

Advanced biodegradable antimicrobial coatings by combining biopolymers and natural antimicrobials for the active packaging and extending the shelf-life of fish and other seafood products: a review

Hariharan S. Melarcode^{1,*}, Onur Ertugrul², Paola C. Alzate Calderon³, Joseph P. Kerry³, and Michael A. Morris¹

¹Advanced Materials and Bioengineering Research (AMBER) and School of Chemistry, Trinity College Dublin, Dublin, D02W085, Ireland

²Department of Metallurgical and Materials Engineering, Izmir Katip Çelebi University, Cigli, Izmir, 35620, Turkey

³Food Packaging and Material Sciences Group, School of Food and Nutritional Sciences, University College Cork, Cork, T12 Y337, Ireland

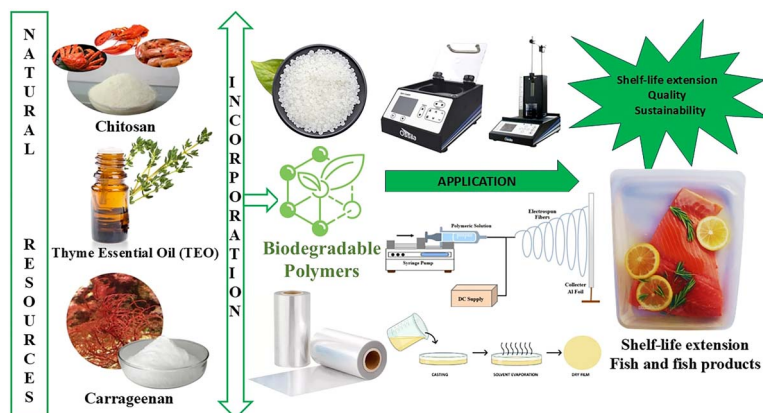
*Corresponding author: Advanced Materials and Bioengineering Research (AMBER) and School of Chemistry, Trinity College Dublin, Dublin 2, D02W085, Ireland.
Email: melarco@tcd.ie

Abstract

Fish and other seafood products are an integral and valuable part of human diets. In advanced nations, the transportation of fish and other seafood products to customers requires packaging and display. Due to their intrinsic physicochemical properties, seafood products are particularly susceptible to microbial growth, resulting in shorter shelf lives and higher spoilage rates compared to other animal-derived proteins, such as meat. To prevent microbial spoilage and preserve the freshness and quality of fish and other seafood products, petroleum-based plastic materials are commonly used in combination with packaging technologies, such as vacuum packaging and modified atmosphere packaging. However, as these fossil fuel-based materials are non-biodegradable, non-recyclable, and associated with damaging environmental footprints, there is a clear need to develop advanced and smart sustainable packaging solutions that can meet environmental and climate demand while being able to extend the shelf-life of seafood products beyond current expectations. Various biopolymers, including polylactic acid, polyvinyl alcohol, chitosan, and polyethylene glycol, have been widely utilised in the development of advanced, sustainable, and biodegradable packaging materials for seafood. However, most of these biopolymers inherently lack antimicrobial properties, limiting their ability to effectively extend the shelf-life of seafood products. Recent studies have shown that incorporating natural antimicrobial materials into biopolymers significantly enhances their antimicrobial properties. As a result, the development of active packaging materials for seafood preservation has emerged as a growing area of research. In this review, we examine approaches that have been used in this area with an emphasis on active antimicrobial-containing systems based on non-fossil fuel plastics. This work will centre on reviewing the literature to understand and analyse the techniques used to create these advanced active biodegradable and sustainable packaging materials.

Keywords: seafood products, sustainable packaging, biopolymers, antimicrobial activity

Graphical abstract



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Introduction: Overview

Seafood is a rich source of nutrients with specific aromas and tastes. Increasing global seafood production reflects a growing consumer demand for freshly packaged seafood products (Laorenza et al., 2022). Fish oils and oily fish are considered to be major food sources that provide the necessary levels of Omega-3 fatty acids (Das et al., 2024). Omega-3 fatty acids (n-3 polyunsaturated fatty acids [n-3 PUFAs], including EPA [eicosapentaenoic acid] and DHA [docosahexaenoic acid]) are dietary fatty acids with a plethora of health benefits (Swanson et al., 2012). Eicosapentaenoic acid and docosahexaenoic acid have been shown to prevent and treat various diseases, including coronary heart disease and rheumatoid arthritis. Omega-3 fatty acids are known to be beneficial to health due to their anti-inflammatory, hypolipidemic, antithrombotic, and antiarrhythmic properties (Lange, 2020). Various research studies have shown that EPA and DHA are necessary for brain development. Neurodevelopmental disorders in humans have been connected to low maternal consumption of Omega-3 fatty acids. EPA and DHA have been suggested to help maintain mental health and prevent brain and heart-related diseases, such as Alzheimer's disease, epilepsy, stroke, schizophrenia, Parkinson's disease, depressive symptoms, and autism spectrum disorders (Madore et al., 2020). However, fish/seafood products are highly perishable and deteriorate faster than meat proteins due to a combination of various microbial and biochemical breakdown mechanisms, as well as their high-water content, mild pH, and high content of non-protein nitrogen compounds (Sullivan et al., 2020). Environmental factors like changing climatic conditions and handling conditions post-mortem may also contribute to the short shelf-life of these seafood products (Gustavsson et al., 2011).

Advanced seafood packaging technology plays a key role in improving the quality and the shelf-life extension of seafood by providing a unique interaction between food, technology, and the environment (Laorenza et al., 2022). Research is focused on the application of active and intelligent packaging, as well as modified atmosphere packaging, with various functions, forms, and materials manufactured using state-of-the-art technology to preserve the seafood quality and extend the shelf-life of seafood products. Active packaging focuses on preventing and reducing seafood deterioration, such as the reduction of microbial growth and chemical oxidation (Jiang et al., 2023). It has been reported that over 35% of all seafood is wasted through the supply chain in Europe due to spoilage (Gustavsson et al., 2011). In recent years, active packaging has become more commercialised. Seafood packaging should be designed not only to preserve product quality but also to extend the shelf-life of seafood products, and active packaging materials can significantly and effectively inhibit microbial growth in seafood products. Several studies have investigated microbial inhibition in seafood products using active packaging (Caba et al., 2019; Mohan et al., 2010; Narayanan et al., 2024; Singh et al., 2016). Microbial activity in seafood products can be controlled via active packaging containing antimicrobial agents that delay post-harvest metabolism and extend the shelf-life of seafood products.

Synthetic fossil fuel-based polymers like polyethylene (PE), polypropylene (PP), polystyrene (PS), and polyvinyl chloride (PVC) are widely used for various food packaging applications because of their low cost, ease of manufacturing, and wide availability (Siracusa et al., 2008). Many studies have been conducted on the use of synthetic fossil-based polymers like low-density polyethylene (LDPE) and PP to extend the shelf-life of various fish and seafood

products (López et al., 2023). As these synthetic fossil-based polymers are non-biodegradable and non-renewable (causing significant environmental challenges), there is a rising demand for developing non-toxic, biodegradable, sustainable, and environmentally friendly packaging materials that extend the shelf-life of fish and fish products (Deeraj et al., 2022). Among all the other available biopolymers, polylactic acid (PLA), polyvinyl alcohol (PVA), and chitosan (CS) have been most widely used and studied for seafood packaging applications because of their biodegradability, non-toxicity, and high mechanical and barrier properties. Polyvinyl alcohol is a biodegradable polymer that is non-toxic, transparent, and, in dry conditions, has high oxygen barrier capacity, making it highly suitable for food packaging. Based on its molecular properties, PVA films can be readily obtained by solvent casting and other thermal-processing techniques (Andrade et al., 2022). Polyvinyl alcohol can also serve as a carrier matrix for the incorporation of many active materials. Smart materials for packaging fish and fish products have been created by incorporating active antimicrobial materials, such as essential oils (EOs) and natural plant extracts, into the PVA matrix (Gowsia et al., 2022). However, the application of PVA as a pure polymer for packaging fish and fish products is restricted due to the high degree of hydroxylation in its molecular chain, which leads to an increase in their hydrophilicity and poor barrier properties, particularly to water (Ghaderi et al., 2020). To obtain materials with good barrier properties, it has been suggested that the lamination of PVA with biodegradable polyesters like PLA will enhance its properties, making it suitable for packaging fish and fish products (Andrade et al., 2022). As PVA has a high affinity towards CS, it has been combined with CS, creating PVA/CS composites that have been successful in extending the shelf-life of fish and fish products (Ghaderi et al., 2020).

Polylactic acid is a hydrophobic polymer formed by fermenting renewable plant resources, followed by the polymerisation of the lactic acid produced. High mechanical strength, non-toxicity, non-irritating nature, biodegradability, and biocompatibility make PLA a better alternative for packaging fish and fish products (Wu et al., 2023). As both PLA and PVA lack inherent antibacterial properties, the incorporation of antimicrobials in the polymer matrix enhances the antibacterial properties of the polymer-antimicrobial composite materials.

This review focuses on the development of novel sustainable packaging systems using state-of-the-art sustainable manufacturing processes that employ novel green biodegradable materials as the structural matrix. Different combinations of polymers and antimicrobials are currently being studied to find the potential combination of polymers and antimicrobials to create active packaging films with higher mechanical, thermal, biocompatible, and barrier properties, along with the slow and sustained release of the antimicrobials to extend the shelf-life of fish and seafood products.

Polymers used for seafood packaging Synthetic petroleum-based polymers

Traditionally, many synthetic petroleum-based polymers like PE, PP, PS, and PVC have been widely used for packaging fish and fish products. These synthetic polymers provide several advantages, including availability from inexpensive raw materials, high tensile and mechanical strength, good oxygen and water vapour barrier properties, and high processability (Siracusa et al., 2008). These polymers are available in high quantities and at low costs with

Table 1. Typical examples of LDPE, polyamide (PA), and polypropylene (PP) films used to extend the shelf-life of fish and fish products.

Product	Packaging material	Storage conditions and shelf-life extension	Reference
Salmon muscle	LDPE with tocopherol	21 days with stabilisation of lipid oxidation at 40%	(Barbosa-Pereira et al., 2013)
Carp fish fillets	polyamide/polyethylene	70%CO ₂ /20%N/10%O ₂ and CO ₂ at 27 days and 50% CO ₂ /50% N ₂ at 24 days. Edible coating maintained for 18 days of storage	(Hosam et al., 2018)
Megalobroma amblycephala	LDPE	Shelf-life extension up to 21 days at 4 °C	(Song et al., 2011)
Rainbow trout	LDPE-nanoclay	Shelf-life extension up to 18 to 20 days at 4 °C with a significant delay in lipid oxidation rate	(Bahmani et al., 2021)
Argentine croaker	LDPE	Fish protein hydrolysates successfully bound to the PE surface due to their known antioxidant activity	(Romani et al., 2020)
Iced Nile tilapia fillets	PE with natural garlic extract	Fish fillets were stored in ice. Shelf-life extension of up to 6 days more than the control	(Jiménez-Ruiz et al., 2023)
Salmon fillets	Polypropylene with 1,018 K6 antimicrobial	Fish fillets were stored at 4 ± 1 °C for 7 days. Higher shelf-life than the control	(Ambrosio et al., 2022)
Shrimps	LDPE-based active film containing rosemary essential oil (REO) and cinnamon essential oil (CEO)	1% w/w REO +1% w/w CEO showed the highest antimicrobial effects	(Dong et al., 2018)
Blended film (REO + CEO) extended the shelf life of packaged shrimps up to 4 days			

very well-defined chemical, physical, and mechanical properties, making them materials of choice. It is the barrier properties of these plastics that increase shelf-life by reducing infiltration of contamination or allow formation of vacuum seals or seals that allow formation and retention of CO₂ and other gas atmospheres that limit bacterial growth (Daniloski et al., 2019). Research studies carried out on the effectiveness of synthetic petroleum and fossil fuel-based polymers like PP, PA, and LDPE in extending the shelf-life of fish and seafood products are compiled in Table 1.

Low-density polyethylene is an ideal base material for these applications due to its low glass transition temperature. Most frozen products become hard with sharp edges under freezing conditions due to their transition into an amorphous glassy state. This can cause penetration through the films and loss of package integrity. Glass transition temperature (T_g) plays a key role in the choice of packaging material properties. Polymers with a higher T_g than freezing also become brittle at the storage temperatures used for frozen products. Low-density polyethylene has a low glass transition temperature of around -100 °C, ensuring it remains flexible at typical freezer temperatures, making it a suitable packaging material for products stored at low temperatures (Laorenza et al., 2022).

The studies described in Table 1 have demonstrated that LDPE, PA, and PP can be used efficiently to extend the shelf-life of a wide variety of fish. However, these can be challenging to recycle following use and cause serious environmental and disposal challenges, including climate impacts, microplastic generation, and litter production (Moshood et al., 2022). To overcome these environmental issues, scientists and researchers have started to develop biodegradable polymers with the inclusion of antimicrobials, such as silver, zinc oxide, and copper oxide, to extend the shelf-life of chicken, fish, and other meat products (Garcia et al., 2018). These advanced biodegradable polymers incorporated with the nanomaterials-based inorganic antimicrobials like

silver and zinc oxide were able to deposit the antimicrobials on the surface of the fish and other meat products in a slow and sustained manner without deteriorating the quality and the taste of the fish and other meat products, and also causing no damage to the environment when they are disposed of after their use. Azlin-Hasim et al. spray-coated nanosilver onto commercial LDPE substrate using Pluronic surfactant and PS-PE oxide block copolymers to produce the modified materials. Their study found that when compared to the control films, the silver-coated LDPE films extended ($p < .05$) the shelf-life of chicken breast fillets and enhanced ($p < .05$) the oxidative stability of the chicken breast fillets packaged using the silver-coated LDPE films (Azlin-Hasim et al., 2018). However, these inorganic antibacterials have human toxicity issues, and their use as food contact materials has been limited by regulation (Onyeaka et al., 2022). To overcome these issues, researchers are exploring the use and the advantages of using naturally derived antimicrobials and biopolymers to extend the shelf-life of fish and seafood products (Valizadeh et al., 2020). These systems are explored further below.

