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Edible Polymers for Essential Oils Encapsulation: Application in Food Preservation

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ABSTRACT

According to the World Health Organization, every year 2 billion people worldwide get sick because of unsafe food. For this reason, food preservation from fungi, bacteria and other contaminants, is important to ensure the safety and quality of food to the consumers.

Now a day, there is an increasing demand for natural preservatives which are safe in the food industry. Among these natural compounds with antimicrobial activity, essential oils have aroused

considerable interest in the food industry. Unfortunately, essential oils have numerous limitations in their use, mainly due to their hydrophobic property, which need to be encapsulated before use. The main goal of this specific review is to focus on the encapsulation of essential oils using edible polymers.

Keywords: Food preservation; antimicrobials; essential oils; encapsulation; edible polymers.

1. INTRODUCTION

The U.S. Food and Drug Administration define food antimicrobials agent as added 'chemical compounds to foods or food packaging able to inhibit activity of bacteria and also to reduce growing process of spoilage microorganisms'.¹

The need to preserve food has been felt since ancient times by using simple and inexpensive methods such as drying (of salty meat and fish), smoking process generally used for fishes and sugar based conservation process (adapted to fruits) but these methods are inefficient to completely protect foods from degradation. During storage, transport and post-harvest processing, microbial contamination such as viruses, fungi, bacteria, moulds, mycotoxin and yeasts cause a loss of quantity and quality of food products.

For this reason, chemicals preservatives were introduced in order to avoid food contamination and spoilage caused by different microorganisms but most of these compounds have shown toxic effects on human health and the environment leading to an increase in the demand for natural origin preservatives. In recent years, food industry developed a great interest in research on antimicrobial compounds, which guarantee food safety and microbiological quality and allows extending the shelf life of food products. Particular attention has been placed to natural antimicrobials but these compounds lose their properties during the processing and conservation phases. In order to solve these problems different strategies have been studied; in particular microencapsulation technique

represents a good strategy to protect antimicrobials compounds.² In recent past, Essential Oils (EOs) and their major bioactive compounds have been widely studied to be use as food preservatives alternative to synthetic ones.^{3,4,5} The main advantage of using essential oils in the presence of additional chemical compounds is the possible synergistic effect, which may induces long time preservation.⁶

The antimicrobial effectiveness of numerous essential oils has been evaluated by Alboofetileh et al. (2014). The used essential oils are clove, coriander, cumin, cinnamon, marjoram, and caraway and evaluated against *E. coli*, *S. aureus*, and *L. monocytogenes pathogens*. The study consists also examining the effect of encapsulating three essential oils in chitosan/clay films on some pathogens. The antibacterial activity was found to be more marked for clove, cinnamon, coriander, cumin, and caraway. Interestingly, the antibacterial activity was preserved when the essential oils are incorporated in the films and influences by the incorporated essential oil nature and amount.

In addition, it was reported that the films containing essential oils were more active against the Gram-positive bacteria (*S. aureus* and *L. monocytogenes*) than the Gram-negative organism (*E. coli*).

For the highest concentration of marjoram essential oil, a complete inhibition of growth was observed which confirms the antimicrobial activity of the essential oil and validates its increase in stability once incorporated in the films.⁷ Chitosan and carob gum polymeric matrices were previously used with different *Citrus* essential oils, and their inhibition potential against *Aspergillus Flavus* (Trichocomaceae) was evaluated with *in vitro* and in artificially infected fruits as reported by Aloui et al. (2014). In addition, to ensure the lack of unusual odors and flavors, the effect of the treatment on the sensorial characteristics of the fruits was studied. Results showed that both bergamot and orange essential oils significantly reduce spore germination at 2% (v/v) concentration. Reductions of 87% and 90% were obtained for bergamot and orange oil respectively. However, bergamot oil was more effective in reducing bacterial growth comparing to orange oil. *Citrus* essential oils, rich in limonene, sabinene, linalool, octanal, myrcene, and other compounds,

