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Review

# Nanoengineered sustainable antimicrobial packaging: integrating essential oils into polymer matrices to combat food waste

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**Summary** As global populations rise, the need for sustainable food security and waste reduction solutions becomes crucial. Antimicrobial food packaging presents a strategic defence against spoilage and pathogens and has the potential to minimise foodborne illnesses and global food losses. Essential oils (EOs), known for their inherent antioxidant and antimicrobial properties, are utilised as natural additives in active food packaging materials. These oils, rich in terpenes and hydrocarbons, are promising for food longevity but face challenges like reduced effectiveness in packaging films, highlighting the necessity for nanoengineered solutions to improve their stability and function in polymer matrices. This article aims to review recent work on the use of EOs incorporated in active food packaging. The focus is on the use of nanotechnology to enhance the antimicrobial efficacy of plant-derived essential oils as preservatives. This article illustrates the nanoscale interactions among essential oils, polymers and food substances, aiming to elucidate how technologies such as nanoencapsulation, nanoemulsions and nanocomposites enhance antibacterial and antioxidant performance while improving packaging structural integrity.

**Keywords** Antimicrobial packaging, encapsulation, essential oils, nanostructure, sustainable food packaging.

## Introduction

Food packaging helps extend the shelf life of perishable goods by protecting against environmental factors such as oxygen, water vapour, microbiological and chemical contaminants (Xiao *et al.*, 2014; Ahmad *et al.*, 2023; Garnett *et al.*, 2023). In recent years, researchers have increasingly focused on developing active packaging systems with an aim to enhance foods' shelf life and preserve their sensory attributes (Ahmed *et al.*, 2022; Alves *et al.*, 2023; Thirupathi Vasuki *et al.*, 2023). Active food packaging integrates compounds that are designed to either release into food or absorb any undesirable components released from the food. Active packaging can be broadly categorised into chemoactive and bioactive, depending on the additive added to the food packaging material. Chemoactive packaging, which employs chemicals as active agents, has raised concerns due to its potential adverse health effects and negative impact on the recyclability of packaging materials (Sharma *et al.*, 2021; Shruti *et al.*, 2023). Due to these challenges associated with chemoactive packaging, researchers have explored alternative approaches such as using bioactive compounds sourced from natural origins (Amit *et al.*, 2017). Active packaging incorporating natural bioactive compounds presents a promising alternative to reduce food safety hazards (Hamed *et al.*, 2022; Versino *et al.*, 2023).

Historically, aromatic plants have been inextricably linked with various traditional medicinal practices and culinary traditions across the globe (Giannenas *et al.*, 2020). These plants are praised for their antimicrobial and antioxidant qualities, influenced by factors like extraction techniques, solvents used, cultivation conditions and the parts of the plant utilised (Vieira *et al.*, 2022). Phenolic compounds, ubiquitous across a plethora of edible plants, are instrumental in conferring the biological properties of these plants

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(Shi *et al.*, 2022). These compounds, known as secondary plant metabolites, play a crucial role in protecting plants from environmental challenges, predators and diseases. They also contribute to the plants' colour and sensory properties. Among these natural phenolic compounds are phenolic acids, flavonoids, phenylpropanoids and complex polymers like melanin, lignin and tannin. Flavonoids are the most common and abundant subgroup, resulting from the shikimate and acetate metabolic pathways (Kumar & Goel, 2019; Bié *et al.*, 2023).

Essential oils are renowned for their rich composition of bioactive compounds, including volatile substances, antioxidants and antimicrobial agents (Abdollahi *et al.*, 2012; Bhavaniramya *et al.*, 2019; Jiang *et al.*, 2020; Ni *et al.*, 2021). These compounds are secondary metabolites produced by plants and have been utilised throughout history for their therapeutic, aromatic and cosmetic virtues (Ni *et al.*, 2021; Mohamed & Alotaibi, 2023). Characteristically, essential oils are clear, liquid at ambient temperatures and notable for their unique fragrances. Their identification is facilitated by evaluating their refractive index and pronounced optical activity. Due to their lipophilic nature, essential oils can form emulsions with hydrophilic solvents like ether, alcohol and various oils (Sadgrove & Jones, 2015; Pavoni *et al.*, 2020). Furthermore, essential oils may undergo physical processes that preserve their chemical integrity. The methods for extracting essential oils are diverse, encompassing hydro-distillation, steam distillation, hydro-diffusion and solvent extraction, each contributing to the isolation of these valuable phytochemicals (Turek & Stintzing, 2013; Aziz *et al.*, 2018; Sharma *et al.*, 2021; Saldaña-Mendoza *et al.*, 2022).

Their physical characteristics make them suitable for use in the food industry (Xiao *et al.*, 2014; Santos *et al.*, 2017; Carpena *et al.*, 2021; Rather *et al.*, 2021). In the biological aspect, essential oils (EOs) possess valuable properties including antimicrobial, antifungal, antiviral, antiparasitic and insecticidal or antioxidant properties (Nieto *et al.*, 2018; Hao *et al.*, 2021; Li *et al.*, 2022; Mohamed & Alotaibi, 2023). EOs exhibit antimicrobial properties by permeating through the membranes and layers due to their native lipophilic characteristic (Nazzaro *et al.*, 2013; Khameneh *et al.*, 2019). Thus, incorporating EOs can enhance antimicrobial and antioxidant activity in food packaging, or reduce the water vapour permeability of the packaging.

Incorporating EOs into polymer matrices for food packaging presents challenges, including their stability and sustained release over an extended period. These challenges arise from their inherent immiscibility with many polymer materials. This immiscibility often leads to phase separation, volatility of the essential oils and inconsistent release rates, which can compromise the effectiveness of active packaging solutions. Nanotechnology presents a potential solution for addressing these challenges. For instance, essential oils can be encapsulated within nanocarriers. These nanocarriers can be seamlessly integrated into polymer matrices, ensuring the stability of the essential oils, protecting them from volatility and degradation and facilitating a controlled and sustained release. Nanocarriers not only prevent the degradation of essential oil but also facilitate controlled release of essential oil over time.

This review article provides a comprehensive overview of the strategies and applications of essential oils (EOs) as additives in active food packaging. The use of EOs can be used as the antioxidant and antibacterial additives for food packaging material. Through this review article, we have illustrated the mechanisms by which the efficacy and sustainability of EO-incorporated food packaging material can be enhanced. Some of the nanocarriers that can be used for essential oils are discussed. This article particularly focuses on the use of nanotechnology to improve the antimicrobial efficacy of active food packaging. Potential health, safety and environmental impacts associated with these advanced packaging solutions are discussed in this article. This review is organised as follows: The second section discusses the chemical components and types of EOs that have been used to produce active food packaging. The third section discusses some of the nanocarriers used to encapsulate essential oils. Some of the critical parameters that influence the performance of the EOs and how to improve the effectiveness of EOs are discussed in the next section. After that, common fabrication methods used to produce EO-incorporated active packaging are discussed. Finally, conclusions and future perspectives associated with active packaging are discussed.

