil        | Fillets of sea bream fillets wrapped in the composite film extended shelf life to 6–8 days | [26]       |
| Polycaprolactone                    | Chitosan/Rutin (PCL-CS-R)                                | • Reduced microbial population of E. coli, S. aureus, and L. monocytogenes by 18.39%, 19.27%, and 17.45% as compared to polycaprolactone film alone in rainbow trout packaging | [27]       |
| Cellulose nanofiber/Carboxymethyl cellulose | Shikonin                                 | Discoloration of shikonin at weak acid and weak alkali to monitor fish freshness | [28]       |
| Pectin/Sodium alginate/Xanthan gum | Raspberry pomace extracts                                | Changes its color (pH-sensitive) used to monitor protein-rich foods           | [29]       |
| Pectin                              | Oregano essential oil, Ginger essential oil              | • Significant protective effect on protein oxidation, prevention of endogenous enzyme activity | [30]       |
| Chitosan                            | Tarragon essential oil 10% purple tomato anthocyanins    | • Decreased bacterial deterioration of seafood during chilling                | [31]       |
| Chitosan/Methyl cellulose           | Saffron petal anthocyanins                              | Used to indicate the freshness of milk and fish/ monitoring food freshness/spoilage (intelligent film) | [32]       |
| Chitosan/Methyl cellulose           | Anthocyanin                                              | • Antimicrobial against E.coli and S.aureus, antioxidant activity, and monitor lamb meat freshness during storage (smart packaging) | [33]       |
| Bacterial cellulose                 | Pelargonidin dye                                         | Monitors changes in total volatile base nitrogen (TVB-N)                      | [35]       |
| Sodium alginate                     | Purple onion peel extract (POPE)                         | 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% TiO2 Rosemary Essential oil (REO)                     | Exhibited antimicrobial by reducing the psychrotrophic bacteria count 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, TiO2                                                | Good active food packaging films retained high-quality bananas (weight loss, firmness, and surface color) | [38]       |
| K- Carrageenan                      | Red cabbage extract (Brassica oleracea)                  | • Helped to monitor freshness and quality of rainbow trout fillets (Oncorhynchus mykiss) because of color change when quality deteriorates | [39]       |

starch conserves frozen pack fish fillets for 360 days better as compared to low-density 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.\textsuperscript{[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\textsuperscript{[45]} K- Carrageenan, alginate, and agar are widely used to develop coatings and films due to their gel-forming abilities.\textsuperscript{[46]} Oregano essential oil (0.4\%) added into pectin increases the shelf-life of yellow croaker from 20 to 27 days.\textsuperscript{[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 extensively 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.\textsuperscript{[52]} 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.\textsuperscript{[48]} Different proteins and other biopolymers-based fish packaging materials incorporated with different bioactive molecules are presented in Table 2.

| Proteins              | Applications                                                                 | References   |
|-----------------------|------------------------------------------------------------------------------|--------------|
| Gelatin               | Eugenol nanofiber and Polylactic acid                                          | [49]         |
| Gelatin/ Kappa-       | Changes its color based on changes in pH, act as antimicrobial, antioxidant   | [50]         |
| Carrageenan           | and monitors fish freshness during storage                                    |              |
| Fish gelatin          | Fish gelatin containing 1\% of haskap berries extract showed antioxidants and  | [51]         |
|                       | color change as a function of the pH of intelligent packaging to monitor the  |              |
|                       | freshness of shrimp                                                          |              |
| Gelatin/ Oxidized     | Antioxidant activity and have great potential in monitoring freshness of      | [52]         |
| chitin nanocrystals   | shrimp and hairtail (smart packaging), high protein foods                    |              |
| Gelatin/Chitosan      | Retarded lipid oxidation and microbial growth of bluefin tuna                 | [53]         |
| (carboxymethyl       |                                                                               |              |
| chitosan)             |                                                                               |              |
| Gelatin/Chitosan      | Improves antibacterial and UV barrier capability                               | [54]         |
| 0.4\% TiO\textsubscript{2} |                                                                               | [55]         |
| Gelatin/Chitosan      | Effective controlled six foodborne pathogens (E. coli, K. pneumoniae, S.    | [56]         |
| e-Polylysine          | enteritidis, P. aeruginosa, S. aureus, and L monocytogenes)                   |              |
| Gelatin               | Demonstrated superior color retention as compared to curcumin and               | [57]         |
|                       | anthocyanins color change with pH change application for intelligent packaging|              |
| Soy Protein           | The population of S.aureus, B.cereus, S. typhimurium, L.monocytogenes, and E. | [58]         |
| Isolate/Gelatin       | coli was diminished via the inclusion of 20\% Zataria multiflora active      |              |
| Gelatin               | food packaging, extended shelf life                                            |              |
| Diallylde hydroxy     | Films containing a 1:2 ratio of gelatin films (GEL)/DAK- car and thymol-     | [59]         |
| kappa-carrageenan     | loaded zein nanoparticles (ZNP)                                               |              |
| Whey protein isolate  | The film preserves food against oxidation and microbial spoilage improving   | [60]         |
| (WPI)                 | shelf life and quality of foodstuffs                                           |              |
| Gelatin               | Antimicrobial against S.aureus, higher antioxidant activity due to the         |              |
|                       | presence of green tea extract                                                 |              |
Gelatin is one of the first biopolymer materials proposed to carry bioactive compounds. 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. 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. 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. 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. 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. Due to its hydrophobic nature, Zein protein has low water vapor permeability and solubility.