when coated with chitosan based formulations, showed a reduction of fungal degradation of infected fruits by 52% - 62% after 12 days. Plus, undesirable flavors or odors were not detected in fruits. These results indicate that combining essential oils with polymeric matrices can be a promising technique for the production of antifungals designed to control the post-harvest growth of *A. flavus*.⁸ In another study, films based on the mucilage of quince seeds loaded with thyme essential oil at a concentration of up to 2% were prepared. Physical properties, mechanical properties, barrier properties, antibacterial activity, and antioxidant activity were evaluated. 11 species of food-related bacterial were used for the antimicrobial activity study using agar disc diffusion method. It was found that all studied bacteria are sensitive to the formulation at 1% essential oil concentration. Plus, the strongest inhibitory effect was observed against *Shewanella putrefaciens* (Shewanellaceae), *L. monocytogenes* (Listeriaceae), and *S. aureus* (Staphylococcaceae). Additionally, the antioxidant activity increased when adding the thyme essential oil to the quince seed mucilage, and the glass transition decreased. The formed structure were studied by scanning electron microscopy.⁹ These formulations offer a new strategy for the protection of food products against degradation following harvesting.

2. ANTIMICROBIAL COMPOUNDS USED AS FOOD PRESERVATIVES

Numerous chemical compounds have been used in the past as food preservatives, such as benzoic acid and p-hydroxybenzoic acid, propionic acid, nitrites, sulfites, butylated hydroxyanisole (BHA) and butylated hydroxytoluene (BHT) (see Table 1 in supporting information). Preservatives have revolutionized eating habits.¹⁰ They are subdivided according to their mechanism of action in antimicrobials, antioxidant and antibrowning.¹¹

Preservatives are accepted if they do not compromise the health of consumers (Council Directive 89/107/EEC). Their possible toxic effects on the organism depends on the dose, route of administration and their chemical form.³² On the other hand, legislation reduced the use and the

doses of chemical ingredients in food and beverage.³³ Despite their effectiveness in food preservation, there are many doubts about their safety on human health; in fact, they can interact with nutrient altering their absorption, transport, metabolism and elimination.

For example, nitrate and nitrite are largely used for “curing” meat; they inhibit anaerobic bacteria, such as *Clostridium botulinum*;³⁴ nitrite is also responsible of the bright pink colour and of a good flavour of meat. Nitrate can be reduced to nitrite by microorganisms present in raw meat or in oral cavity; it inhibits the growth of some microorganisms. Nitrites are converted into nitrosamines, in the acidic environment of the stomach: nitrosamines could be carcinogenic.²² Several studies have shown that nitrite is dangerous for human health: nitrite in fact, can cause methemoglobinemia, by oxidation of oxyhemoglobin to ferrihemoglobin.^{20,35} High consumption of nitrates and derivatives and a possible increase of the risk of gastric cancer has been hypothesized in different studies but more in-depth studies are necessary to establish a secure correlation.^{21,36} A correlation between a high consumption of red meat and a significantly increased risk of development of colorectal cancer has been shown in numerous studies (World Cancer Research Fund [WCRF] 2007).³⁷

3. ESSENTIAL OILS WITH ANTIMICROBIAL PROPERTIES

In the last few years, there has been an increase in consumers' rejection of synthetic preservatives; this caused the need to find natural antimicrobials for food preservation. Among natural compound Essential oils have aroused particular attention in researchers for their widely documented antimicrobial activity.^{38,39} Essential oils (EOs) are natural, aromatic, oily, liquids and volatile compounds obtained from different parts of the plant (flowers, buds, seeds, leaves, twigs, bark, herbs, wood, fruits and roots).³ They possess antibacterial⁴⁰, antifungal⁴¹ and antiviral⁴² activity. There is a big demand of essentials oils from food industry; in fact, due to their antimicrobial activities even against multi-resistant bacteria, they are used for food preservation.³ In the table 2 (see supporting information) are listed different essential oils with antimicrobial activity.

3.1 Chemical composition

Essential oils have very complex chemical composition. They contain volatile compounds in different percentages divided into four categories based on their chemical structure: terpenes, terpenoids, phenylpropenes and “others”. Terpenes are organic compounds constituted by several isoprene units (C_5H_8). They have cyclic structures constituted by hydrocarbon backbone. They are classified in monoterpenes ($C_{10}H_{16}$), sesquiterpenes ($C_{15}H_{24}$) and diterpenes ($C_{20}H_{32}$) based on the number of carbon atoms. Terpenoids are obtained by the enzymatic modification of terpenes into alcohols, esters, aldehydes, ketones, ethers, phenols and epoxides.