## Chemical components and types of essential oils

Expanding our exploration of nanotechnology's impact on improving the stability and release of EOs within food packaging, it is essential to delve into the chemical composition of these oils. This section focuses on understanding the chemical makeup of EOs, primarily categorising them into two main groups, *viz*. terpenes and hydrocarbons.

### Terpenes

Terpenes are composed of isoprene units, which serve as the building blocks defining their structure and function (Ninkuu *et al.*, 2021; Xavier *et al.*, 2023). The classification of terpenes is determined by the number of these isoprene units present in their molecular framework (Masyita *et al.*, 2022). Remarkably, monoterpenes are the predominant class, making up approximately 90% of all essential oils. This category includes the essential oils derived from *Lavandula luisieri* (Zuzarte *et al*., 2022), *Cymbopogon citratus* (Rojas-Armas *et al*., 2020), as well as those extracted from both white and green tea (Lin *et al*., 2021), showcasing the diversity within the monoterpene group. Additionally, terpenes can be further divided based on their cyclic structure into acyclic (non-cyclic), monocyclic (single-ring) and bicyclic (two-ring) groups (Ninkuu *et al*., 2021, Xavier *et al*., 2023). Terpenoids represent a specialised subclass of terpenes, characterised by the incorporation of oxygen into their molecular structure, which often results in enhanced biological activity and solubility.

### Hydrocarbons

Essential oils also contain hydrocarbons, which are molecules composed solely of carbon and hydrogen (Sell, 2020). The structural variety of hydrocarbons allows their classification into aliphatic, alkanes and aromatic types, each contributing distinct characteristics to the oils. For instance, citrus oils are known for their unique acidic aroma, a quality primarily attributed to aliphatic hydrocarbons (Bhavaniramya *et al*., 2019; Sharma *et al*., 2021). Meanwhile, octanal aldehydes play a pivotal role in defining the citrusy fragrance of orange oil (Qi *et al*., 2020). Although aliphatic compounds form a minor component of essential oils, their oxygenated functional groups are instrumental in endowing these oils with their distinctive scents (Eslahi *et al*., 2017). This diversity in hydrocarbon structure not only underscores the chemical complexity of essential oils but also highlights the intricate interplay of components that contribute to their multifaceted aromas and therapeutic properties.

### Types of EOs

Essential oils comprise diverse mixtures distinguishable by their aromatic compounds. Varieties of essential oils encompass *Thymus vulgaris* (thyme), *Eucalyptus globulus* (eucalyptus), *Lavandula angustifolia* (lavender), *Azadirachta indica* (neem), *Syzygium aromaticum* (clove), *Citrus limonum* (lemon), *Cinnamomum zeylanicum* (cinnamon), *Melaleuca alternifolia* (tea tree), *Brassica nigra* (mustard), among others (Ramsey *et al*., 2020; Álvarez-Martínez *et al*., 2021; Sharma *et al*., 2021; Padhi & Routray, 2023). These essential oils, as depicted in Fig. 1, are frequently employed in active food packaging applications. They utilise their volatile nature to combat microbial proliferation, and thus, significantly enhance the preservation and shelf life of food items. Many EOs that are used for active food packaging applications and their components have been recognised by the food and drug administration (FDA) of the USA as generally recognised as safe (GRAS) substances. These include EOs obtained from clove, oregano, thyme and basil, etc.

## Nanocarriers that are used to encapsulate essential oils

One of the major limitations of EOs is its volatility. It is known that more than 90% of the whole EOs are volatile. The volatile constituents of EOs comprises of monoterpene, sesquiterpene hydrocarbons, aliphatic aldehydes, alcohols and esters. Due to the presence of volatile components in EOs, the bioactivity of EOs can be compromised when exposed to air, heat, irradiation and UV illumination. The volatility of EOs also negatively affects their antibacterial properties. EOs that contain high amounts of monoterpene are also easily oxidised when exposed to air.

It is known that the practical application of essential oils for food packaging applications can be enhanced when they are encapsulated within nanocarriers. When essential oils are dispersed within nanocarriers, the enhanced surface-to-volume ratio plays a critical role in improving their activity, absorption, controlled release and targeted delivery compared to neat essential oils. This section discusses some of the common nanocarriers that are used to encapsulate essential oils.

### Nanoemulsions

Nanoemulsions are colloids that are a mixture of two or more immiscible fluids. These immiscible fluids are stabilised with the help of an emulsifier. Typically, the emulsions that have been used to encapsulate bioactive compounds are either oil-in-water or water-in-oil emulsions. The type of emulsion system used is solely dependent on the bioactive compound that is used. For example, oil-in-water nanoemulsions are typically used to encapsulate antibacterial essential oils. In such systems, essential oils are used as the oil phase, which gets dispersed into a water phase that contains a known quantity of emulsifier. Proteins such as gelatin and soybean protein, and polysaccharides such as chitosan, pectin and maltodextrin have been used to produce essential oil-encapsulated oil-in-water nanoemulsion. Some studies have also used a combination of protein and polysaccharide to produce a complex biopolymer system for encapsulation of essential oil. This type of nanoemulsion finds its application in active food packaging as the complex biopolymer system acts as a multilayer shield. It can control the release of essential oils from the packaging and into the food during storage.