Biopolymers

Bio-based polymers or biopolymers can be described as polymers obtained from natural and renewable resources rather than fossil fuels. They should be differentiated from the term biodegradable, as not all biopolymers are biodegradable. Some of the biopolymers are naturally available, some are obtained via materials sourced from nature and synthesised in a laboratory, while others are synthesised using micro-organisms.

Polysaccharides, such as CS, starch, and carrageenan, microbial-based biopolymers like polyhydroxyalkanoates (PHAs), polyhydroxybutanoates (PHBs), pullulan, and xanthan gum, protein-based biopolymers like alginate, gelatin, soy protein, whey protein, and collagen, chemically synthesised biopolymers like PLA and PVA, as shown in Figure 1. They are widely used for packaging

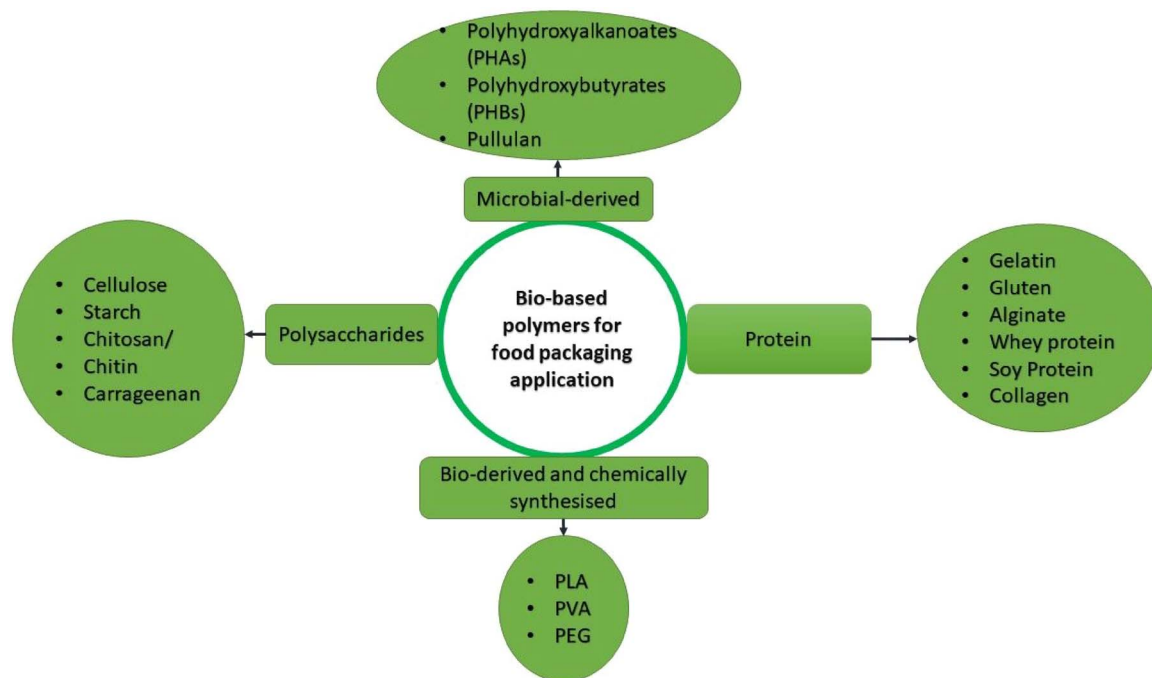


Figure 1. Major types of bio-based and biodegradable polymers, including microbial-derived, polysaccharides, bio-derived and chemically synthesised, and protein-based, used for packaging seafood products.

fish and fish products because of their biodegradability, non-toxicity, low carbon footprint, ability to form three-dimensional networks, and low processing and production costs (Andrade et al., 2022). Despite the range of biopolymers now available, many of these biopolymers are made in too low a volume and at too high a cost for use in the low-cost food packaging market, and the packaging sector is focused on a limited number of these biopolymers (de Jong et al., 2022). For example, the polymer polyethylene furan-2,5-dicarboxylate has superior barrier properties to polyethylene terephthalate (PET), but it is currently too expensive for widespread commercial use (de Jong et al., 2022).

Bio-derived and chemically synthesised bio-based or biodegradable polymers

Polyvinyl alcohol obtained from sugar cane and other crops has been widely used in packaging food products due to its low cost, high polarity, and biodegradability (Oun et al., 2022). Polyvinyl alcohol gained more importance as a packaging material because it offers superior mechanical strength, chemical resistance, biocompatibility, and high film-forming capacity compared to other conventional synthetic polymers. However, due to its high water solubility and dispersibility, this material cannot be extensively used on its own for packaging fish and other seafood products (Oun et al., 2022). Polylactic acid is more widely used for food packaging (Hussain et al., 2024). Unlike PVA, PLA is a hydrophobic polymer that has better mechanical strength than PVA, is non-toxic, biodegradable, and biocompatible, and has superior thermal processability than PVA, along with low cost (Hussain et al., 2024). However, PLA is often combined with other biopolymers and antimicrobials to be effectively used for seafood packaging and shelf-life extension of seafood products (Hussain et al., 2024). Polyethylene glycol (PEG) is another biodegradable polymer used for seafood packaging applications (Jafarpour et al., 2023). Below, we review some of the more commonly used biopolymers.

Biodegradable PVA (Bio-PVA) for seafood shelf-life extension

Biodegradable PVA (Bio-PVA) is an abundantly available, bio-compatible polymer obtained from various biological sources through hydrolysis (Rahman Khan et al., 2024). Unlike fossil fuel general PVA, bio-PVA may contain modifications such as bio-based monomers, copolymers, or enzyme-sensitive linkages that enhance degradation (Rahman Khan et al., 2024). Figure 2A shows the chemical structure of PVA. Polyvinyl alcohol has been classified as Generally Recognised As Safe (GRAS) by the Food and Drug Administration (FDA) and the EU regulations for food contact applications (Hussain et al., 2024). Bio-PVA has been extensively used for various applications, including coating, medical devices, and as a potential packaging material (Rahman Khan et al., 2024). When compared to other available polymers, PVA has higher mechanical strength, chemical resistance, gas-barrier, and film-forming properties, which offer advantages when used for packaging fish and fish products. As shown in Figure 2B, the properties, like the antimicrobial activity, UV-barrier property, colorimetric property, moisture/gas barrier, mechanical strength, and biodegradability, of the composite films can be enhanced by combining PVA with other polymers to form non-soluble, water-resistant products for packaging, and shelf-life extension of fish and other seafood products (Hussain et al., 2024; Oun et al., 2022). Polyvinyl alcohol films can be prepared using several techniques like solution casting, spin coating, electrospinning, and common moulding techniques (Rahman Khan et al., 2024).

As PVA is hydrophilic, containing hydroxyl groups in its chemical backbone, it is highly compatible and easily mixed with other hydrophilic biopolymers like CS through hydrogen bonding (Dudek et al., 2020).

To enhance the application of PVA as a potential food packaging material, many chemical cross-linkers are added to improve its properties (particularly mechanical strength) (Suganthi et al., 2020). These include malic acid, boric acid, glyoxal,

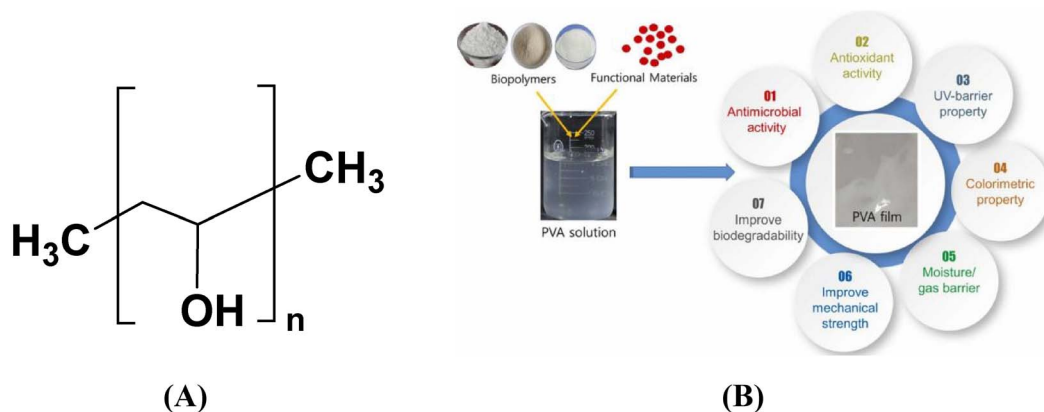


Figure 2. (A) The chemical structure of PVA and (B) improved properties of PVA films after blending with various biopolymers and reinforcement with functional materials (Oun et al., 2022).

glutaraldehyde, formaldehyde, methacrylate, and citric acid, which have been extensively studied in enhancing the shelf-life of fish and other seafood products (Suganthi et al., 2020). Mustafa et al. (2020) studied the effect of glutaraldehyde as a cross-linker to improve the properties of PVA film for food packaging applications (Mustafa et al., 2020). The authors observed that glutaraldehyde reacted in intermolecular spaces and increased the cross-link density of the matrix (Mustafa et al., 2020). The study proved that because PVA has hydroxyl groups in its molecular chain, it was able to form strong hydrogen bonds. This led to an increase in the intermolecular bonding, thereby enhancing the water vapour transmission rate (WVTR) of the resulting composite matrix compared to a neat PVA matrix (Mustafa et al., 2020). The composite also had enhanced antibacterial activity (Mustafa et al., 2020). Singha et al. combined PVA with corn starch, which was reinforced at 15 wt.% with *Grewia optiva* fibre grafted with methyl methacrylate (Singha et al., 2015). This combination enhanced the adhesion of PVA, improving its mechanical and thermal stability (Singha et al., 2015). Furthermore, this composite showed enhanced biodegradability by losing 29% of its weight after being decomposed in the soil for 120 days (Singha et al., 2015). When fillers like glass flakes were added to the PVA matrix, a significant reduction in the film's WVTR was observed when compared to the neat PVA matrix (Channa et al., 2022). These fillers also enhanced PVA's biodegradability and demonstrated greater efficacy in food preservation when compared to commercial LDPE bags (Channa et al., 2022).

PLA biopolymers for seafood shelf-life extension

Among all the biopolymers used for food packaging, PLA, PHAs, pullulan, and xanthan gum have been most widely used for seafood packaging (Abelti & Tekka, 2022). Polylactic acid is synthesised from the fermentation of renewable plant resources and polymerisation of lactic acid (Channa et al., 2022). Polylactic acid is chemically represented as shown in Figure 3. Polylactic acid has a high mechanical modulus, which makes it better than other biopolymers that are used as a better alternative to other petroleum-based polymers used in food packaging applications (Channa et al., 2022). Polylactic acid has higher mechanical strength and better appearance when compared to other polymers, such as polyurethane, PS, and PP (Channa et al., 2022). Along with having a high strength modulus, PLA is also non-toxic and non-irritating (Channa et al., 2022). Polylactic acid is classified as GRAS by the FDA and the EU regulations for contact with food applications (Abelti & Tekka, 2022). When compared to other

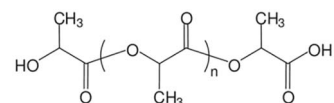


Figure 3. Chemical structure of PLA.

degradable biopolymers, e.g., PEG, PHA, and poly(ϵ -caprolactone) (PCL), PLA has superior thermal processability and is a hydrophobic polymer (Abelti & Tekka, 2022). It can be processed via blow moulding, thermoforming, electrospinning, film extrusion, injection moulding, and film forming techniques (da Silva Pens et al., 2024). However, owing to its brittle nature and poor barrier properties, the application of PLA in the seafood packaging industry has been limited (da Silva Pens et al., 2024). Thus, researchers are focusing on the further development of PLA to overcome these limitations, which can be achieved by incorporating bioactives, such as EOs (Qin et al., 2017). The incorporation of these bioactives, such as EOs, into PLA helps not only to increase the biodegradability of PLA and improve its plasticity (reduced brittleness) but also aids in increasing the antimicrobial activity and the barrier properties of PLA (da Silva Pens et al., 2024; Qin et al., 2017).