Monoterpenoids and sesquiterpenoids are the most abundant terpenes present in essential oils. Phenylpropenes consist of an aromatic phenol group with six carbon atoms and three-carbon propene tail of cinnamic acid. Phenylpropenes and terpenoids are the EOs component with the greatest antimicrobial activity.⁶⁴ Cosentino et al. (1999), established that phenolic components are the main responsible for the antibacterial activity of EOs.⁶⁵ Moreover, antimicrobials activity depends on the substituents on the aromatic ring. Finally, EOs contain a series of degradation components such as allicin and allyl isothiocyanate.⁶⁴ The amount of single component of EOs and their chemical composition are responsible for their antimicrobial activity.⁶⁶ It is important to highlight that harvesting seasons and geographical source influence the chemical composition of a particular plant species.^{51,67}

Gas chromatography/mass-spectrometry analysis is used in order to determine the composition of the EOs.⁵⁰ EOs are extracted from plant raw component by different method that can be classified into conventional methods and innovative methods. Conventional methods are based on water distillation by heating, include hydrodistillation, entrainment by water steam, organic solvent extraction and cold pressing; the main limitation for their use is the thermo-lability of EOs. Conversely, the innovative methods are faster and less expensive such as: supercritical fluid extraction (SCFE), subcritical extraction liquids (H_2 and CO_2), extraction with subcritical CO_2 , microwave extraction process and the instant controlled pressure drop.⁶⁸ Another important feature

of the EOs to be taken into consideration is the possibility that there are interactions between their various chemical components because they are complex mixtures of numerous molecules. For these reasons, it is necessary to keep in mind that the antimicrobial effect of essential oils may be due to the component present in greater quantity or to a possible interaction between different molecules.⁶⁶

3.2 Possible antimicrobial mechanisms

EOs mechanism of action has not yet been clarified but seems to be due to their hydrophobic nature.⁶⁹ Due to the large number of different chemical compounds is more difficult to describe the exact mechanism of antimicrobial action. A possible mechanism of antibacterial action is schematized in figure 1. Due to their hydrophobic nature, EOs component can interact with the lipids of the bacterial cell membrane and mitochondria, altering the normal bacterial structure by increasing the membrane permeability and causing loss of cellular content.^{3,70} Furthermore, there is a reduction of the cytoplasmic pH, a decrease of the ATP synthesis and a change in membrane potential. All these events cause bacterial death.⁶⁹

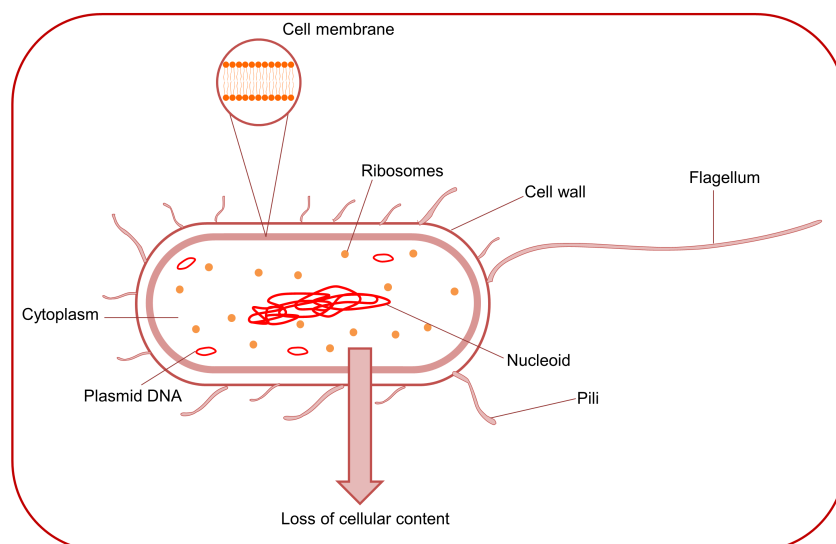


Figure 1. Representation of essential oils mechanism of action. The arrow in the figure indicates the loss of cellular content caused by the possible interaction of the EOs components with the cell membrane.