### Nanoliposomes

In the literature, nanoliposomes have been employed for encapsulating essential oils. These nanoliposomes

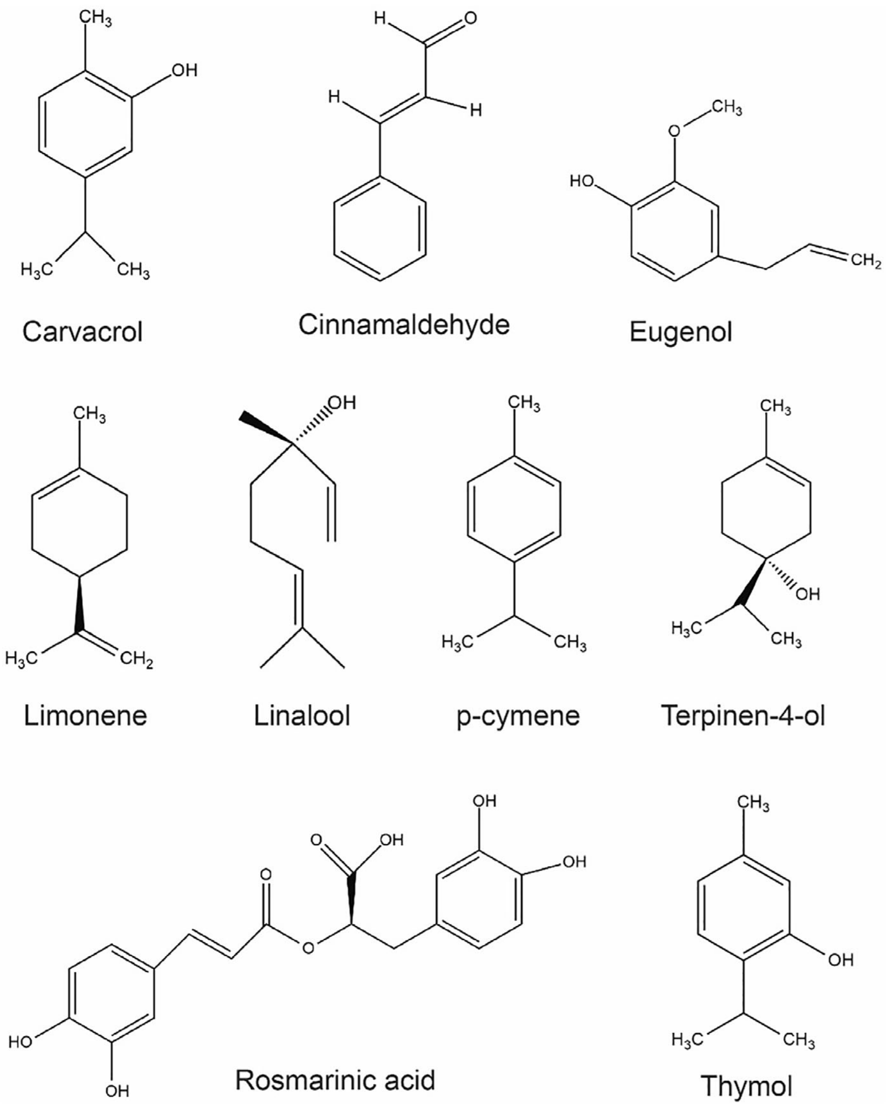

**Figure 1** Essential oils, derived from a variety of botanical sources, have garnered attention for their potent antimicrobial, antioxidant and sensory-modifying properties, making them invaluable in the development of innovative packaging solutions aimed at addressing contemporary food safety and sustainability challenges.

comprise phospholipid bilayers arranged concentrically, creating enclosed spherical vesicles that contain an aqueous core within their structure. Another study (Hammoud *et al.*, 2019) used ethanol injection method to incorporate essential oils such as estragole, eucalyptol, thymol, terpineol, etc. into Lipoid 100 and cholesterol-based liposomes. They show that the liposomes were stable even after 10 months of storage at 4 °C. They show that the use of liposomes enhanced the entrapment of essential oil components. The system is shown to have a controlled release of essential oils from the liposomes. Similarly, Lin *et al.* (2019a) show that the stability of eucalyptus oil can be enhanced by encapsulating it into a liposome. Their results show that the stability and the shelf life of eucalyptus oil are improved when it is encapsulated into solid liposomes. Such essential oil-encapsulated nanoliposomes have been explored for active food packaging applications. For example, chrysanthemum essential oil is encapsulated in chitosan-pectin triple layer liposome and shown to have antibacterial properties against *Campylobacter jejuni* on chicken (Lin *et al.*, 2019b). The results show that the triple-layered liposomes are more stable with excellent release rate and phospholipid oxidation properties compared to single or double-layered liposomes.

### Solid lipid nanoparticles

Solid lipid nanoparticles are composed of a solid lipid matrix that is biocompatible and biodegradable and that is dispersed in an aqueous phase. The structure of the solid lipid nanoparticles are found to be similar to the nanoemulsions. In the case of solid lipid nanoparticles, the oil core is a solid lipid, which contrasts with the liquid oil in the nanoemulsions. Due to this solid oil core, the solid lipid nanoparticles have major advantages over the nanoemulsions. This allows solid lipid nanoparticles to have high encapsulation efficiency and controlled release. Jajarm *et al.* (2021) used homogenisation followed by ultrasonication method to produce solid lipid nanoparticles for food packaging applications. They fabricated *Ziziphora chinopodiodes* essential oil loaded solid lipid nanoparticles that had size less than 100 nm. These solid lipid nanoparticles are shown to have higher toxicity towards red flour beetles compared to free oil.

### Nanostructured lipid carriers

Unlike solid lipid nanoparticles, nanostructured lipid carriers are made up of a mixture of solid and liquid lipids. Due to the presence of both solid and liquid lipids in the core, the core is less crystalline than the core consisting of only solid lipids. This allows the loading of higher concentrations of bioactive molecules in it. Solid lipid in the core provides higher colloidal stability to the nanostructured lipid carriers. Nahr *et al.* (2018) used low-energy emulsification along with high-shear homogenisation and sonication methods to produce cardamom essential oil-loaded nanostructured lipid carriers. They used cocoa butter as solid lipid and olive oil as liquid. They show that the packaging displayed high antibacterial behaviour against *E. coli* and *S. aureus*. These cardamom-loaded nanostructured lipid carriers are shown to have encapsulation efficiency of over 90%. Similarly, Bashiri *et al.* (2020) used a hot homogenisation method to produce chitosan-coated nanostructured lipid carriers and demonstrated its potential as an antioxidant for milk enrichment. These chitosan-coated nanostructured lipid carriers are shown to display enhanced stability and better resistance against aggregation. The encapsulation efficiency is shown to be greater than 84% even after a storage period of 30 days.

## Critical parameters when incorporating essential oils into packaging

### Controlled release of active compounds from packaging

Figure 2 illustrates how food packaging materials, infused with nanotechnology-engineered active compounds, provide robust protection against gases, moisture and multiple forms of degradation such as biological, chemical and physical spoilage. The nanoencapsulation of active ingredients enables a controlled release mechanism, ensuring the prolonged efficacy of these compounds while mitigating direct contact with food, thereby reducing the risk of toxicity (Bhunia *et al.*, 2013). To evaluate the safety and effectiveness of these nanotechnology-enhanced packaging systems, release tests are conducted (Paseiro-Cerrato *et al.*, 2019; Schmid & Welle, 2020). These tests are tailored to the packaged food's characteristics, considering specific time and temperature conditions reflective of its expected usage and storage environments.