**Polylactic acid and polyhydroxyalkanoates-based biopolymers for fish product packaging**

Polylactic acid (PLA) is synthesized from lactic acid monomers that can be widely used for fish packaging during chilling storage. Polyactic 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.

**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. 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. Bioactive molecules change their color based on changes in pH, temperature, light, time, and ammonia which making which acts as an indicator in monitoring the spoilage of packaged fish products. 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.

Some of the pH-sensitive bioactive molecules (Table 5) are anthocyanins, betalains, carotenoids, carotenoids clowse oil, curcumin, turmeric, rosemary, oregano, thyme, green tea, sage, basil, ginger, coriander, garlic, nutmeg, mace, savory and fennel. The most intensively studied bioactive molecules as colorimetric indicators are anthocyanins, curcumin, and carotenoids. Anthocyanins incorporated into biopolymer films have been used as smart indicators to monitor fish spoilage in Grass carp, rainbow trout, Hair-tail, Spade nose shark, Pangasius, and Atlantic mackerel. 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                               | • Damaged the cell wall membrane leading to change in cell membrane permeability, protein synthesis, and expression of Shewanella putrefaciens fresh-keeping material with antibacterial properties | [68]       |
| Polylactic acid (PLA)           | Nanocellulose/Chitosan                                      | • Polylactic 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                                                         | • 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                                              | • An Indian anchovy (*Stolephorus indicus*) fish packed in PLA/nanocellulose demonstrated increased shelf life as compared to neat PLA and LDPE during chilling storage for 20 days | [71]       |
| Polylactic acid (PLA) / Sawdust (SD) particles | Bacteriocin from Weissella hellenicca BCC 7293 was diffusion coated onto PLA/SD | • Retarded the growth of Gram-positive (*Listeria monocytogenes*, *Staphylococcus aureus*) and Gram-negative bacteria (*Pseudomonas aeruginosa*, *Aeromonas hydrophila*, *Escherichia coli*, and *Salmonella typhimurium*) antimicrobial packaging for pangasius fish fillets | [72]       |
| Poly (butylene adipate co-terephthalate)–PBAT | Oregano (*Origanum vulgare*) essential oil (OEO)            | • Revealed antibacterial activity (reduced growth of coliforms, *Staphylococcus aureus*, and psychrotrophic microorganisms) and prolonged storage quality of fish fillet for 10 days | [73]       |
| Poly lactic acid (PLA)-polyoxybutyrate (PHB) | Cinnamaldehyde (5%)                                        | • 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]       |
| Polylactic acid (PLA)           | Silver montmorillonite (Ag-MMT)                             | • Poly (lactic acid) (PLA)/polyoxybutyrate (PHB) nanocomposite films 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   | • Inhibit protein degradation, microbial growth and lipid oxidation of stored sea bass (*Lateolabrax maculatus*) 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) | *Clitorietatea* (CT) extract                               | • 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]       |
| Nanofillers | Embedded polymers | Characteristics of nanofillers | Mechanisms of action of nanofillers | References |
|------------|-------------------|-------------------------------|-------------------------------------|------------|
| TiO₂/Ag    | Rhinobatoscemiculus gelatin | Antioxidant properties | • Embedding 4% TiO₂-Ag nanoparticles into gelatin film gave good metal chelating and DPPH scavenging activity | [93] |
| TiO₂/ZnO  | 4A-Zeolite Gelatin | Antimicrobial properties | • 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 | • Antimicrobial against Listeria monocytogenes > Escherichia coli > Staphylococcus aureus > Pseudomonas fluorescens though releasing ions and catalysis the formation of active oxygen species | [95] |
| TiO₂      | Polyactic acid Modified with Lycopene | Antimicrobial and antioxidant activity | • 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. | [96] |
| ZnO       | Chiosan/Melissa officinalis essential oil | Antimicrobial and antioxidant activity | • Due to the presence of a positive charge in chitosan polymer, citronellal and geraniol in Melisa essential oils increases the permeability of the cytoplasmic membrane to ATP which leads to cell death (antimicrobial biodegradable composite film for food packaging) | [97] |
| TiO₂      | Gelatin/Grape fruit extracts | Antibacterial against E. coli, L. monocytogenes and antioxidant | • 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/SiO₂  | Chitosan | Antibacterial | • UV-barrier, antimicrobial (Escherichia coli and Listeria monocytogenes) and antioxidant activity | [99] |
| TiO₂      | K-Carrageenan/Xanthan gum/Gillam gum | Antibacterial | • Mold growth of Escherichia coli, IRAQ 3 and Staphylococcus aureus, S33R has been prevented via the inclusion of 5% ZnO-SiO₂ | [100] |