The antimicrobial tests currently available on the market, used to study the antimicrobial activity of essential oils, are hydrophilic due to the hydrophilic nature of the culture broth (for example, RPMI and Mueller-Hinton broth)^{71,72} while essential oils are hydrophobic. For this reason, surfactants, such as tween 80, are used to mix a hydrophilic phase with essential oils.⁷¹ Tween 80 and other surfactants, could themselves cause a decrease in bacterial growth affecting the results obtained from the antimicrobial activity due to the studied essential oil. There is therefore a need to produce and market new tests to study the *in vitro* antimicrobial activity of essential oils and other lipophilic compounds.

3.3 Limitation in the use of essential oils as food preservatives

The U.S. Code of Federal Regulations and FDA classified several specific chemical active molecules of essential oils such as linalool, thymol, eugenol, carvone, cinnamaldehyde, vanillin, carvacrol, citral, and limonene as generally recognized as safe (GRAS). Clove, oregano, thyme, nutmeg, basil, mustard, and cinnamon are among EOs considered as GRAS.⁷³

Despite their documented antimicrobial power, the use of essential oils in food preservation presents numerous obstacles such:

- i) Hydrophobic component of essential oils can interact with food components such as fat, starch and proteins.⁶⁴
- ii) The high quantity of essential oils to be used to have an antimicrobial effect can alter the taste of food.⁷⁴
- iii) EOs are volatile, photosensitive and temperature sensitive and moreover high temperatures, UV light and oxidation compromise their biological activity.⁶⁸
- iv) The dose necessary to obtain antimicrobial effects could induce toxicity.⁷⁵

These problems represent a limitation in their use, therefore it is necessary to find solutions that can allow the use of EOs in order to have maximum effectiveness and safety.

4. ENCAPSULATION OF ESSENTIAL OILS

Encapsulation techniques of essential oils represent an excellent strategy to overcome the problems related to their use. Encapsulation technique offers many advantages:

- 1) Protecting EOs from degradation;
- 2) Encapsulation of hydrophobic EOs into hydrophilic polymers makes them soluble in a watery environment;
- 3) Masking the strong aroma of EOs that could negatively alter the taste of food.⁷⁶
- 4) Avoiding the interactions with food components.⁷⁷
- 5) Targeting antimicrobials compounds where microorganisms are preferentially located with consequent increasing of their bioactivity and reducing the amount necessary to achieve the antimicrobial effect.⁷⁷

Different encapsulation techniques were used for delivery of EOs used as preservatives in food. In the table 3 (see supporting information) are listed the main technique used in food preservation.

Recently, two new techniques for the encapsulation of essential oils to be used in food preservation have been developed. They are randomly methylated beta cyclodextrins (RAMEB) and Pickering emulsions.

In a recent work, Kfoury et al. (2014), prepared a complex of native cyclodextrins (α -, β -, γ -cyclodextrin) and randomly methylated-cyclodextrin (RAMEB) with the main compounds of basil and terragon essential oils. They obtained a stable system to improve the use of essential oils in food preservation.⁹⁰ Zhou et al prepared and characterized oregano essential oils Pickering emulsion stabilized using cellulose nanocrystals to study their antimicrobial activity on bacteria responsible for food contamination. Although the emulsions antimicrobial activity was slightly less effective compared with pure essential oil; the obtained results encourage further research to improve this type of emulsions in order to use them in food preservation.⁹¹

It has been shown that nano-encapsulation is more effective than microencapsulation because the smaller nanosystems, compare with microcapsules, are more stable and more compatible with food matrix.⁸⁵ In fact, microparticles represent good systems for protecting essential oils but not for increasing antimicrobial activity while nanosized particles are able to increase the passive cellular absorption mechanisms with a consequent greater antimicrobial activity.⁷⁵ Furthermore, unlike microparticles, nanoparticles allow active targeting of the bacterial surface.⁸⁵ Essential oils have also been encapsulated in Solid Lipid Nanoparticles⁹⁰ but these nanosystems have not been applied in food-preservation field.

Table 4 (see supporting information) contains examples of different particles loaded with essential oils, which have different applications, not just antimicrobial.