It is known that EOs exhibit their antimicrobial activities in liquid as well as in vapour phases. It is shown that EOs have higher antimicrobial properties in vapour phases compared to their liquid phases. This is attributed to the presence of gaseous state of functional groups in EOs. In addition to this, the vapour pressure allows the functional groups to cross the microbial cell membranes. Thus, low concentrations of EOs in vapour phase are effective in inhibiting bacterial growth (López *et al.*, 2005). Another study evaluated the antimicrobial activities of EOs against bovine respiratory pathogens (Amat *et al.*, 2017). They showed that ajowan and thyme EOs inhibited the growth of all bovine respiratory pathogens. Ajowan EOs is shown to inhibit the growth of Gram-negative bacteria such as *E. coli* and *Klebsiella*, and Gram-positive bacteria such as *S. aureus*. The vapour phase of ajowan EO is also shown to display antifungal activity. Vapour phase of thyme EO displayed antimicrobial activities against a range of Gram-negative and Gram-positive bacterial species. Similarly, the vapour phase of cinnamon leaf EO is shown to inhibit *Bacillus cereus* along with several Gram-negative and Gram-positive bacteria. Amat *et al.* (2017) show that ajowan, thyme and cinnamon EOs showed vapour phase antimicrobial property against *M. haemolytics* S1, *H. somni* and *P. multocida*. On the other hand, ginger grass EO displayed weaker inhibition against the bovine respiratory pathogens compared to ajowan, thyme and cinnamon EOs. In a similar study, Boukhatem *et al.* (2014) investigated the antifungal activity of lemon grass EO using both liquid and vapour phases. They show that the lemon grass EO in vapour phase displays higher antimicrobial activity compared to lemon grass EO in liquid phase.

### Essential oil aroma in the food packaging

The antimicrobial efficacy of essential oils against bacteria is typically assessed *in vitro* using methods such as disk diffusion, agar well diffusion, agar dilution and broth dilution techniques (Balouiri *et al.*, 2016). These methods allow for the evaluation of essential oils'

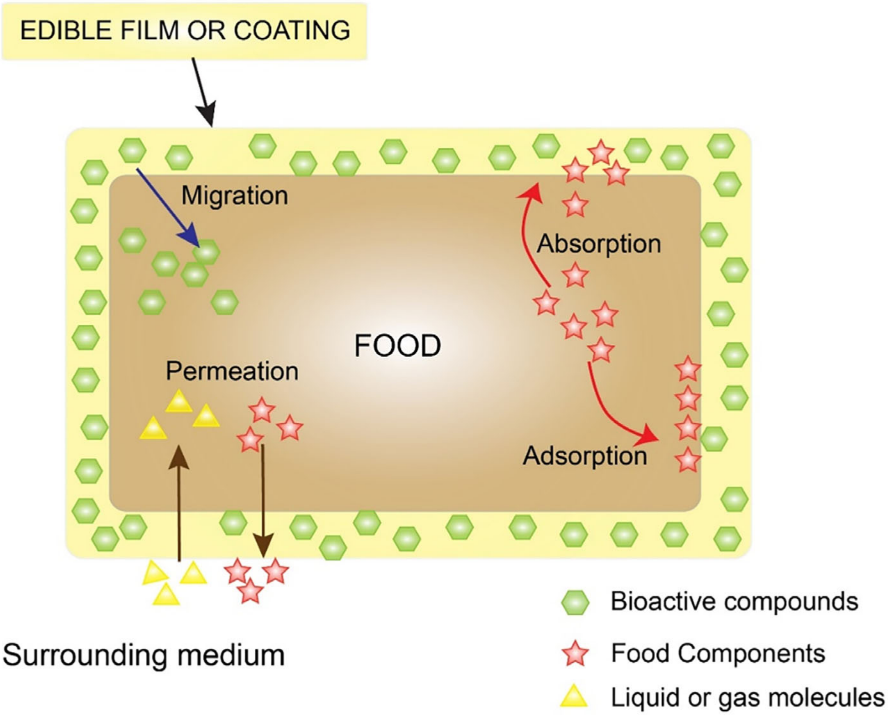

**Figure 2** Food packaging material embedded with active agents offers protection against gases, vapours and various forms of deterioration, including biological, chemical and physical.

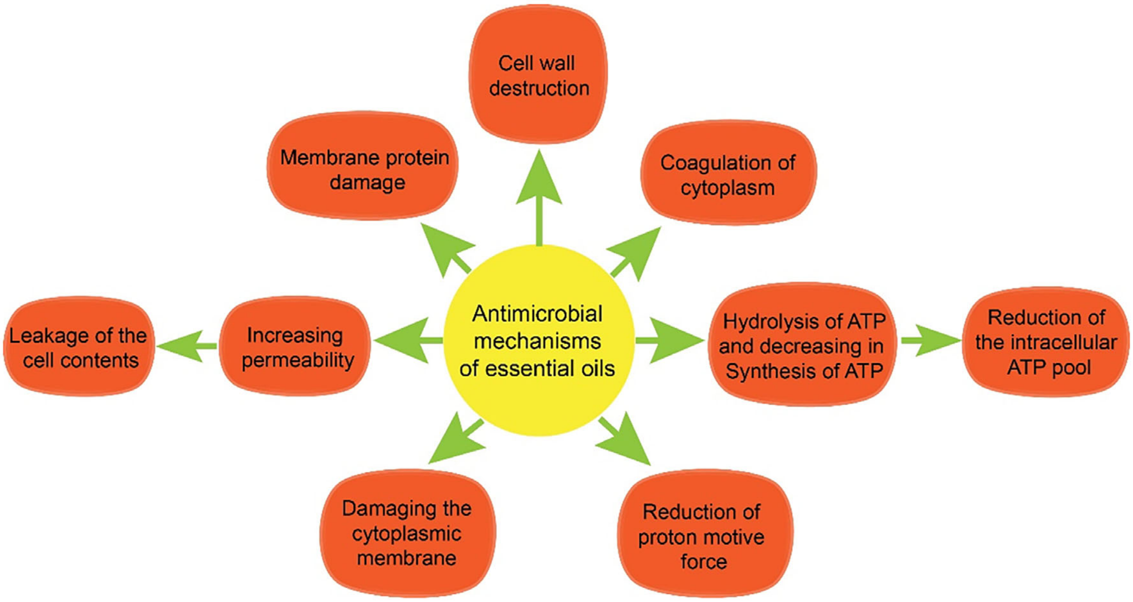

**Figure 3** Comprehensive illustration of the various mechanisms employed by essential oils to inhibit bacterial growth and enhance food safety.