| ZnO       | Polyactic acid | Antibacterial | • Protection from UV light, partial antimicrobial activity against Staphylococcus aureus | [100] |
| ZnO       | Carboxymethyl cellulose/Grape seed extract (5 wt%) | Antibacterial | • Exhibited antibacterial activity against Escherichia coli and Listeria monocytogenes during storage of minced fish paste | [101] |
|           |                   |                               | • Exhibited UV-light and water vapor barrier properties | [102] |
|           |                   |                               | • Zinc oxide nanoparticles increased water and mechanical properties | |
|           |                   |                               | • Antibacterial against L. monocytogenes and E. coli | |
|           |                   |                               | • It preserves high fat meat products | |
|           |                   |                               | • Less water vapor barrier and antioxidant activities | |
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 |
|---------------------|-------------------|---------------------------------|---------------------------------------------|------------|
| *Echium amoenum* anthocyanins | Bacterial cellulose film | • Color change spotted to identify freshness/ spoilage of shrimp for fresh (violet), use soon (gray), and spoiled (yellow) shrimp | • 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 | • Gelatin-chitosan-gallic acid-clove oil prolonged the shelf life of salmon fillet for five days | • Better gas and oxygen barrier properties, inhibition of microbial growth, antioxidant effects of coating | [104] |
| Oxidized chitin nanocrystals Curcumin | Chitosan | • The color of the films was changed due to a change in pH which enabled identify spoilage of the hairtail and shrimp | • 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 | • Enhanced the storage quality of fish/chicken from three days for control to 12 days for amaranthus leaf extract treated film | • Antioxidant, antimicrobial, and intelligent packaging (visible color change from red to yellow on spoilage) | [84] |
| Betacyanin | Glucomannan | • The film had changed its color from purple to yellow when TVB-N reached 39.74 mg/ 100 g | • Smart packaging during fish Osphronemusgouramystorage to monitor freshness | [106] |
| Lyophilized alga *Fucus spiralis* powder | Gelatin | • 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 | • 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) | • New interaction between cellulose and PSPA appeared | • Changed its color from red to blue and is sensitive to ammonia | [85] |
| | • PSPA was uniformly dispersed in CMC and Starch | • Intelligent packaging to monitor fish freshness | |
| | • The addition of PSPA increased tensile strength, decreased elongation at break and moisture content | | |
| Starch Jackfruit seeds anthocyanins | • Higher tensile strength, reduced resistance | • Fish freshness indicator | [108] |
| | | • Intelligent packaging | |
| | | | |
| Starch Roselle anthocyanins | • Anthocyanins were immobilized into starch film, water content and tensile strength decreased, elongation at break increased | • The film was sensitive to ammonia and monitors fish freshness at 4°C | [109] |
| | | • Intelligent packaging | |
| | | | |
| Bacterial nanocellulose Black carrot anthocyanins | • Immobilization of black carrot anthocyanins decreased the mean diameter of nanofibers | • Monitors the freshness of common carp and rainbow trout at 4°C | [110] |
| | | • Consumers realize spoilage by the naked eye due to color changes Intelligent packaging | |
| Zein nanofibers Alizarin | • Alizarin is incorporated in to the zein matrix through hydrogen bonding | • Monitor the freshness of trout through changing its color in response to microbiological and color changes | [111] |
| Chitosan/ Xylan/ Hydroxyapatite/ Curcumin | • Dehydration temperature, protein unfolding and glass transition reduced | • Monitor freshness in Indian oil sardines | |
| | • Immobilization of curcumin into the polymers did not alter the surface of the film | • Smart packaging | [112] |

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

### 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    | • Improved contact angles, mechanical properties, water vapor transmission, lower moisture content, and oxygen transmission rate | • Promising active packaging materials | [113] |
| Chitosan Rice berry phenolics extract     | • Chitosan- rice berry phenolics extract films exhibited increased thermal stability, mechanical resistance, hydrophobicity, barrier, and antioxidant properties | • 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 | • Showed remarkable antibacterial, antioxidant, and air barrier activity | • Zn\textsuperscript{2+} ions damage the cell membrane and interact with intracellular contents of E. coli | [115] |
| Gelatin/Cellulose/Arabic gum TiO\textsubscript{2} Garlic extract | • Enhanced the mechanical, antimicrobial, thermal, water vapor, and oxygen transmission rate | • Bionanocomposite film synergistically controlled the growth of microbes during refrigeration of tilapia fillets | [116] |
Disclosure statement

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

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