As PLA is brittle, has high glass transition temperature ($T_g \sim 55^\circ\text{C}$), high oxygen and water vapour permeability, and lacks inherent antimicrobial property, the combination of PLA with other compatible biopolymers, NAMs, and plasticisers like PEG have been known to be one of the most efficient and cost-effective methods to enhance the properties of PLA-based composites for packaging fish and fish products (Ismaili et al., 2024). Copolymerisation of PLA with a nanofiller or a plasticiser has been shown to be an economic and effective way to overcome these limitations of the PLA matrix. Active PLA-based composite films have also been obtained by combining PLA with different antimicrobials like propolis ethanolic extract (PEE) and *Tanacetum Balsamita* essential (TBE) oil along with cellulose nanocrystals (CNC) to extend the shelf-life of vacuum-packed cooked sausages (Figure 4). Addition of CNC enhanced the mechanical property of the PLA composite films, while TBE enhanced the antibacterial property of the composite films (Khodayari et al., 2019). Monika et al. (2018) synthesised a ternary bionanocomposite of PLA/polybutylene succinate (PBS)/1D Functionalised CS (FCH) using melt extrusion (Monika et al., 2018). The study showed that the static contact angle (CA) had significantly increased when compared to PLA, good dispersion of FCH in polymer domain helped to establish a proper balance in the mechanical properties with improved crystallisation efficiencies and the tight networks formed by the

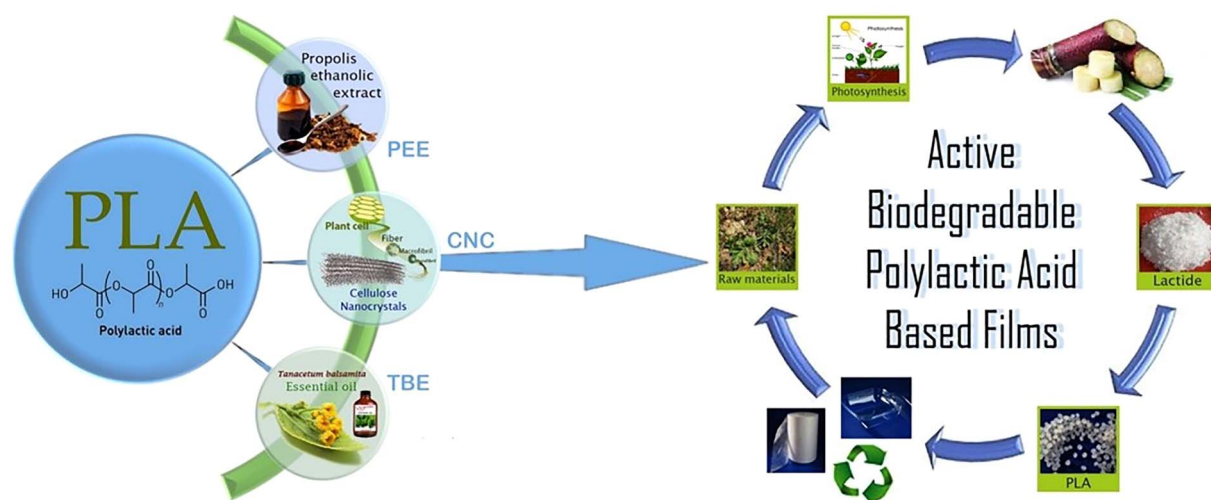


Figure 4. Biodegradable active PLA-based composite films developed by incorporating PEE, TBE, and CNC for enhanced functional performance (Khodayari et al., 2019).

ternary bionanocomposite helped to block the UV-C, which makes this bionanocomposite to be used as a potential fish packaging material (Monika et al., 2018).

Poly(lactic acid) was combined with different fossil and non-fossil-based polymers that act as bioplasticisers, such as poly(methyl methacrylate) (PMMA), PCL, and PET to enhance its thermal and mechanical properties (Gonzalez-Garzon et al., 2018). Many researchers have studied PLA/PMMA blends for various biological applications, including food packaging. Studies by Gonzalez-Garzon et al., (2018) showed that the miscible PLA/PMMA system has a synergistic effect on the polymer blend and can enhance the glass transition temperature (T_g) and the tensile strength of the PLA (Gonzalez-Garzon et al., 2018). In another interesting study by Mazuki et al. (2023), a significant decrease in the glass transition temperature (T_g) value was observed upon addition of PLA into PMMA, indicating increased flexibility of the polymer backbone chain. Fourier-transform infrared analysis of the composite showed that strong dipole-dipole interactions were observed between PLA and PMMA matrices (Mazuki et al., 2023). However, the miscibility of the PLA and PMMA matrix was highly dependent on the crystalline nature of PLA, the technique used for the synthesis of the PLA-PMMA blend and the molecular interaction force between the PLA and PMMA matrix. In an interesting study, Zhang et al. (2003) found that the PLA and PMMA had similar poor miscibility because PLA was crystalline, and as a result, only amorphous D-lactide-containing PLA and PMMA were partially miscible (Zhang et al., 2003).

Poly(lactic acid) and PET are polymers with similar barrier properties and are often compared with each other for their application as a food packaging material. Even though PET has been widely used for packaging food materials and for creating portable and durable products to store food products, the increase in the cost of production and the purchasing cost of PET has led to the development and use of PLA as a potential alternative to PET (Ismaili et al., 2024; Qin et al., 2017). La Mantia et al. (2012) studied the influence of small amounts of PLA on the recycling properties of post-consumer use PET bottles, where only up to 5% can be included without affecting the PET recycling, largely related to the clarity and transparency of recycled products (La Mantia et al., 2012). The rheological studies showed that the rheological properties and the behaviour of PET during spin coating and blow moulding worsened after the addition of even small amounts of

PLA (La Mantia et al., 2012). The overall behaviour of the PLA-PET blends depended on the quantity of PLA present in the blends, the immiscibility between PET and PLA matrices and the degradation of PLA, which has a melting point (~ 170 to 180 °C) lower than the processing temperature of PET (~ 260 °C) (La Mantia et al., 2012). You et al. (2018) overcame this issue of immiscibility of PLA/PET blends by using 0.7 phr (parts per hundred resin) of SA-GMA, which acted simultaneously as a chain extender and compatibiliser (You et al., 2018). This low quantity addition of SA-GMA ensured that the bio-based carbon content was maintained. The authors concluded that combining SA-GMA with PLA/PET blends was an alternative way to replace some of the PET and thus lower the carbon footprint of fossil-based PET (You et al., 2018).

Synthetic biodegradable PEG polymer for seafood shelf-life extension

Phase-change materials, or PCMs, are materials that can store a considerable amount of heat by changing their phase (Yang et al., 2022). Many PCMs are generally used to store latent heat energy (Yang et al., 2022). Phase-change materials have gained a lot of interest because of their high energy density, low cost, chemical stability, non-toxicity, non-corrosiveness, and low temperature variations throughout the heat storage process (Farid et al., 2021). Phase-change materials that are most frequently used include hydrated salts, paraffin waxes, eutectics, and organic compounds, such as polymers, like PEG and fatty acids. Due to a high degree of crystallinity, PEG is often used as a PCM (Farid et al., 2021). Jafarpour et al. (2023) encapsulated PEG400, as a PCM, in alginate using calcium chloride (CaCl_2) (Figure 5). The alginate was able to effectively encapsulate PEG, and the encapsulated PEG, which was used as a PCM, helped to preserve pH, total volatile basic nitrogen (TVB-N), and thiobarbituric acid (TBA) value, thereby helping to increase the shelf-life of packaged fish (Jafarpour et al., 2023).

Poly(ethylene glycol) is often described as a biopolymer, but it is, in fact, synthesised from fossil-fuel-derived ethylene oxide polymerisation. Poly(ethylene glycol)-based polymers are flexible, water-soluble, non-toxic, biocompatible, and biodegradable, making PEG an interesting and useful material for various biological applications, including drug delivery, wound healing, and food packaging. Poly(ethylene glycol) is chemically represented as:

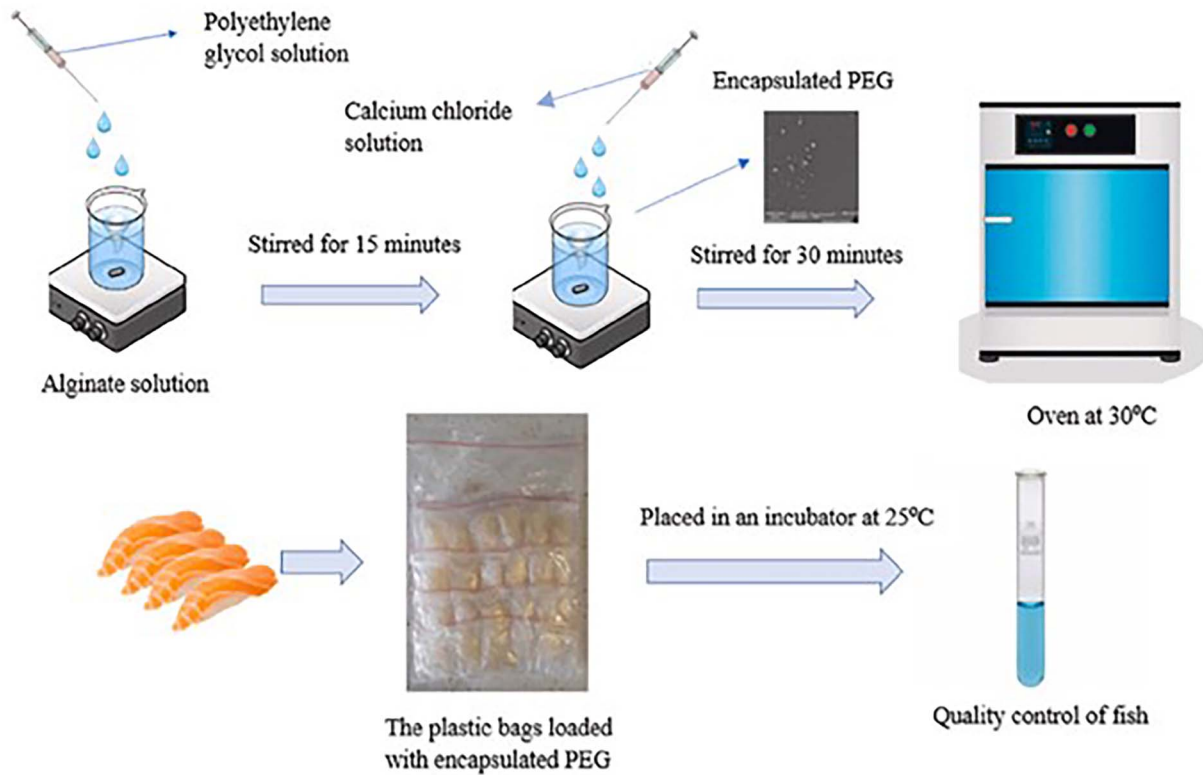
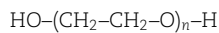


Figure 5. Encapsulation of PEG as a phase change material using alginate microbeads to package fish (Jafarpour et al., 2023).



where n is the number of oxyethylene groups. They are non-irritating and have no negative impact on human health (Birca et al., 2019). Thus, they are widely used as additives and plasticisers, thereby having a wide variety of industrial applications, including cosmetics, drug delivery, and as food-coating materials to prevent microbial attachment and biofilm formation on the contact surfaces of food products (Shi et al., 2021). The application of PEG mainly depends on its molecular weight (Shi et al., 2021). Among all PEGs, PEG400 is commonly used as a plasticiser to enhance the flexibility of the polymers, thereby increasing the mobility of the polymer chains by helping overcome the brittleness of the polymer and enhancing their applicability as food packaging materials (Sajjan et al., 2020). An interesting study was carried out by Sajjan et al. (2020), where they prepared PVA-gelatin/PEG400 and used formaldehyde to increase the barrier properties. The authors found that these films had low water solubility, low water transmission rate, and high water retention capability. The incorporation of PEG-400 enhanced the amorphous nature of the film (Sajjan et al., 2020). In other interesting research by Byun et al. (2010), where they prepared PLA films with α -tocopherol, butylated hydroxytoluene (BHT), and PEG using a film casting extruder, they found that the addition of PEG caused a decrease in T_g of the films (Byun et al., 2010). Apart from this observation, the authors also found that the addition of plasticisers, PEG400 and BHT, increased the water vapour permeability and decreased the oxygen permeability of the films (Byun et al., 2010). Through this study, the authors were able to show that BHT and PEG400 were added not only to prevent the degradation of the PLA films during extrusion but also to increase the flexibility of the PLA films (Byun et al., 2010).

Along with PVA and PLA, PEG400 has also been combined with natural biodegradable polymers like CS to enhance the flexibility and the barrier property while being used as a seafood packaging material (Ismaili et al., 2024; Sun et al., 2020). An interesting study by Sun et al. (2020) showed that to enhance the flexibility, mobility, and applicability of the CS polymer chains (by forming new hydrogen bonds and free volume between the chains), plasticisers with good compatibility and water solubility, like glycerol, sorbitol, and PEG400, can be added to the CS polymer (Sun et al., 2020). Among these plasticisers, PEG400 promoted better barrier properties. A decrease in crystallinity and an increase in permeability were observed with an increase in PEG400 concentrations (Sun et al., 2020). Zidan et al. (2023) synthesised biodegradable films of CS/PEG using a simple casting method on a glass petri dish, as shown in Figure 6. Here, the CS and PEG solutions were prepared separately and then mixed on a hot plate magnetic stirrer. Once the solutions were fully dissolved, the mixture was poured into a glass petri dish and allowed to dry. Upon drying, the films were peeled off, labelled, and stored in a glass desiccator (Zidan et al., 2023). In another interesting study, Fathima et al. (2018) used PLA/CS films for packaging Indian white prawn (Fathima et al., 2018). Here, PVA was used as a plasticiser, and PEG was used as a crosslinking agent. PEG400 reacted with hydrophobic PLA, while PVA reacted with hydrophilic CS (Fathima et al., 2018). PEG400 lowered the tensile strength of the film, thus making the films more flexible and also enhanced the heat-sealing capacity of the films (Fathima et al., 2018). The study concluded that the microbial and biochemical quantity indices of prawn packed in these films were retained, and thus the PLA/nano CS films, where PEG was used as a crosslinking agent, can be used to extend the shelf-life of fresh prawn (Fathima et al., 2018). The study by Yang et al. (2018) also showed that PEG/EG PCM had excellent

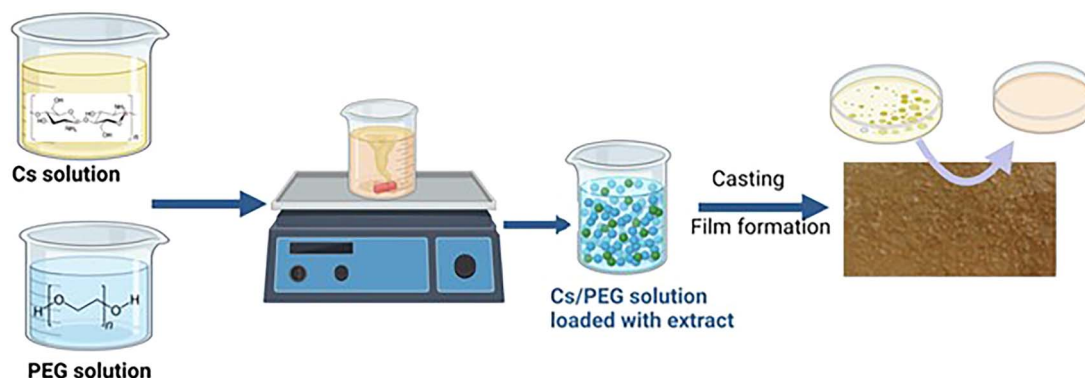


Figure 6. Preparation of CS/PEG films using the solvent or solution casting process (Zidan et al., 2023).

heat-sealing properties and excellent heat storage capacity (Yang et al., 2018).