capacity to inhibit microbial growth. Figure 3 illustrates some of the antimicrobial mechanisms associated with essential oils. Their strong aromas may sometimes limit the application of essential oils in biodegradable active food packaging (Atarés & Chiralt, 2016; Sharma *et al.*, 2021). Nonetheless, the integration of essential oils within the matrix of food packaging materials can significantly enhance the antimicrobial properties of the packaging. Nanotechnology enables the encapsulation of essential oils within nanocarriers, which are then embedded into the packaging film's polymer (Sharma *et al*., 2021; Chacha *et al*., 2022). This encapsulation not only enhances the stability and controlled release of the antimicrobial agents but also reduces their direct contact with food, thus maintaining the food's sensory attributes. The controlled release and diffusion of these antimicrobial agents into the food are influenced by several factors, including electrostatic interactions between the agents and the polymer matrix, osmosis, physical changes in the packaging material and environmental conditions (Singh *et al*., 2016; Duda-Chodak *et al*., 2023).

## Enhancing antioxidant properties through essential oil integration in food packaging

Oxidation processes, particularly lipid oxidation, are significant factors in food spoilage that lead to detrimental changes to food's sensory and nutritional attributes (Sahraee *et al*., 2019). Foods rich in fatty acids are especially vulnerable to oxidative damage, which manifests as discolouration, texture alteration, rancidity, nutrient loss and the production of potentially harmful by-products (Domínguez *et al*., 2019). In combating oxidation and extending the shelf life of food products, the integration of natural antioxidants into active packaging emerges as a superior alternative to synthetic additives, potentially increasing consumer trust and product safety (Wang *et al*., 2023).

Essential oils, with their robust bioactive and recognised antioxidant capabilities, play a pivotal role in this strategy (Nieto *et al*., 2018; Sahraee *et al*., 2019; Li *et al*., 2022; Ebrahimzadeh *et al*., 2023). They are increasingly incorporated into edible films and coatings, leveraging their ability to scavenge free radicals and facilitate the transfer of protective compounds to the food surface (Basavegowda & Baek, 2021; Sharma *et al*., 2021). To evaluate the antioxidant efficacy of essential oils in packaging, several analytical techniques can be utilised. The ferric-reducing antioxidant power (FRAP) and the 2,2-diphenyl-1-picrylhydrazyl (DHHP) free radical assays are among the most prevalent methods (Munteanu & Apetrei, 2021). The FRAP assay measures the antioxidant's capacity to reduce $Fe^{3+}$ to $Fe^{2+}$ in a colorimetric reaction at low pH, forming a blue ferrous-probe complex (Benzie & Strain, 1996). The intensity of this complex's colour, measured at 593 nm, correlates with the sample's total antioxidant power. Conversely, the DPPH assay quantifies the ability of antioxidants to neutralise free radicals, a fundamental property of natural antioxidants in plant and food extracts. These methodologies provide a quantitative basis for assessing the potential of essential oils to improve the antioxidant characteristics of food packaging, offering a natural solution to mitigate oxidation and enhance food preservation (Brand-Williams *et al*., 1995; Baliyan *et al*., 2022).

## Impact of incorporating essential oils on the microstructure of food packaging materials

A packaging material is considered biodegradable if it can fully break down into natural elements upon disposal (Moshood *et al*., 2022). Unlike conventional packaging materials, which are primarily hydrophobic plastics, biodegradable and edible packaging is often made from polysaccharides and proteins (Mohamed *et al*., 2020). For example, edible films can be made by spreading an aqueous solution onto a non-stick silicone flat surface and then allowing it to dry at a steady temperature (Sharma *et al*., 2021). The essential oils can be added to the polymer matrix through various techniques, including emulsification or homogenisation (Ebrahimzadeh *et al*., 2023). Within the aqueous phase, polymers that contain essential oils typically form fine emulsions, and upon drying, lipid droplets are integrated into the polymer structure (Lovell & Schork, 2020; Yammine *et al*., 2023).

The final microstructure of the packaging material can be influenced by the structural arrangement of its components, which may experience transformations such as coalescence, creaming and droplet flocculation during the drying phase (Sharma *et al*., 2021). Additionally, the film-forming polymer affects the retention of essential oils (Atarés & Chiralt, 2016). Consequently, the interaction between the polymer and essential oils bolsters emulsion stability, resulting in a markedly enhanced microstructure of the film.

The examination of the microstructure of food packaging materials, enriched with active compounds like essential oils, can be conducted using scanning electron microscopy (SEM), transmission electron microscopy (TEM) or atomic force microscopy (AFM) (Vasile & Baican, 2021). These microscopy methods allow for a detailed examination of the films' structural composition, revealing the integration of lipid droplets, polysaccharides and proteins at the micro and nanoscale. Such detailed analysis provides profound insights into the synergistic interactions between essential oils and biopolymer matrices, elucidating the mechanisms behind the enhanced stability and functionality of the resulting biodegradable packaging solutions. Through this microscopic evaluation, the transformative impact of incorporating essential oils into biodegradable packaging materials is vividly illustrated, showcasing the potential for creating more sustainable, effective and consumer-friendly food packaging options.

### Challenges in employing essential oils in food packaging

While EOs have the potential to reduce or eliminate the need for synthetic preservatives, their antimicrobial effectiveness observed *in vitro* may not consistently persist when applied to actual food products (Carpena *et al*., 2021). This discrepancy arises from the complex interplay of various factors inherent to food, such as pH, water activity, additives, salts, fat and protein content, which are unique to each type of food product. These elements can modify the interactions between the food and the biomolecules integrated into the packaging (Vasile & Baican, 2021). Additionally, external factors such as storage temperature and the atmospheric composition surrounding the food product warrant consideration. If the concentration of EOs in the product is excessive, it might surpass the organoleptically acceptable threshold, leading to alterations in the food product's natural flavour (Hyldgaard *et al*., 2012; Novais *et al*., 2022).

Recently, with the rising demand for materials in active food packaging, essential oils are being utilised to improve properties of food packaging materials (Sharma *et al*., 2021; Rout *et al*., 2022). There are a variety of studies that exploit the benefits of these chemicals in the different approaches. Oregano oil is rich in carvacrol and thymol, two phenolic compounds that have strong antimicrobial activity against a variety of foodborne pathogens, including *Salmonella*, *E. coli* and *Listeria* (Pesavento *et al*., 2015; Hao *et al*., 2021). In a reported study, the authors created nanocomposite films based on low-density polyethylene (LDPE) involving carvacrol with the organo-modified montmorillonite (MMT) was applied as fillers (Persico *et al*., 2009). The combination protected the plant-based substance from thermal degradation and delayed its release from the films. The film exhibited antimicrobial activity against *Brochotrix thermosphacta* and *Listeria monocytogenes*. Moreover, the film also inhibited the growth of *Carnobacterium*, which was damaged in the sensitive bacterial cells by the antibacterial mechanism from phenolic compounds.