Ge et al. (2022) blended zein and PLA to create an active biodegradable packaging material. PEG was used as a plasticiser and a compatibiliser (Ge et al., 2022). Four kinds of EOs, namely eugenol, cinnamaldehyde, carvacrol, and tea tree EO, were added as antimicrobial and antioxidant agents (Ge et al., 2022). The authors found that PEG not only enhanced the flexibility and workability of the films by reducing the intermolecular binding and the cohesion between the polymer chains, which led to a decrease in the tensile strength of the films, but also promoted the interfacial adhesion between PLA and zein by acting as a compatibiliser (Ge et al., 2022). In another interesting study by Mendes et al. (2023), nanofiber mats of PLA/PEG blends incorporated with Peppermint EO were prepared using the solution-blow-spinning (SBS) technique (Mendes et al., 2023). In this study, the authors showed that the peppermint EO can be released in a slow and sustained manner, thereby increasing the shelf-life of strawberries stored at 25 °C (Mendes et al., 2023).

All these research studies have shown that PEG has the potential to be used as a plasticiser to increase the flexibility or mobility of the films and as a compatibiliser to enhance the adhesion between two or more polymer chains (Ge et al., 2022; Mendes et al., 2023). They can also help in the slow and sustained release of the antimicrobials from the polymer films, thereby increasing the shelf-life of food products (Ge et al., 2022; Mendes et al., 2023).

Polysaccharide-based biopolymers (CS, carrageenan)

Chitosan is a natural biopolymer with high antimicrobial properties obtained from the deacetylation of chitin (Hejazi & Amiji, 2003). It is an abundant polymer found on the exoskeleton of shellfish, insects, crustaceans, and fungi (Hejazi & Amiji, 2003). The positively charged amino groups at the C-2 position of the glucose monomers enhance the antimicrobial property of CS (Martins et al., 2014). The free amino group in the glucosamine chain, as seen from the chemical structure of CS shown in Figure 7, can be easily protonated (Meshalkin et al., 2022). The protonated or cationic CS interacts with the negatively charged phospholipids, lipopolysaccharides, peptides, and amino groups present on the surface of the microbial membranes (Meshalkin et al., 2022). This is why CS effectively kills gram-negative bacteria more than gram-positive bacteria (Martins et al., 2014). Another probable mechanism by which CS is expected to impede bacterial growth is through its interaction with the genetic material (DNA or RNA) of the microbial cells (Ke et al., 2021). In an interesting study carried out on *Candida albicans* using CS, Ke et al. observed that the mitochondrial biogenesis of *C. albicans* can be inhibited by

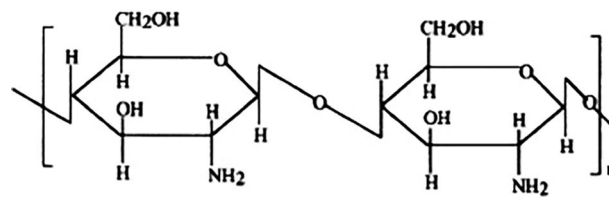


Figure 7. Chemical structure of CS (Meshalkin et al., 2022).

CS, suggesting that the antifungal and antimicrobial property of CS is effectively carried out by the suppression of mitochondrial function and the subsequent ATP generation, leading to the death of the micro-organism (Ke et al., 2021). Due to these unique properties and the ability to bind with water, absorb nutrients used by the bacteria for their growth and development, interacting with the negatively charged molecules on bacteria's membrane, and its superior film-forming abilities, CS has been potentially favoured for packaging food products (Maghsoudi et al., 2009). However, the characteristic behaviour of CS in solution is variable as it relies upon its degree of deacetylation, chain length, and the position of the acetyl groups along the chain (Narasagoudr et al., 2020). Many attempts have been made to identify and understand the exact antimicrobial mechanism of CS, but only approximate findings have been reported so far (Rabea et al., 2003).

Chitosan films can be obtained using techniques, such as solvent casting and extrusion. Chitosan films have demonstrated high water vapour, UV, and oxygen barrier properties, along with greater mechanical strength, making them suitable biopolymers for use in food packaging applications (Foster & Butt, 2011). Although CS has shown promising antimicrobial properties against medically important bacteria and other microbes, an interesting study by Foster and Butt suggests that films formed using CS solutions are not antimicrobial (Foster & Butt, 2011). The CS solution-based films appear not to inhibit the growth of common bacteria, such as *Staphylococcus aureus*, *Escherichia coli*, or *Staphylococcus epidermidis* (Foster & Butt, 2011). The insolubility of CS, which is akin to chitin, in organic solvents and water complicates the synthesis of film-forming solutions. Acidic solutions with a pH below 6.2 are employed to help overcome this issue (Leffler & Müller, 2000). Cruz-Romero et al. (2013) demonstrated in their intriguing study that acetic acid, used to dissolve CS, exhibited strong antibacterial effects. The synergistic effect between acetic acid and CS facilitated the dissolution of CS in a 1% acetic acid solution (Cruz-Romero et al., 2013). The results obtained from their study also suggested that the determination of the antimicrobial activity of CS may be influenced to a certain

extent by the molecular weight of the CS used (Cruz-Romero et al., 2013). To enhance the antimicrobial, mechanical, and optical properties of the CS films and to make them more effective for use as food packaging materials, other biopolymers and NAMs, such as EOs, plant extracts, and nanomaterials, were incorporated into the CS films to create stronger films with advanced properties for efficient food packaging applications.

Chitosan is compatible with many hydrophilic biopolymers like PVA and can interact with other NAMs and nanomaterials to form composites with improved properties to extend the shelf-life of fish and other seafood products (Sébastien et al., 2006). Sébastien et al. (2006) prepared a CS-PLA film by blending CS with PLA dissolved in chloroform using PEG400 as a plasticiser and casting them onto a glass plate. Although the film showed antifungal and antibacterial properties, the authors found that the compatibility of CS and PLA was very low, indicating that a compatibiliser must be used to increase these polymers' interfacial adhesion (Sébastien et al., 2006; Suyatma et al., 2010). In the study on the preparation of CS/PLA/PVA films using solution mixing and film casting techniques by Grande and Carvalho, they found that the binary blends of PVA/PLA and CS/PLA were incompatible, while the binary blend of CS/PVA was compatible because of high synergy between CS and PVA (Grande & Carvalho, 2011). Many research studies have been reported on the successful blending of functionalised-CS with PVA for packaging fish and fish products (Lei et al., 2021; Narasagoudr et al., 2020). These studies have shown that the incorporation of functionalised-CS in the PVA matrix has led to the improvement of tensile strength, UV-transmittance and the elongation at break of the PVA polymer (Lei et al., 2021; Narasagoudr et al., 2020). Lei et al. and coworkers were the first to report a new method to prepare catechol-functionalised CS (C-CS)/PVA composite films using a solution blending technique in a neutral aqueous solution for food packaging (Lei et al., 2021). The authors found that the incorporation of C-CS into the PVA matrix led to composite films that had tensile strength and elongation at break of 45.2 MPa and 153%, respectively, which were 26.3% and 46.3% greater than pure PVA films (Lei et al., 2021). The authors also found that the water resistance, UV barrier, and the antibacterial properties of the final composite films had improved significantly (Lei et al., 2021). Narasagoudr et al. incorporated ethyl vanillin, an ethanol-soluble synthetic food additive, in CS/PVA blend films of various ratios synthesised using the solvent casting technique (Narasagoudr et al., 2020). The authors found that the incorporation of EV in the CS/PVA blend matrix had a strong impact on the WVTR, oxygen and UV-barrier properties of the film (Narasagoudr et al., 2020). The authors also found that there was an increase in the hydrophobicity of ethyl vanillin incorporated CS/PVA (CPEV) blend films as the concentration of CS was increased (Narasagoudr et al., 2020).

Apart from combining CS with biopolymers like PLA and PVA, another biopolymer with which CS has been combined and widely studied is gelatin (López-Caballero et al., 2005). Gelatin is also obtained through natural sources, mainly from fish skins and bones (López-Caballero et al., 2005). Both gelatin and CS are hydrophilic biopolymers with high affinity and good compatibility with each other (López-Caballero et al., 2005). A decrease in the TVB-N and the microbial cell count of specifically gram-negative bacteria proved that the liquid form of the CS/gelatin coatings prevented the spoilage of cod patties (López-Caballero et al., 2005). Feng et al. (2016) synthesised a coating by combining fish gelatin and CS to prevent the myofibril degradation of golden pomfret fillets during cold storage (Feng et al., 2016). The authors found that during the storage of golden pomfret at 4 °C for 17 days,

its deterioration was significantly prevented by both the coatings made up of CS only and gelatin/CS combined (Feng et al., 2016). Overall, the authors found that among all the other coating combinations of CS and gelatin, during cold storage, the best effect of preserving the quality of fish fillet was demonstrated by the edible coating made of 7.2% gelatin and 0.4% CS (Feng et al., 2016). However, in a study conducted by Gómez-Estaca et al. (2009) to evaluate the effectiveness of edible films made of 8% gelatin and 8% gelatin/CS combined with 7.5% clove EO (CEO) on salmon fillets, it was found that the film made of gelatin alone was highly effective on both *Listeria innocua* and *E. coli* bacteria than the film composed of gelatin/CS mixture (Gómez-Estaca et al., 2009). The solubility of the resulting CS/gelatin film was reduced significantly due to the formation of ionic and hydrogen bonds between the films (Gómez-Estaca et al., 2009). This observation contradicted their previous studies, where the authors observed that the low water solubility of the gelatin/CS matrix provided the required stability when it was used in contact with the fish during cold storage (Gómez-Estaca et al., 2009).

While CS is the predominant polysaccharide-based biopolymer employed for prolonging the shelf-life of fish and fish products, carrageenan has also been utilised for this purpose, albeit less extensively and effectively than CS. Carrageenan is regarded as a renewable and sustainable biopolymer, characterised by very simple and efficient extraction processes (Mathew et al., 2024). Carrageenan is a polysaccharide obtained from red seaweed *Rhodophyceae*, renowned for its superior gelling and film-forming capabilities (Mehnath et al., 2021). This polysaccharide is linear, consisting of repeated units of α -D-galactopyranose (residue A) and β -D-galactopyranose (residue B) linked by 1,3 and 1,4 links (Mehnath et al., 2021). Carrageenan also includes sulphate hemi ester groups, methoxy groups, and pyruvate groups in addition to galactopyranose (Mathew et al., 2024; Mehnath et al., 2021). Carrageenan is divided into two groups according to its gelling (kappa and iota) and thickening (lambda) properties (Lochhead, 2017). *Eucheuma cottonii*, *Eucheuma spinosum*, *Chondrus crispus*, and *Gigartina* sp. are among the most prevalent carrageenan-rich seaweed species (Cheng et al., 2022). Carrageenan is a popular ingredient in medicines, cosmetics, and the food industries due to its gelling, thickening, and stabilising properties (Cheng et al., 2022). Carrageenan-based composites, for example, can be created by including functional materials, such as polyphenols, pigments, and enzymes due to their high hydroxyl content (Khan et al., 2023). Furthermore, the sulphate group in carrageenan contributes significantly to the strength of the gel generated by carrageenan (Cheng et al., 2022; Khan et al., 2023).

Carrageenan-based materials are gaining popularity in the food packaging industry due to their capacity to improve food quality and safety (Aga et al., 2021). One potential application for carrageenan-based materials is the development of active packaging, which can help extend the food product's shelf-life (Maroufi et al., 2021). Carrageenan-based systems have been investigated as possible delivery systems for active chemicals, such as antimicrobials or antioxidants, that can be combined with packaging materials to enhance the quality and safety of food (Mathew et al., 2024). Originally created as biodegradable films, carrageenan-based films have demonstrated good oxygen barrier properties that can help prevent oxidation, thereby extending the shelf-life of various food products (Koca & Bayramoğlu, 2022). They are combined with clay, EOs, and other seaweed-based polysaccharides like glucomannan and agar to enhance their properties and extend their application for packaging various food products (Qiao et al., 2023).