Additionally, it is essential that the release kinetics of the EOs from the packaging films should match the food deterioration kinetics. The release kinetics parameters of the EOs depend not only on the type of EO used but also on the processing method adopted to produce the packaging. It is argued that packaging films produced using extrusion and compression moulding techniques lead to the production of higher crystallinity within the packaging film. EOs encapsulated within such polymer materials display lower diffusion coefficient values. On the other hand, packaging films produced using solvent casting exhibit lower barrier properties towards mass transfer. This is attributed to the lower crystallinity of solvent cast films compared to extruded films. The lower crystallinity of the films is shown to be beneficial to the diffusion of the EOs through the polymer matrix (Rojas *et al*., 2021). Packaging films produced using supercritical impregnation lead to low degrees of crystallinity. This allows for a faster diffusion of EOs through the film's structure.

## Fabrication techniques used to incorporate EO into packing films

To incorporate essential oils into packaging films, several fabrication techniques are employed. These methods can be broadly categorised into direct and indirect incorporation. Direct incorporation methods involve blending essential oils with the polymer matrix during the film fabrication process. The typical methods of this category are listed as follows.

### Solvent casting

Solvent casting is a technique used to incorporate essential oils into a polymer matrix (Huang *et al*., 2022; Mondello *et al*., 2022; Gonon *et al*., 2023; Li *et al*., 2023; Tampau *et al*., 2020). The process involves dissolving the essential oil in a compatible solvent and mixing it with the polymer matrix (Fig. 4a). The mixture is then cast into a film and solvent is evaporated, leaving the essential oil evenly distributed throughout the film. This technique is commonly used in the production of biodegradable polymeric materials for food packaging. In this particular research, the solution-casting approach was used to create composite films from zein and PLA in various ratios (Huang *et al*., 2022). PEG was used as a plasticiser and compatibiliser in the composite membrane. A biodegradable film was developed using a bacterial biopolymer, specifically poly (3-hydroxybutyrate-co-4-hydroxybutyrate), or P(3HB-co-4HB), and incorporated thyme essential oil as an antibacterial component, aiming to extend the shelf life of bread through active packaging (Sharma *et al*., 2022). The morphological, thermal, chemical, mechanical, barrier and antibacterial properties of thin solvent-cast films containing 10%, 20% and 30% v/w thyme oil were investigated. Infrared spectroscopy showed the existence of oil in P(3HB-co-4HB) films and their cleanliness, with no visible remnants of solvent, that is, chloroform. Similarly, when stored in a controlled environment, the time-bound release of volatile thyme oil from the films was illustrated. The plasticising effect of thyme oil on films was evidenced by a decrease in tensile strength and crystallinity, while there was an increase in the elongation at break and water vapour permeability of the films containing the oil.

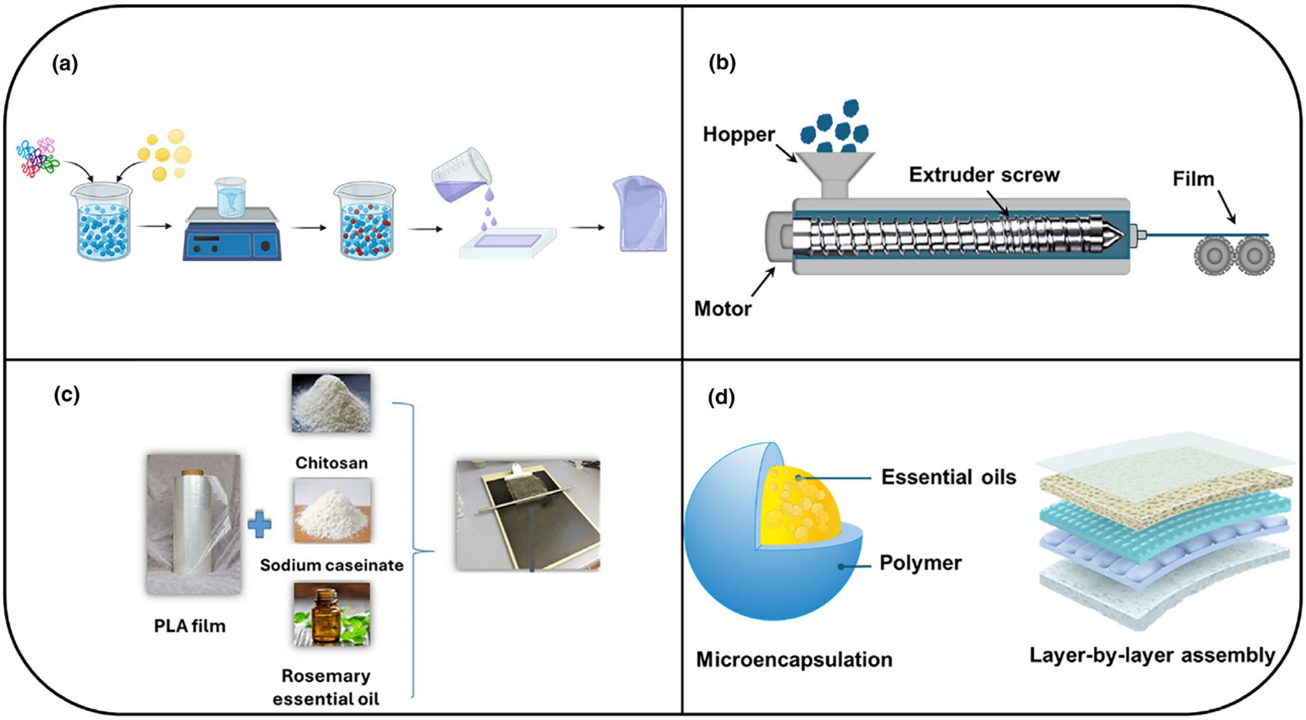

**Figure 4** The techniques applied to incorporate EOs into packing films. (a) Solvent casting technique. (b) Melt extrusion technique. (c) Coating technique. (d) Indirect incorporation including microencapsulation and layer-by-layer assembly technique.

## Melt extrusion

Melt extrusion is a technique used to incorporate essential oils into a polymer matrix (Giannakas, 2020; Aragón-Gutiérrez *et al.*, 2021; Tambe *et al.*, 2021; Granados *et al.*, 2022; Mukurumbira *et al.*, 2022). In this process, essential oils are melted and mixed with the polymer in the molten state before being extruded into a film (Fig. 4b). This process ensures good dispersion of essential oils within the film. Melt extrusion encapsulation is a promising technique for the encapsulation of volatile, unstable and organic compounds including essential oils. This technique is a one-pot process that does not require the use of organic solvents and has decreased energy and water consumption compared to spray drying. Melt extrusion is recognised as a viable and versatile technology for the mass production of encapsulated materials. Recently, a new active film made from ethylene vinyl alcohol copolymer (EVOH) and cinnamaldehyde (CIN) was effectively produced through a hybrid method (Aragón-Gutiérrez *et al.*, 2021). This method included a two-stage process that started with the solvent-casting of an EVOH-CIN polymeric masterbatch, followed by the creation of bioactive EVOH-based films using melt extrusion processing. The vaporisation of the bioactive ingredient caused a minor decrease in the thermal stability of EVOH, according to thermogravimetric measurement. The bioactive properties of the films were examined as well. It was found that the residual cinnamaldehyde in EVOH post-extrusion process endowed the films with some radical scavenging capabilities, as identified through the DPPH assay. Additionally, the films demonstrated antifungal efficacy in vapour phase against *Penicillium expansum* (Aragón-Gutiérrez *et al.*, 2021). In different works, using two separate procedures, direct osmosis, and the green evaporation–adsorption procedure, nanostructures were created by combining thyme oil with natural natrium-montmorillonite and organo-montmorillonite. For the first time, such nanostructures were combined with poly-L-lactic acid in an extrusion moulding technique to create a new packaging film. Research suggests that incorporating clays and essential oils into Poly-L-lactic-acid could enhance the quality of active packaging films (Giannakas *et al.*, 2022). The resulting product has food odour control and shelf-life extension properties and it could be used in active packaging. The films based on OrgMt clay appear to have more potential, and the addition of thyme oil increases their performance as active packaging. Among the examined samples, the PLLA/3%TO@OrgMt and PLLA/5%TO@OrgMt films were identified as the most promising materials for this purpose (Giannakas *et al.*, 2022). Solano & Gante (2012) used extrusion method to disperse EOs in low-density polyethylene. Briefly, EOs are preblended with low-density polyethylene to obtain a master batch using blender at 110 °C at 50 r.p.m. Fragments are then produced using this master batch. Following this, a single screw extruded is used to blend the fragments with virgin resin pellets to obtain the antimicrobial films.

## Coating

Coating is another technique used to incorporate essential oils into a packaging film (Fiore *et al.*, 2021; Mukurumbira *et al.*, 2022; Zhang *et al.*, 2022). In this method, a thin layer of essential oil is coated directly onto the surface of the packaging film (Fig. 4c). This method is often used to create scented or antimicrobial films and provides controlled release properties. Essential oils can be utilised in active packaging as either films or coatings. Their natural derivation and functional properties, including antioxidant and antimicrobial effects, make essential oils a compelling component for biodegradable food packaging solutions. The film surface was coated with chitosan or a chitosan/caseinate combination supplemented with rosemary essential oil at 1% and 2% concentrations to create an antioxidant polylactic acid film (Fiore *et al.*, 2021). The microstructure, water vapour permeability, tensile characteristics and antioxidant capability of the films were all evaluated. Minced chicken meat samples were refrigerated at 4 °C and subjected to *in vivo* analysis at intervals of 0, 4, 7, 11, 14 and 21 days. The presence of the coating lowered the rate of water vapour transmission, with a coating supplemented with 2% rosemary essential oils. The films demonstrated an antioxidant property of up to 6%, which is comparable to 6.25 g of REO/mL (Fiore *et al.*, 2021). *In vivo* tests revealed that active films could decrease meat oxidation during storage in anaerobic-adjusted atmosphere conditions. Samples enclosed in active film maintained steady malondialdehyde (MDA) levels and colour for up to 14 days and also exhibited decreased concentrations of heptanal and ethanol (by 72% and 90%, correspondingly) in comparison to those wrapped in control film (Fiore *et al.*, 2021). In another study, the researchers looked at the microbiological, chemical and sensory properties of turkey breast flesh covered with chitosan and infused with 1% *Origanum vulgare* essential oils (oregano EOs) and 1% or 2% grape seed extract (GSE) and put in the cool condition during 20 days (Mojaddar Langroodi *et al.*, 2021). According to the gas chromatography–mass spectrometry (GC–MS) analysis, oregano EO is high in phenolic compounds, particularly carvacrol and thymol.

Lipid oxidation was markedly reduced ($P < 0.05$) in treatments that combined oregano essential oil (EO) and grape seed extract (GSE), as evidenced by the levels of thiobarbituric acid reactive substances (TBARS) and total volatile basic nitrogen (TVBN) at 0.71 MDA/kg and 10.04 mg N/100 g, respectively. This was observed in chitosan treatments with 2% GSE and 1% oregano EO. Furthermore, on the 20th day of refrigerated storage, the lowest numbers for total viable count (TVC), Enterobacteriaceae, Pseudomonas spp., lactic acid bacteria (Abedi-Firoozjah *et al.*, 2023) and yeast-mould were recorded in these treatments, showing a range of 3.54–4.51 Log CFU/mL. Due to the efficient delaying of microbial and oxidative activities, these combination treatments also received the highest sensory scores (the total acceptability was approximately 7). As a result, a chitosan-based covering containing GSE and oregano EOs can improve the microbiological, chemical and organoleptic aspects of refrigerated fish and turkey meat (Mojaddar Langroodi *et al.*, 2021).

## Indirect incorporation

Indirect incorporation involves the use of carrier materials or encapsulation to protect and deliver essential oils within the packaging films including:

### *Microencapsulation*

Microencapsulation is a technique used to incorporate essential oils into a polymer matrix (Desai & Jin Park, 2005; Ghosh, 2006; Aguiar *et al.*, 2020; Marques *et al.*, 2021). In this method, essential oils (EOs) are encapsulated within micro-sized particles, which are then dispersed within the polymer matrix (Fig. 4d). Microencapsulation ensures controlled release and protection of essential oils, improving their stability. The most commonly used technique for microencapsulation is spray drying (Desai & Jin Park, 2005). The emulsion solvent evaporation process was used to create PLA capsules containing three different active components, *Cinnamomum cassia* essential oil (CEO), eugenol (EEO) and linalool (Giannenas *et al.*, 2020; Campini *et al.*, 2021). The capsules demonstrated two steps of release, with efficacy against infections lasting up to 28 days, indicating an acceptable interior morphology. This research described a method for encapsulating antimicrobial chemicals that might be used in active food packaging. CEO-PLA capsules achieved the greatest results in terms of stability and antibacterial activity (Campini *et al.*, 2021).