Microbial-based biopolymers (PHAs, PHBs)

Polyhydroxyalkanoate is a natural polyester produced via bacterial fermentation of sugar, glucose, or vegetable oil feedstock (Zytner et al., 2023). As their name indicates, polyhydroxyalkanoates, or PHAs, are hetero- or homopolymers of esters made up of several hydroxyalkanoates (HAs). A variety of bacterial and algal genera construct PHAs (Zytner et al., 2023). Polyhydroxyalkanoate is a thermoplastic polymer that is nontoxic, optically active and biocompatible. Being easily manufactured from renewable resources, their production is considered environmentally sustainable (Zytner et al., 2023). Polyhydroxyalkanoate has an isotactic structure with piezoelectric properties (Gupta et al., 2020). Polyhydroxyalkanoate is one of the most current and commonly utilised biodegradable polymers for food packaging applications in different industries (Gupta et al., 2020; Shaikh et al., 2021).

Several bacteria can accumulate PHAs as intracellular reserve materials. Some organisms accumulate PHA from 30% to 80% of their cellular dry weight (Gupta et al., 2020). Its T_m ranges from 40 to 180 °C depending on the monomers used for synthesis (Gupta et al., 2020). Depending on the carbon source and the microbial strain, PHAs can be biosynthesised with mechanical properties ranging from rigid and brittle to rubber-like (Shaikh et al., 2021). A good barrier property film is formed when PHA is blended with zein (Shaikh et al., 2021). An increase of 39%–48% and 27%–35% was seen in the water vapour permeation coefficient and oxygen permeation coefficient, respectively (Shaikh et al., 2021). Polyhydroxyalkanoates are completely biodegradable through the esterase activity that breaks down the polymer's linkages from the chain ends (Gupta et al., 2020; Shaikh et al., 2021).

Polyhydroxybutyrate (PHB), which is created when 3-hydroxybutyrate is polymerised, is the most prevalent type of PHA (Savenkova et al., 2000). PHA and PHB both belong to the category of polyesters that are extracted intracellularly by strains of both bacteria and fungi. Polyhydroxybutyrate, having a T_m of 180 °C and a T_g of 55 °C, is renowned for its exceptional UV-resistivity and high optical characteristics. Polyhydroxybutyrate possesses a crystallinity of over 50% (Savenkova et al., 2000). Polyhydroxyalkanoate is the more popular choice among industry professionals and academics due to its greater applicability (Savenkova et al., 2000). In comparison to PHA, PHB is less flexible, has lower thermal stability and biodegrades more slowly (Savenkova et al., 2000). It is commonly recognised that PHB becomes unstable at temperatures below 180 °C and that it can experience molecular weight decrease at even lower temperatures, restricting its processability (Savenkova et al., 2000). Several techniques, including copolymerisation with other alkanoates, the addition of biodegradable polymer, or mixing with a second polymer, are used to improve the processability conditions (Shaikh et al., 2021). With the use of various bacterial strains, numerous researchers have successfully isolated PHA and PHB. Polyhydroxyalkanoate is often made up of repeated (R)-3-hydroxy fatty acid units with a linear chain that are connected by ester bonds. According to the length of its carbon chains, PHA is divided into two categories: short chains (C3–C5) and medium chains (C6–C14) (Sharma et al., 2024).

Polyhydroxybutyrate is biodegraded by bacteria, fungi, and algae in a variety of environments (Shah et al., 2010). The hydrolytic breakdown produces 3-hydroxybutyric acid at a low rate (Shah et al., 2010). The copolymer polyhydroxybutyrate-valerate (PHBV) is created by adding propionic acid to the feedstock (Shah et al., 2010). It is harder and less stiff; hence, it is utilised for packaging (Shah et al., 2010). It degrades in a microbiologically active environment in 5–6 weeks, resulting in

CO₂ and H₂O in aerobic circumstances (Shah et al., 2010). The breakdown is rapid, with methane produced under anaerobic conditions (Shah et al., 2010). Polyhydroxybutyrate and PHBV are commercialised under different trade names: Biopol from Monsanto, Nodax from Procter & Gamble and Kaneka corporation, Eamat from Tianan, and Biomer-P from Biome (Shaikh et al., 2021).

Natural antimicrobials (EOs, essential oils)

Currently, there is an increase in the consumers' demand for natural products, such as EOs, to be used in food packaging materials over synthetic ones (Sharma et al., 2021). Essential oils have been known to be a potential source of antimicrobial compounds (Sharma et al., 2021). Among the various natural sources for obtaining antimicrobial compounds, plant-derived materials—particularly EOs, such as clove, cinnamon, garlic, thyme, oregano, ginger, *Aloe vera*, and black pepper—have proven especially effective. These EOs are considered safe, environmentally friendly, nontoxic, and multifunctional. In addition to enhancing the sensory qualities of food products, they also contribute significantly to their antimicrobial efficacy, making them attractive candidates for natural food preservation strategies (Sharma et al., 2021). The plant EOs are also classified as GRAS by both the FDA and the most recent European regulations (Pezantes-Orellana et al., 2024). Essential oils can be explained as “volatile oils or essences derived from vegetation and characterised by distinctive odours and a substantial measure of resistance to hydrolysis” (Pezantes-Orellana et al., 2024). In general, EOs are complex mixtures of volatile compounds that are present in aromatic plants (Pezantes-Orellana et al., 2024). These compounds can be isolated from distinct anatomic parts of the plants, mainly by distillation and pressing (Pezantes-Orellana et al., 2024). The main components in EOs are terpenes, but aldehydes, alcohols, and esters are also present as minor components (Pezantes-Orellana et al., 2024). Essential oils are synthesised by plants to protect themselves from pests and micro-organisms, to attract pollinating insects, and for signalling processes, but recent studies have demonstrated that EOs might have beneficial effects on human health. For example, cinnamon EO is highly effective against an anti-inflammatory bowel disease colitis; thyme EO is effective against prostate cancer, breast cancer, and lung cancer cells; and garlic and ginger EOs are anti-inflammatory and aid in digestion (Pezantes-Orellana et al., 2024).

Incorporation of EOs with highly desirable smells, such as lime, ginger, garlic, or oregano, into the packaging also reduces the fishy aroma of seafood (Laorenza et al., 2022). Essential oils are aromatic organic compounds extracted from plants that exist in various forms, including aldehydes, alcohols, ethers, ketones, acids, amines, and other volatiles (Laorenza et al., 2022). Seafood products contain high amounts of fat and moisture that increase the release of phenolic compounds from EOs, such as aliphatic hydrocarbons (8–10 carbon atoms) in citrus oil, aliphatic molecules (6 carbon atoms) in leafy-green scented floral oils, or octanal aldehyde in orange oil, which are responsible for odour migration from packaging to seafood (Laorenza et al., 2022).

Biopolymers and synthetic biodegradable polymers incorporated with natural antimicrobials for seafood shelf-life extension

The hydrophilicity and the lack of inherent antimicrobial properties limit the application of PVA for packaging fish and seafood

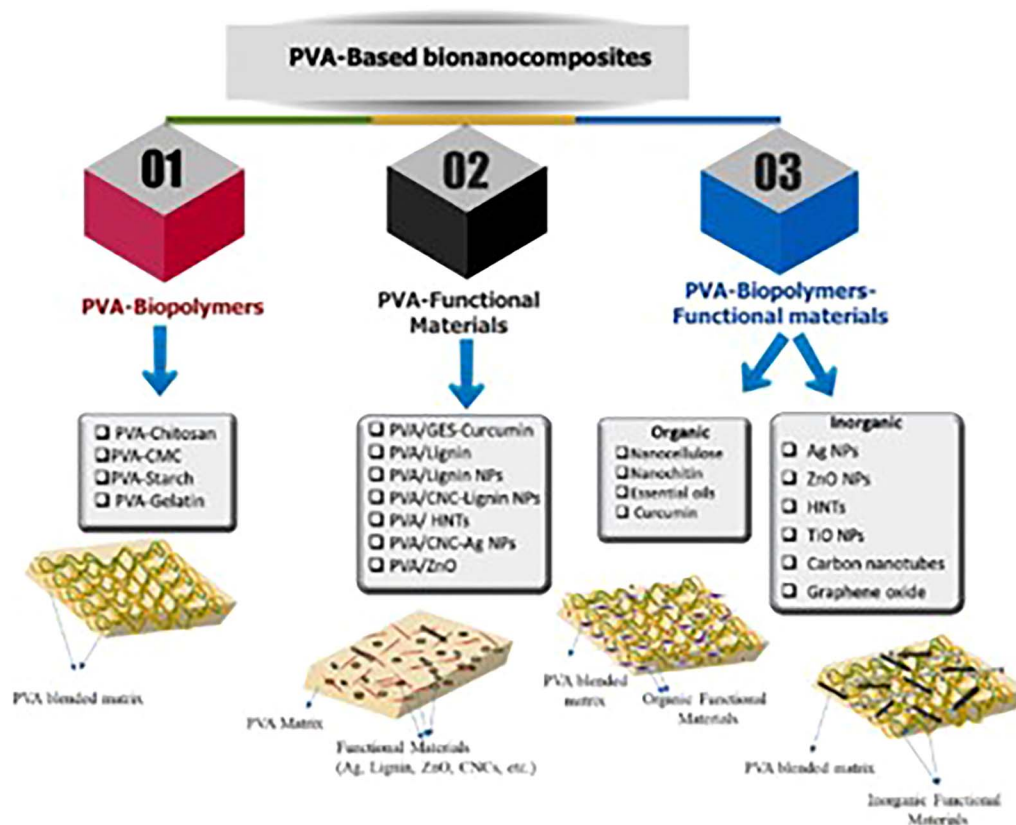


Figure 8. Classification of PVA-based bionanocomposites prepared by blending PVA with various biopolymers and functional materials for various applications, including food packaging (Oun et al., 2022).

products (Malhotra et al., 2015). To overcome this limitation, PVA is combined with other biopolymers like CS, starch, and gelatin, nanoparticles (NPs) or nanomaterials like Lignin NPs, nanocellulose, nanochitin, silver NPs, zinc NPs, titanium oxide NPs, carbon nanotubes, graphene oxide, and NAMs like EOs and curcumin EO (Figure 8), which helps to enhance its thermal, physical, mechanical, and antimicrobial properties (Malhotra et al., 2015).

Polyvinyl alcohol was also combined with many other NAMs like CS, cinnamon oil, "rhubarb" ethanolic extracts, carvacrol EO, and CEO (Ali & Ahmed, 2021; Göksen et al., 2021). These combinations are used as a potential packaging material to extend the shelf-life of food products by reducing the microbial growth on the food's surface. Göksen et al. (2021) developed electrospun PVA containing two different EOs and used citric acid as a cross-linker (Göksen et al., 2021). This active packaging material was used to extend the shelf-life of chicken breast fillets (Göksen et al., 2021). The authors found that even though some amount of EO degraded after annealing, the remaining amount of EO prevented the lipid oxidation process by as much as 68% and showed enhanced antimicrobial properties (Göksen et al., 2021). Many EOs like rosemary EO (Mustafa et al., 2021), oregano EO (OEO) (Kwon et al., 2017), cinnamon EO (Wen et al., 2016), clove oil (Muppalla et al., 2014), black pepper, and ginger EOs (Liu et al., 2024) have been incorporated into the PVA matrix to enhance the antibacterial, antioxidant, and UV barrier properties of the PVA composites thus enabling these NAMs incorporated PVA-based composites to be used as a potential packaging material to extend the shelf-life of fish and fish products (Göksen et al., 2021; Kwon et al., 2017; Liu et al., 2024; Muppalla et al., 2014; Mustafa et al., 2021; Wen et al., 2016).

Mendes et al. (2023) produced nanofiber mats of PLA/PEG blend incorporated with peppermint EO (PEO) at different ratios using the SBS technique to be used for potential food packaging applications (Figure 9). The addition of PEO not only enhanced the antibacterial activity and the CA of the PLA/PEG blend but also decreased the water vapour permeability (Mendes et al., 2023). The incorporation and the effect of EOs on the polymer matrix not only depend upon the type and the concentration of the EOs used, but also on the compatibility of the EOs and the polymer matrix (Virág et al., 2022). Virág et al. (2022) found a significant difference in the adsorption of EOs in the PLA matrix (Virág et al., 2022). It was observed that the difference in the adsorption was due to the difference in the composition of EO solutions, which was determined by the Hansen solubility parameter (HSP) (Virág et al., 2022). Amr et al. incorporated TEO, rosemary EO and oregano EO (OEO) into the PLA matrix using the solvent casting method (Zeid et al., 2019). The authors showed that the PLA-EO composites slowed down the lipid oxidation and extended the shelf-life of rainbow trout fish fillets (Zeid et al., 2019). Min et al. (2021) loaded TEO onto the electrospun PLA nanofibers coated with PVA/PEG blends to evaluate the controlled release of TEO from the composite system (Min et al., 2021). The authors demonstrated that the release rate of TEO can be controlled by the modification of humidity conditions (Min et al., 2021).

Similar to PLA and PVA, PEG has also been combined with NAMs like EOs to package fish and other seafood products (Silva et al., 2020). Silva et al. (2020) developed novel antimicrobial films of PHB blended with PEG and incorporated with CEO to determine its antimicrobial efficacy and its potential migration into the food products (Silva et al., 2020). The authors showed

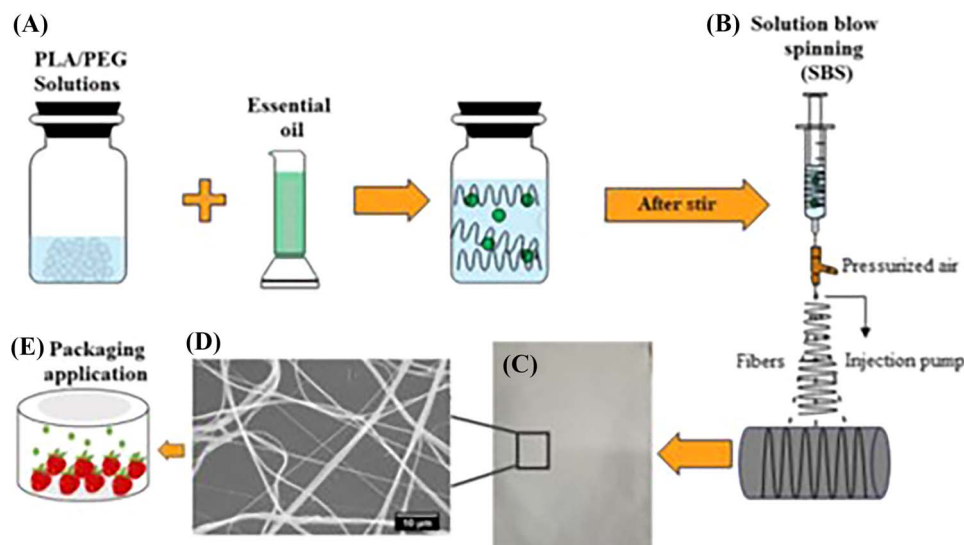


Figure 9. Solution blow spinning for food packaging applications using PLA/PEG solutions encapsulated with peppermint EO (PEO) synthesised. (A) PLA/PEG/PEO solutions prepared for solutions-blow-spinning (SBS) process. (B) Solution-blow-spinning (SBS) process to produce PLA/PEG/PEO nanofibers. (C) Image of PLA/PEG/PEO nanofibers mat. (D) SEM image of PLA/PEG/PEO nanofibers. (E) PLA/PEG/PEO nanofibers used for food packaging (Mendes et al., 2023).

that CEO efficiently inhibited the growth of *E. coli*, *S. aureus*, and *Enterobacter aerogenes* bacteria (Silva et al., 2020). Polyhydroxybutyrate is used widely due to its biodegradability, biocompatibility, superior water vapour permeability, non-toxicity, aroma barrier, and UV-resistance (Silva et al., 2020). However, it is very brittle, which limits its application (Silva et al., 2020). Thus, PEG was added as a plasticiser to increase its flexibility (Silva et al., 2020). The authors concluded that PHB/PEG films incorporated with CEO have the potential to be used for antimicrobial protection packaging applications (Silva et al., 2020). There are very limited studies on the combination of plain PEG with EOs, because PEG is most commonly used as a plasticiser (Silva et al., 2020). Primarily for various applications, including food packaging, PEG is often combined with other biopolymers like PLA, PVA, and CS (Ismaili et al., 2024; Mustafa et al., 2020; Sun et al., 2020).

Apart from these bio-derived or chemically synthesised biopolymers, polysaccharides like CS are combined with EOs like TEO and OEO (Figure 10) to enhance the antimicrobial properties of these films (Hosseini et al., 2015). Gelatin/CS films incorporated with OEO demonstrated a strong antimicrobial activity by inhibiting the growth of *E. coli*, *S. aureus*, *Bacillus subtilis*, and *Bacillus enteritidis* (Hosseini et al., 2015). Incorporation of OEO in the gelatin/CS films reduced the tensile strength and the elastic modulus of the films but increased the light barrier and the water vapour barrier properties of the films (Hosseini et al., 2015). Chamanara et al. (2012) conducted a study to understand and evaluate the effect of CS and CS combined with TEO to extend the shelf-life of butterfly-shaped rainbow trout, which was refrigerated for 15 days (Chamanara et al., 2012). The authors observed that the coating made of CS incorporated with TEO lowered the pH and the TVB-N values when compared to the other samples (Chamanara et al., 2012). The authors also found that the CS-TEO not only formed an edible coating layer over the sample's surface but also helped to extend the shelf-life to more than 14 days (Chamanara et al., 2012). Similar studies were conducted by Aref et al. (2022), where they evaluated the effect of CS, nano-CS, and clove oil on the shelf-life extension of grey mullet steaks stored under refrigerating conditions for 24 days

(Aref et al., 2022). The authors found that the nano-CS coatings with or without clove oil lowered the pH, TVB-N value, and the growth of psychotropic bacteria (Aref et al., 2022). The authors concluded that nano-CS coatings with clove oil were proven to be the best at extending the shelf-life and maintaining the quality of mullet steaks under refrigerating storage conditions for 24 days (Aref et al., 2022). Karakosta et al. (2022) combined CS with laurel EO (LEO) to extend the shelf-life of water buffalo meat stored for 18 days under refrigerated conditions at 4 °C (Karakosta et al., 2022). This study showed that CS/LEO composite lowered the pH of samples, reduced the growth of bacteria, lowered the TVB-N value, and significantly extended the shelf-life of the water buffalo meat to 13–14 days (Karakosta et al., 2022).

Another polysaccharide that is combined with NAMs, such as EOs, and studied for its application in seafood packaging is carrageenan. According to the research study by Kim et al. (2008), carrageenan can produce translucent films with superior mechanical and physical qualities (Kim et al., 2008). Sharma et al. (2021) reported that an 80% increase in UV blocking efficacy was shown by clove oil composite film (Sharma et al., 2021). Prasetyaningrum et al. (2021) crosslinked CaCl_2 with sodium alginate/ κ -carrageenan incorporated with CEO and studied the modification effects on the physiological, morphological, antioxidant, and antibacterial properties of the composite (Figure 11) (Prasetyaningrum et al., 2021). The authors reported that the water solubility and the tensile strength of the films were reduced by the incorporated CEO (Prasetyaningrum et al., 2021). The incorporated CEO also enhanced the antibacterial and antioxidant activities of the film (Prasetyaningrum et al., 2021). Carrageenan was the appropriate material to be combined with alginate for better film characteristics (Prasetyaningrum et al., 2021). Dewi & Purnamayati (2019) studied the application of carrageenan and garlic EO(GEO) to preserve the shelf-life of catfish sausages stored in chilling conditions for 10 days (Dewi & Purnamayati, 2019). In this study, the authors showed that the increase in the garlic EO reduced the tensile strength and increased the elongation of the films, which led to an increase in the plasticity of the films (Dewi & Purnamayati, 2019). The

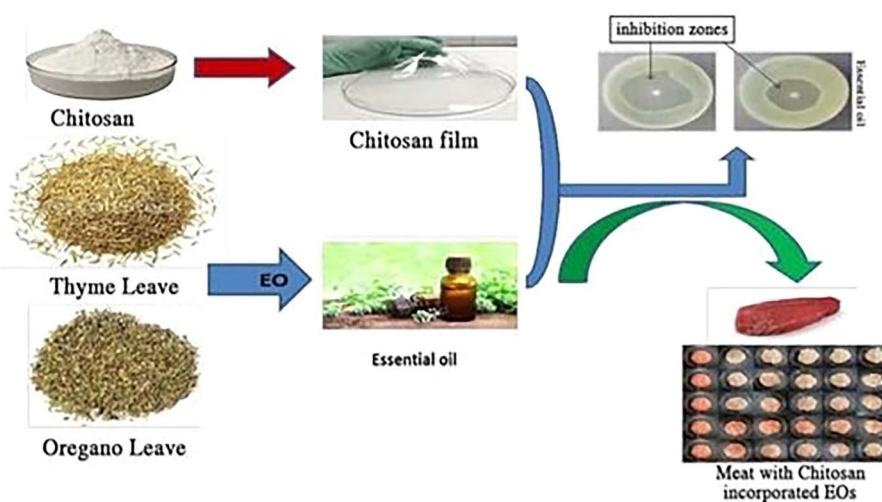


Figure 10. Chitosan film loaded with TEO and OEO to inhibit microbial growth and preserve the quality of beef meat (Gaba et al., 2022).

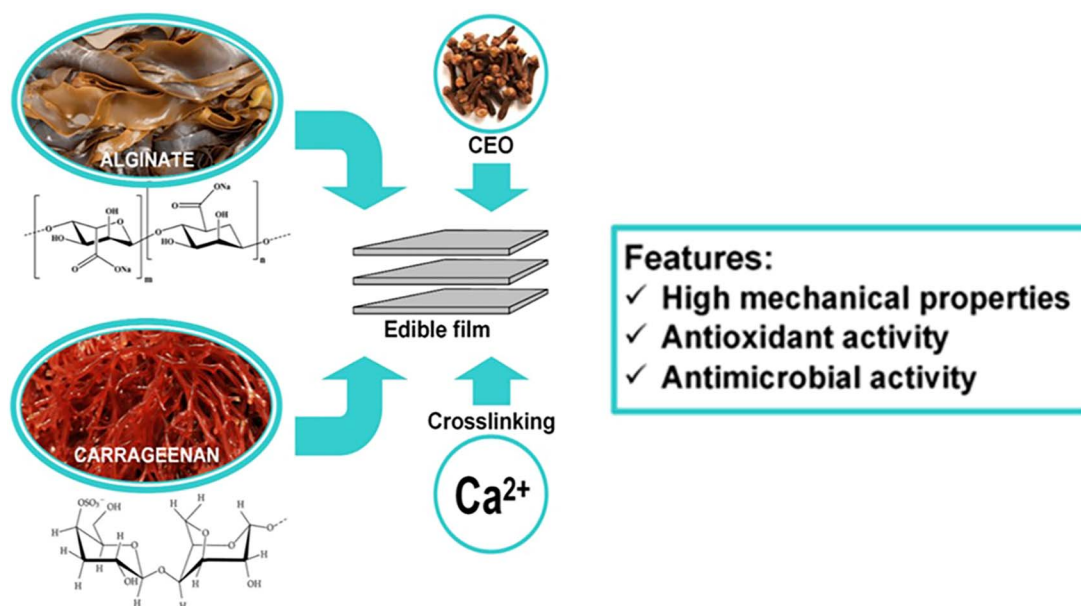


Figure 11. Alginate/ κ -Carrageenan-based edible films incorporated with clove EO, and their superior properties (Prasetyaningrum et al., 2021).

authors also observed that carrageenan edible film containing 0.6% GEO had the highest elongation value of $3.97 \pm 0.33\%$ (Dewi & Purnamayati, 2019). Volpe et al. (2015) studied the application of carrageenan and lemon EO on the shelf-life extension of trout fillets (Volpe et al., 2015). According to this study, fresh trout fillets coated with carrageenan coating and carrageenan coating incorporated with lemon EO exhibited low lipid oxidation and good antimicrobial activity while being stored at 4 °C for 15 days (Volpe et al., 2015). The findings of their study also showed the effectiveness of lemon EO at reducing lipid oxidation and the microbiological growth, while the carrageenan polymeric matrix worked well for preserving the fish fillets' fresh and shiny appearance for longer than seven days in cold storage (Volpe et al., 2015).

Processes for depositing antimicrobials onto the biopolymer films

In this literature review paper, two methods most commonly used to deliver NAMs to a food package, either inclusion into the bulk

or by addition as a coating, are reported. Coating can reduce the amount of NAMs used while providing a rapidly available source. It is worthwhile providing a review of the coating techniques used in academia and industry, as this can influence measurements, such as shelf-life and antimicrobial testing, and the coating techniques need careful consideration in the assessment of the work. Coating of antimicrobial materials has proven to be beneficial not only for the significant reduction in microbial growth on the food surface but also for the shelf-life extension of the food product. Spin-coating and electrospinning are two simple yet versatile techniques that have been used not only for producing polymeric films but also for coating antimicrobials over the polymer surfaces but are not widely used in industry. Dip coating and solvent casting or film casting are widely used in industry. Here, we review a range of these techniques and point to appropriate examples of work in the literature that adopt them.

Spin coating process

Although not used commercially, spin coating is an effective method of making test samples for laboratory work, thus

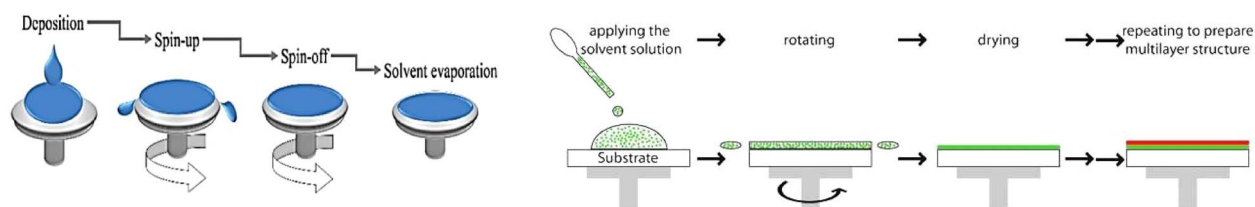


Figure 12. Schematic diagram of thin film formation using the spin coating method. 4 basic steps involved: Deposition or applying the solution, spin-up or rotating, spin-off, and drying or solvent evaporation (Kandjani et al., 2015; Shojaeiarani et al., 2020).

making it a simple and quick thin-film preparation technique (Shojaeiarani et al., 2020). Ultrathin polymer films with uniform thickness can be obtained by optimising the spin speed and the polymer solution concentrations (Shojaeiarani et al., 2020).