In different approach, a composite film was created using potato starch (St) and apple peel pectin (Pec) (Sani *et al.*, 2021). Microencapsulated *Zataria multiflora* essential oil (MEO) and zirconium oxide ($ZrO_2$) nanoparticles were added to the St/Pec film (St/Pec/MEO/$ZrO_2$). The potential characteristics of film including physicochemical and antibacterial were investigated. The best films were utilised to box quail meat. The results showed that adding MEO to the film enhanced the moisture content while adding $ZrO_2$ to the film reduced the moisture content.

Water vapour permeability enhanced as MEO concentration increased and declined as $ZrO_2$ concentration increased. Packaging quail meat with St/Pec-based active films extended the meat's shelf life. The film with the most MEO and the most $ZrO_2$ had the greatest influence on the stability of quail flesh. The encapsulation of the essential oil has improved the essential oil's efficacy time (Sani *et al*., 2021).

### *Layer-by-layer assembly*

Layer-by-layer assembly is a technique used to incorporate essential oils (EOs) into a polymer matrix. In this method, multilayer films are created using layer-by-layer assembly techniques, and essential oils are incorporated within these films (Fig. 4d) (Ferreira *et al*., 2016; Yan *et al*., 2019). Layer-by-layer assembly is also used in the production of biodegradable polymeric materials for food packaging. The utilisation of EOs in active packaging can be applied to the structures of films and coating. Another study reported that both single and triple-layer nanofiber films using cellulose acetate (CA) nanofibers for the outer layers, encapsulating curcumin (CUR) within chitosan flocculated and directly freeze-dried FSP (CFSP and DFSP) via electrospinning and layer-by-layer (LBL) assembly. This research explored the microstructure, efficacy and curcumin release patterns of these films (Hong *et al*., 2023). For three-layer films, the nanofibers had a homogeneous, smooth and beadless morphology, with a transparent interface but significant contacts between the neighbouring layers. The CA-CFSP/CUR-CA three-layer film demonstrated the best flexibility (elongation at break 6.5%), surface hydrophobicity (water contact angle 130.5° 2.6°) and heat stability, implying that it might be used in food packing. The CUR release kinetics in all nanofiber films were well matched to a first-order model, indicating a concentration gradient-driven release. CUR was shortly released in neutral PBS by the monolayer films. In a simulated gastric-intestinal fluid (SGF-SIF), the three-layer CA-DFSP/CUR-CA film exhibited a diffusion-controlled Fickian release mechanism, effectively resisting gastric acid and achieving a controlled release of 18% curcumin within the first 60 min, with a cumulative release reaching 45.2% over 480 min. This demonstrates significant potential for applications in sustained-release systems. This research will not only enhance the surimi processing sector by recovering and utilising FSP from surimi rinse wastewater, but it will also broaden the application of FSP in the food packaging and medication delivery fields (Hong *et al*., 2023).

## Conclusion and future perspective

Post-harvest spoilage of food products due to microbial pathogens has long been an issue needing resolution. Essential oils derived from plants offer a hopeful solution for managing these microbial pathogens, thanks to their inherent antimicrobial properties. In these active packaging, EOs are typically embedded into the packaging films. Incorporation of EOs into the packaging films instead of addition to the product, enables the inhibition of microbes, formation of unpleasant tastes as well as odour. In some cases, the addition of EOs to the packaging is also shown to improve the mechanical, thermal and barrier properties of the film. Alternatively, EOs can be added to the sachets which can be placed inside the packaging. The volatility of EO ensure that they exist in vapour phase within the packaging material headspace. EOs in vapour phase can act as a biofumigant and can display enhanced efficiency compared to EO in liquid phase. However, as the storage duration extends, the essential oils used in films and coatings diminish, resulting in reduced antimicrobial effectiveness. Therefore, recent studies are concentrating on methods to enhance the persistence of essential oils within polymer matrices post-incorporation. Studies have also started assessing the genotoxic potential of compounds present in EOs. Although EOs have been shown to be antimicrobial and antioxidant, EOs may also have some potential toxic effects. It is essential that the toxic effects of EOs should be assessed before they are used in packaging that come in contact with the food. The presence of EOs in the packaging can also result in a higher human exposure of these compounds.

This article has systematically explored the domain of essential oils, elucidating their chemical constituents, several classifications and the potential applications in the industry of films and coatings, particularly within the ambit of food packaging. Moreover, the natural bioactive compounds such as terpenes and hydrocarbons from the EOs group have been elucidated in terms of their chemical properties, revealing potential applications in the robust development of food packaging films. These films exhibit food preservation qualities and prevent microbial growth. Additionally, the interaction between essential oils, material (polymer) and food products can be presented in the combination and stored process, which was analysed related factors including the influence of essential oil incorporation on antibacterial and antioxidant properties, and the consequent effects on the microstructure of packaging materials. Nevertheless, the limitations inherent in utilising essential oils for food packaging were critically determined, highlighting the necessity for an appropriate approach that weighs their advantages against potential challenges such as stability, efficacy and compatibility with different packaging materials.

These drawbacks can be solved by applying several fabrication techniques employed for integrating essential oils into packaging films. Techniques such as solvent casting, melt extrusion, indirect incorporation, microencapsulation and layer-by-layer assembly were meticulously examined, with each method being discussed in terms of its unique benefits and inherent challenges.

This review highlights the significant role of essential oils in advancing food packaging technology, simultaneously acknowledging the complexity and challenges associated with their application. The information derived from this review is anticipated to catalyse further research and innovation in this field, to optimise the utilisation of essential oils in food packaging, thereby enhancing preservation, safety and environmental sustainability.

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## Author contributions

**The H. Duong:** Writing – original draft; writing – review and editing; funding acquisition. **K. Ba Dinh:** Writing – review and editing; writing – original draft; funding acquisition. **Trong Luu:** Writing – original draft; writing – review and editing; formal analysis. **James Chapman:** Writing – review and editing; writing – original draft. **Avinash Baji:** Conceptualization; writing – original draft; writing – review and editing. **Vi Khanh Truong:** Conceptualization; writing – original draft; writing – review and editing; funding acquisition; project administration.

## Ethics statement

Ethics approval was not required for this research.

## Data availability statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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