Figure 12 is a schematic diagram that gives a general overview of the process of thin film formation by spin coating. The spin coating process consists of four major steps: deposition or applying the solution on the substrate, spin-up or initial rotation to spread the solution across the substrate surface, spin-off at higher speeds to remove excess solution and form a uniform layer of coating, and drying or solvent evaporation. The whole process can be repeated to obtain multiple or thick layers. The existence of surface tension among the molecules of a film and the centrifugal force given by the spin coater often leads to the formation of thin films with appropriate thickness and uniform dispersion of the film over the surface (Xu & Yamamoto, 2012). Polymer solutions are generally coated onto a glass or silicon substrates. Spin coating helps to understand the polymer behaviour over a flat surface, and these films can be analysed and used for various applications (Xu & Yamamoto, 2012). Before beginning the spin coating process, the polymer is first dissolved adequately in a solvent to the extent that it forms a transparent solution (Xu & Yamamoto, 2012). The polymer solution is then deposited onto a solid substrate like a glass slide or a silicon substrate. The substrate is rotated at high speed, where the solvent evaporates, and the substrate is covered by the solid residual polymer solution (Xu & Yamamoto, 2012). The process is repeated, and the parameters are optimised to obtain coatings with even thickness. Xu and Yamamoto (2012) spin-coated poly-L-lactic acid (PLLA) and PCL films on Mg. Using the spin coating technique, the authors were able to obtain uniform, non-porous, amorphous PLLA films and semi-crystalline PCL films (Xu & Yamamoto, 2012). Interesting studies were carried out by Shojaeiarani et al. on the development of PLA and cellulose nanocrystals (CNCs) films by using the spin coating technique to observe their improved performance characteristics to be used as a food packaging material (Shojaeiarani et al., 2020). The authors found that the spin coating method helped to reduce the self-assembly of CNCs. Uniform dispersions of CNCs in the PLA matrix provided higher crystallinity and glass transition temperature (T_g) to the resulting spin-coated films, along with higher CA and lower WVTR (Shojaeiarani et al., 2020).

Electrospinning process

Electrospinning techniques have been appearing to be a state-of-the-art and environmentally friendly technique to manufacture fibrous materials with sufficient physical characteristics to be used for food packaging applications (Bhardwaj & Kundu, 2010). Electrospinning technique offers several advantages, including low production cost, production of very thin fibres from micro to nanometre size ranges with higher surface areas and functionalities, tailored thickness, higher mechanical strengths, higher

barrier properties, and easy processing of even non-electrospin materials (Bhardwaj & Kundu, 2010; Venmathi Maran et al., 2024). During a typical electrospinning process, an electric field is applied between the collector and the needle tip of the syringe containing the polymer solution (Ahmadi Bonakdar & Rodrigue, 2024). The electric field that overcomes the surface tension of the polymer solution electrically charges the polymer, which forms a conical jet from the needle tip of the syringe and gets deposited on the substrate over the collector (Ahmadi Bonakdar & Rodrigue, 2024). The solvent, in which the polymer was dissolved before electrospinning, evaporates between the needle tip and the collector (Ahmadi Bonakdar & Rodrigue, 2024).

Electrospinning can be used to incorporate active compounds into the polymer matrix (Abadi et al., 2022). Figure 13 shows the different types of electrospinning techniques used to synthesise nanofibers. Of these, emulsion electrospinning and coaxial electrospinning are the most widely used electrospinning techniques for food packaging application, where both of these electrospinning techniques are used for incorporating active compounds into the polymer matrix and for releasing the antimicrobial in a slow and sustained manner from the polymer matrix (Abadi et al., 2022). In blend electrospinning, different polymer solutions are blended with functional agents or antimicrobials to form a single solution for electrospinning (Figure 13A) (Abadi et al., 2022). In coaxial electrospinning, the outer chamber of the syringe is used for filling the polymer solution and the inner chamber is used for filling another polymer solution or powders (Figure 13B) (Nista et al., 2015). Nista et al. (2015) used coaxial electrospinning to produce nanofibers of CS-alginate-polyethylene oxide (PEO) polycomplex (Nista et al., 2015). Chitosan and alginate were able to form cross-linked chains easily because of the strong ionic interactions between the chains. The gels were formed at the needle tip quickly, preventing the polymer solution from being ejected from the syringe (Nista et al., 2015). To avoid this problem, strong control over the process parameters and conditions was required (Nista et al., 2015). Pure CS is unable to form fibres with the help of electrospinning owing to the presence of strong hydrogen bonds within and with other polymers, along with poor solubility in organic solvents. So, it is mixed with PEO to make the process more feasible (Nista et al., 2015).

In emulsion electrospinning, two immiscible liquids that are stabilised using an emulsifier are taken in the syringe and electrospun (Figure 13C) (Abadi et al., 2022). In this process, bioactive compounds are first emulsified by adding a surfactant to the solution to create a water-in-oil emulsion. The resulting emulsion is then combined with a polymer or polymer-blend solution (Abadi et al., 2022). The emulsion electrospinning technique can be used to synthesise core-shell nanofibers and other composites that can incorporate functional materials, such as NAMs (Ajalloueian et al., 2014). In addition to achieving targeted delivery and controlled release, emulsion-based electrospun nanofibers can improve the stability, bioavailability, and efficiency

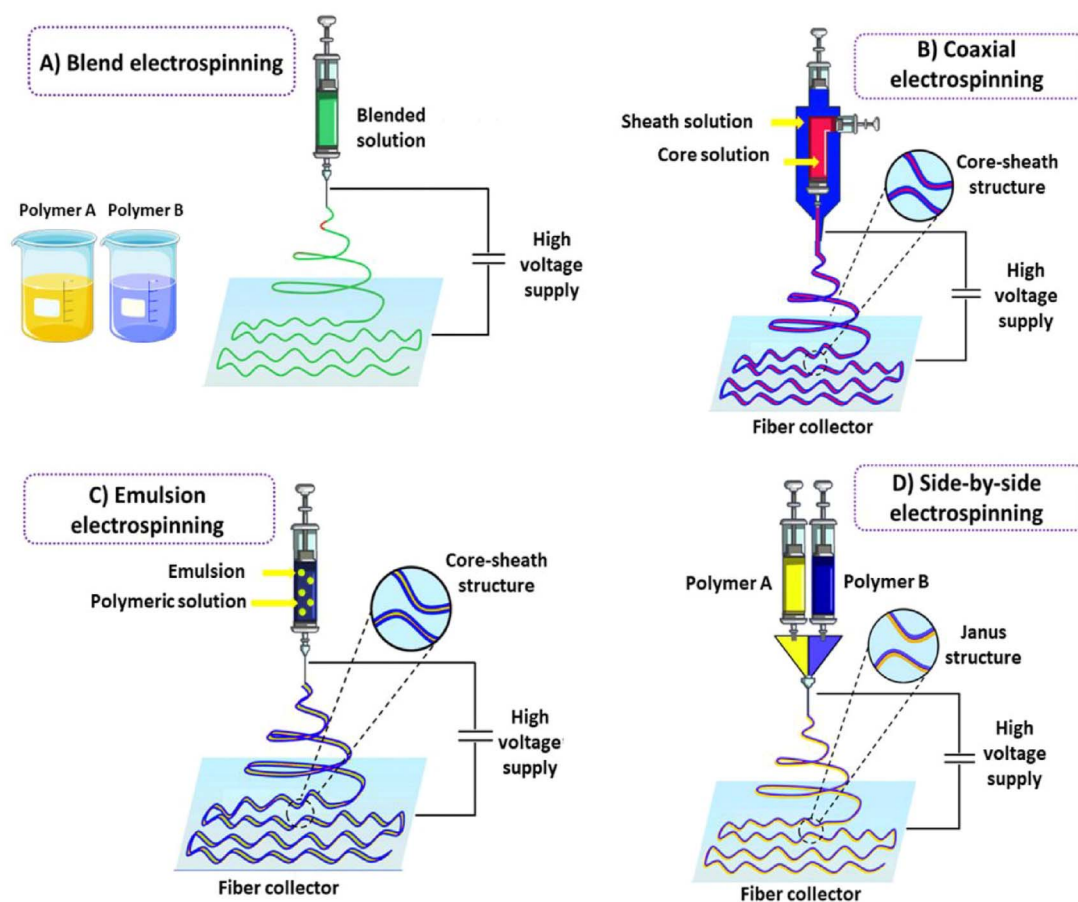


Figure 13. Types of electrospinning techniques used to synthesise nanofibers for biomedical applications: (A) blend electrospinning, (B) coaxial electrospinning, (C) emulsion electrospinning, and (D) side-by-side electrospinning (Abadi et al., 2022).

of encapsulation of bioactive compounds (Ajalloueian et al., 2014). This opens new avenues for improving the barrier performance of emulsion-based electrospun mats when compared to those synthesised using traditional electrospinning techniques (Ajalloueian et al., 2014). Ajalloueian et al. (2014) synthesised a novel blend of poly(lactic-co-glycolic acid) (PLGA) and CS nanofibers using emulsion electrospinning, where the authors used PVA as an emulsifier (Ajalloueian et al., 2014). Upon separating the PVA fibres, PLGA and CS blend scaffolds were obtained (Ajalloueian et al., 2014). Cross-sectional analysis by SEM showed that PLGA and CS were homogeneously distributed in the polymer matrix (Ajalloueian et al., 2014). The CA measurements concluded that the PLGA/CS matrix was more hydrophilic than pure PLGA electrospun fibres (Ajalloueian et al., 2014). A two-compartment approach called side-by-side electrospinning is frequently employed to create Janus nanofibers (Abadi et al., 2022). The two sides of the structure of Janus nanofibers have different compositions (Figure 13D). Unlike the core-sheath structure, both elements have direct interactions with the environment, which can be advantageous for creating new structures (Abadi et al., 2022). In contrast to coaxial electrospinning, the side-by-side electrospinning technique separates two chambers that contain polymers (Abadi et al., 2022).

Solvent casting

Solvent casting, or in other terms, solution casting, is one of the most commonly and extensively used methods for synthesising and manufacturing biodegradable films for seafood packaging

(Lai & Wong, 2022). As seen in Figure 14, biodegradable polymers and plasticisers are first dissolved in an organic solvent to form a film-forming solution (Abelti et al., 2022). This mixture is allowed to stir at either ambient room temperatures or heated to form a homogenous solution. In case of a protein solution, the solution is degassed to remove any air bubbles which might be present in the mixture (Abelti et al., 2022). The solvent mixture is then poured into a petri dish. It is then left to dry in the fume hood for 6 hrs to 2 days, approximately. After the solvent is evaporated, the film is then peeled off and characterised using standard methods (Abelti et al., 2022). Many important parameters must be taken into consideration before carrying out the solvent casting process. The polymer must be selected according to the desired application (Borbolla-Jiménez et al., 2023). While selecting the solvent, it is important to select the volatile solvent or combination of volatile solvents in which the polymer, antimicrobial, and plasticiser are soluble (Borbolla-Jiménez et al., 2023). The viscosity, miscibility, and temperature at which the polymer-antimicrobial solution is dissolved are also important parameters to be considered, while synthesising the polymer-antimicrobial solution for solvent casting (Borbolla-Jiménez et al., 2023). While casting the polymer-antimicrobial solution in a petri dish, it is important to choose the spreading method appropriately, e.g., sonication, centrifugation, etc., (Borbolla-Jiménez et al., 2023). Finally, while drying the solvent-cast films, it is important to note and choose the drying temperature and the time taken to dry the solvent according to the desired application (Borbolla-Jiménez et al., 2023). The solvent casting process has several benefits, such

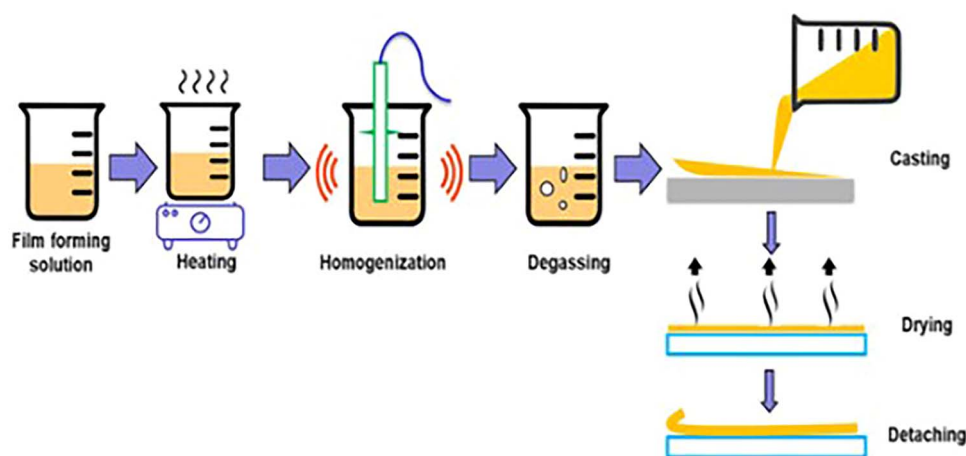


Figure 14. Schematic representation of solvent casting method to synthesise biodegradable polymer films for food packaging (Kumar et al., 2022).

as better physicochemical characteristics, ease of use, inexpensive processing, and sufficient thickness uniformity (Karki et al., 2016). Additionally, it also enables the adjustment of the mechanical and optical characteristics of the film by varying processing parameters like temperature and solvent casting time, which results in the creation of films with high optical clarity and porosity (Anbukarasu et al., 2015). All these advantages of the solvent casting method make it a reliable procedure that is simple to expand industrially.

Solvent casting has been the most widely and frequently used technique to prepare PLA and CS films for packaging and extending the shelf-life of fish and fish products (Zeid et al., 2019). In an interesting study, Zeid et al. (2019) incorporated Thyme, Oregano and Rosemary EOs (EOs) into PLA resin using solution/solvent casting to synthesise antioxidant packaging films to maintain the shelf-life of rainbow trout fish (Zeid et al., 2019). In this study, the TBA test was used to determine the degree of lipid oxidation in fish patties that are covered with PLA films incorporated with EOs. The MDA values obtained showed the measurable antioxidant activity of the PLA films incorporated with EOs when compared to that of the control films (Zeid et al., 2019). The TBA test showed that on day 4 of storage, Thyme EO incorporated PLA films showed a 5.1% decrease in the oxidation degree, while those PLA films incorporated with Rosemary EO showed a decrease of 20.2%, and the PLA films incorporated with Oregano EO showed a decrease of 47.9%, respectively (Zeid et al., 2019). These values showed that the PLA films incorporated with Oregano EO were the most effective at stopping lipid oxidation in minced fish muscle. The authors also suggested that the incorporation of EOs in the polymer matrix may reduce the toxic effects induced by the direct incorporation of EOs as a coating on the packaging film (Zeid et al., 2019). In another interesting study, Qin et al. (2017) studied the effects of different EOs on the antimicrobial effect of the fabricated PLA-based films using solvent casting (Qin et al., 2017). The authors observed and concluded that the EOs not only decreased the glass transition temperature (T_g) of the films but also helped to increase the mechanical and antimicrobial efficiency of the fabricated PLA-based films (Qin et al., 2017). Gulzar et al. (2022) synthesised gelatin/CS coated PLA nanofibers containing nisin for the shelf-life extension of Asian seabass slices (Gulzar et al., 2022). Solvent casting was used to synthesise PLA films, and electrospinning was used to coat these PLA films with gelatin/CS nanofibers containing nisin (Gulzar et al., 2022). The authors observed that when the Asian seabass slices were stored in the pouches made from

these films for 12 days at 4 °C, these films helped to reduce the microbial and chemical deterioration. These films also decreased water vapour permeability but enhanced mechanical properties (Gulzar et al., 2022).

Along with PLA, CS films were also widely produced using the solvent casting technique for seafood packaging applications. Although CS films can also be produced by using the melt extrusion method and other similar methods, pure CS films are prepared only using the solvent casting method (Fathima et al., 2018). Fathima et al. used solvent casting to synthesise PLA/nanoCS composite films by using PEG as a crosslinking agent and PVA as a plasticiser (Fathima et al., 2018). The study showed that the quality of the Indian white prawn samples packed in PLA/nanoCS films was maintained till 15 days of storage, and the CS helped to reduce the microbial growth, thereby aiding in maintaining the shelf-life of the fish effectively (Fathima et al., 2018). In another interesting study by Remya et al. (2016), CS films incorporated with ginger EO were prepared using the solvent casting technique (Remya et al., 2016). Tween 80 was used as an emulsifying agent, and glycerol was added as a plasticiser. The solution was cast onto a square-shaped acrylic plate and then peeled off after drying (Remya et al., 2016). When the steaks of barracuda fish were wrapped in CH-GEO films and stored at 2 °C for 20 days, significant reduction was observed in the growth of both *E. coli* and *S. aureus* bacteria and also a significant reduction was observed in the TVB-N and total mesophilic count of the fish steaks when compared to the steaks wrapped in the synthetic multilayer ethylene vinyl alcohol (EVOH) films (Remya et al., 2016).

However, there are a few limitations of the solvent casting technique (Barik et al., 2024). One major drawback is the higher cost of production of solvent-cast films because of the high energy consumed during the solvent extraction process (Barik et al., 2024). It is also a time-consuming process. Due to these and many other reasons, solvent casting has been widely and frequently used to synthesise films at a laboratory scale. However, significant work is still required to scale this technique up to the industrial scale (Barik et al., 2024).

Dip coating

Among the several techniques for wet chemical thin film deposition, dip coating was the earliest coating technique applied commercially (Ossila Ltd., 2024). Dip coating is a simple and effective technique which is commonly used in manufacturing

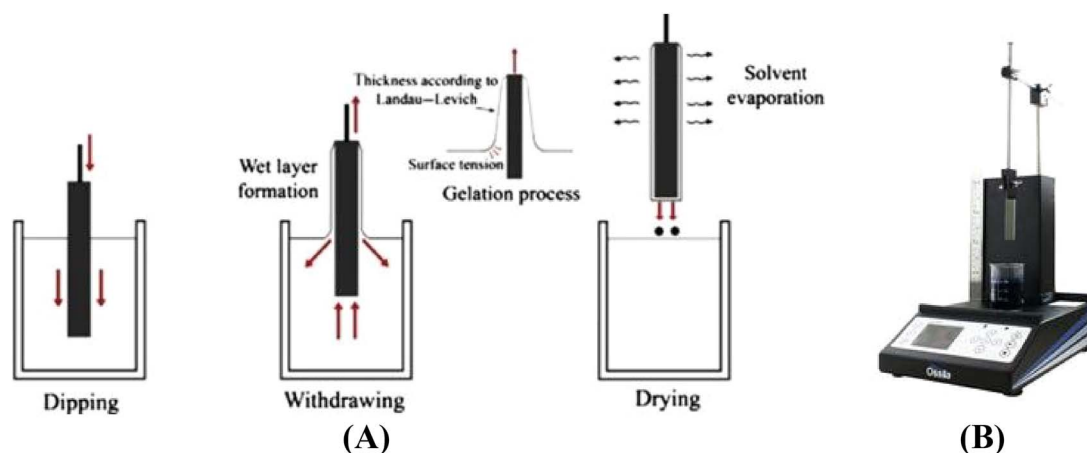


Figure 15. (A) Process steps of the dip coating method (Bobby Zou, 2024) and (B) laboratory-type dip coating device (Ossila Ltd., 2024).

across a wide range of different industries (Ossila Ltd., 2024). Nowadays, sol–gel or more general chemical solution deposition derived coatings are being studied for a range of applications, such as ferroelectrics, dielectrics, sensors and actuators, membranes, superconducting layers, protective coatings, and passivation layers (Brinker, 2013). When the process is optimised, dip coating can be used to produce highly uniform films. Importantly, key factors, such as film thickness, can be easily controlled (Brinker, 2013). One advantage of dip coating over other processing techniques is the simplicity in the design of the instrument used for dip-coating, as seen in Figure 15B. It is low-cost to set up, maintain, and can produce films with extremely high uniformity and a roughness of nanometres (Brinker, 2013).

Dip coating involves the deposition of a liquid film via the precise and controlled withdrawal of a substrate from a solution using a dip coater. The basic dip-coating process consists of three important steps, as shown schematically in Figure 15A (Brinker, 2013).

(1) Immersion (dipping) and dwelling: The substrate is immersed in the precursor solution at a constant speed, followed by a certain dwell time to leave sufficient interaction time of the substrate with the coating solution for complete wetting.

(2) Deposition and drainage (withdrawal): By pulling the substrate upward at a constant speed, a thin layer of precursor solution is entrained, i.e., film deposition. Excess liquid will drain from the surface.

(3) Drying: The solvent evaporates from the fluid, forming the as-deposited thin film, which can be promoted by heated drying. Subsequently, the coating may be subjected to further heat treatment to burn out residual organics and induce crystallisation of the functional oxides.

Besides many of the parameters in the process, the withdrawal speed plays an important role in the final coating thickness and quality of the film (Brinker, 2013). Although there are several approaches/models used to forecast the thickness of the coating film considering withdrawal speed, the viscosity of the solution, gravity force, liquid–vapour surface tension, atmospheric conditions, etc., withdrawal speeds in the range of ~1 to 10 mm/s usually yield successful coatings. In addition to the withdrawal speed, evaporation (drying) plays an important role in dip coating as it determines if the film has a porous or bulk structure (Brinker, 2013). The final film thickness is determined during these two stages by the interplay between the entraining forces, draining forces and the drying of the film. Films are formed in one of three regimes (Ossila Ltd., 2024).

- (1) Viscous flow
- (2) Drainage
- (3) Capillary

The transitions between each of these regimes occur at varying values of withdrawal speed and solution viscosity (Ossila Ltd., 2024). The combination of the three coating regimes ultimately determines the “thickness versus withdrawal speed” behaviour for a thin film. By summing the contributions from the drainage regime and capillary regime, it is possible to obtain an equation that explains the thickness withdrawal speed relationship over a wide range of speeds (Ossila Ltd., 2024). This also allows us to determine the minimum possible thickness that can be coated for a solution. Atmospheric factors, including temperature, airflow, and cleanliness, also play a big part in film quality and must be closely monitored during the dip coating process (Ossila Ltd., 2024).

The advantages and the disadvantages of the coating methods used widely, not only for the synthesis of polymeric films but also for coating antimicrobials on the polymer films for food packaging, are compiled in Table 2.

Conclusions and future recommendations

In this review, we have described the various approaches broadly classified as “sustainable,” which have been used in the development of active packaging materials to extend the shelf-life of fish and other seafood products. As the conventional petroleum-based packaging materials like PP, PA, and LDPE are non-biodegradable, non-renewable, and often cause problems with their recycling process, naturally available biopolymers, and synthetic biodegradable polymers, including polysaccharides, such as CS and carrageenan, microbial-derived biopolymers like PHAs and PHBs, synthetic biopolymers like PLA, and synthetic biodegradable polymers like PVA and PEG, not only have gained significant value but are also being extensively studied and developed to be used as active packaging materials to extend the shelf-life of fish and other seafood products. Various studies show that the mechanical properties, physical properties like hydrophilicity/hydrophobicity, barrier properties, and the thermal stability of single biopolymer-based systems often lag behind those of the petroleum-based synthetic systems. They need to be improved by various methods, like blending two or more different biopolymers. The incorporation of NAMs like EOs (EOs) into biopolymers or their composites can impart novel antimicrobial

Table 2. Advantages and disadvantages of coating techniques, mainly, spin coating, dip coating, electrospinning, and solvent casting, used not only for the synthesis of polymer composite films but also for coating antimicrobials on the polymer films for food packaging.

No.	Method	Advantages	Disadvantages	References
1	Spin coating	<ul style="list-style-type: none">• Formation of thin polymer films with even thickness• Cost-effective as it does not require any high-energy-consuming instruments and processes	<ul style="list-style-type: none">• Cannot be used for large-scale production• Low material efficiency• Difficult to coat on flexible surfaces	(Shojaeiarani et al., 2020; Lors et al., 2025)
2	Electrospinning	<ul style="list-style-type: none">• Robust and simple method to produce fibres from various natural and synthetic polymers with small pores and high surface area• Cost-effective method to produce nanofibres with a high surface-to-volume ratio, high porosity, and enhanced physico-mechanical properties	<ul style="list-style-type: none">• High viscosity causes the solution to get stuck in the tube or at the tip of the needle• Inability to produce fibres as thick as a sheet• Cannot use toxic solvents and high temperatures• Produces porous surfaces that are opaque	(Bhardwaj & Kundu, 2010; Nista et al., 2015)
3	Dip coating	<ul style="list-style-type: none">• Simple and cost-effective method to produce moderately uniform films with roughness of around a few nanometres.• Thickness can be easily controlled by modifying the solution viscosity, immersion time, and withdrawal speed.• Efficient material usage and easy scalability. Suitable for coating large or complex items or substrates.	<ul style="list-style-type: none">• Risk of uneven film thickness with thickness gradients• Can require complex equipment and highly sensitive to conditions• Material shrinkage after the film dries upon dip coating can also crack the films, as seen often while dip coating thicker films	(Ossila Ltd., 2024; Bobby Zou, 2024)
4	Solvent casting	<ul style="list-style-type: none">• Better uniformity of thickness and higher porosity than dip coating and spin coating.• More flexibility of the films and better physical properties than spin-coated and dip-coated films.• Robust process which is easier to scale up industrially.	<ul style="list-style-type: none">• Often, volatile solvents can only be used to dissolve the polymer for solvent casting, where rapid evaporation can lead to phase separation and porous films• High energy consumption during the solvent extraction process• In some cases, heating can be required, which might disrupt the structural stability of the films through differential heating/cooling	(Karki et al., 2016; Anbukarasu et al., 2015; Barik et al., 2024)

and antioxidant properties that these materials do not inherently possess (Panou & Karabagias, 2023).

The techniques widely used to synthesise these multi-component bio-based polymer or biopolymer packaging systems, with improved mechanical properties as well as enhanced antimicrobial activities, such as spin coating, dip coating, solvent casting, and electrospinning, have been discussed, but it should be recognised that these methods are technically challenging to scale towards manufacturing (Ait Ouahioune et al., 2025). The phase separation between the polymers or polymer blends and the incorporated antimicrobials during film production, and the incompatibility between the polymers or polymer blends and the incorporated antimicrobials because of the solvents used to dissolve them, can lead to significant differences between lab-scale equipment and full-scale manufacturing. However, to overcome the various limitations in manufacturing, mechanical properties of the films, and the scaling-up of the films, researchers, regulatory bodies, packaging manufacturers, food firms, and consumers must work collaboratively.

Overall, these recent technological advancements, coupled with producing products that improve sustainability while increasing product shelf-life and reducing waste, provide an imperative to drive research. There is, therefore, an increasing interest in the development of advanced sustainable biodegradable packaging systems, and they are increasingly being relied on to play a significant role in not only extending the shelf-life of fish and other seafood products but also in providing sensory information regarding the freshness of fish and other seafood products to the consumers. This research area thus remains a priority.

Author contributions

Hariharan S. Melarcode (Writing—original draft, Conceptualisation, Methodology, Investigation, Formal analysis), Onur Ertugrul (Conceptualisation, Visualisation, Validation, Formal analysis, Methodology, Writing—review & editing, Supervision), Paola C.Alzate Calderon (Validation, Writing—review & editing,

Supervision), Joseph P. Kerry (Validation, Writing—review & editing, Supervision), and Michael A. Morris (Funding acquisition, Resources, Validation, Writing—review & editing, Project administration, Supervision)

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Conflicts of interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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