5. EDIBLE POLYMER USED FOR FOOD ENCAPSULATION

Edible polymers can be defined as safety, biodegradable and biocompatible materials that can be consumed. They are obtained from renewable, edible ingredients and their functionality depends on their barrier, mechanical, and colour properties. The edible polymer are used as carriers for compounds with antimicrobial and antioxidant activity and for the production of films and coatings.¹²⁸

The choice of the material to be used as coating or carrier for the encapsulation of EOs is crucial; in fact the polymer must protect the oil, guarantee its controlled release, be water-soluble, biocompatible and biodegradable and have a low cost.¹²⁹

Edible polymers are safe for human health and they can be consumed with foods. Edible polymers can preserve the nutritional and sensory characteristics of food, its safety and freshness during storage. They are an alternative to synthetic polymers and plastic materials, thus representing advantages both at the environmental level and for human health. The interest of the food industry

towards edible polymers is constantly increasing mainly due to the continued demand for safe and high-quality food by consumers.¹²⁸

They can be produced from proteins, polysaccharides and lipids. There are four categories of compounds used to obtain edible polymers: hydrocolloids (proteins and polysaccharides), polypeptides, lipids, and composites.¹²⁸ The main edible polymers used in the food industry are listed in table 5 (see supporting information).

Gómez-Estaca et al. (2010) found a strong antimicrobial activity of gelatin-chitosan-based films in which clove essential oil was loaded. The clove-loaded edible films caused inhibition of six studied microorganisms such as *Escherichia coli* and *Listeria innocua*.⁵³

In another research study Wu et al. (2012) prepared zein-based edible nanoparticles in which thymol and carvacrol essential oils were encapsulated: the obtained nanoparticles effectively reduce *Escherichia coli* growth. The obtained results demonstrated how essential oils nanoencapsulation allows their use in food preservation.⁸⁰

6.REGULATORY ASPECT

Actually, there is no established accepted worldwide regulatory plan for nanotechnologies. The safety of nanotechnological products is a fundamental requirement for applications in the food industry. For this reason, in 2012, European Union established that all food ingredients obtained from nanotechnological processes must be approved before being put on the market has been drafted in 2012. The U.S. FDA has developed guidelines on the use of nanomaterials for food application while a list of nanofoods and beverages marketed in Europe is available from 2012.¹⁴⁹

The European Food Safety Authority (EFSA) contributes to EU food surveillance by publishing documents related to the safe application of nanotechnologies in food field. EFSA, in 2011, issued a document about the risks arising from the use of nanotechnologies in the food and feed sectors. Furthermore, in 2012, EFSA provided a Nanonetwork to create an easy way of communication with

EU Member states. The purpose of Nanonetwork is to facilitate the exchange of information concerning the safety of the use of nanotechnologies in food field. EU legislation established that all the nanomaterials contained in food products must be specifically indicated in the final label of the product in order to inform consumers.¹⁵⁰

Nanotechnology provides the opportunity to use essential oils as natural food preservatives protecting them from degradation and increasing their effectiveness.

In any case, further studies are needed to understand the possible toxicological effects of nanomaterials before their application as delivery of natural compounds in food preservation.⁷¹

7. FUTURE DIRECTION

The use of essential oils in combination with emergent technologies, such as high isostatic pressures (HIP), thermal treatments, pulsed electric field and actively sensitive packaging against foodborne pathogens has also been described.¹⁵¹ For instance, Espina and co-workers (2011) demonstrated that combined treatments of low concentrations of orange, lemon and mandarin EOs with a mild heat treatment have a strong synergistic antibacterial activity.¹⁵²

Application of nanotechnology to the development of edible coatings includes various nanosystem such as nanoemulsions, polymeric nanoparticles, nanofibers, solid lipid nanoparticles, nanostructured lipid carriers, nanotubes, nanocrystals, or mixtures of organic and inorganic nano-sized components. Several studies of the encapsulation of antimicrobial essential oils into these nanosystem have been documented, however utilization of these system as edible coating has not been fully explored.^{153,154} Out of these nanosystem, nanotube and nanofiber are novel technology for development of edible coating and could be another promising alternative for preservation of foods. Control release packaging (CRP) technology could be another alternative way to deliver the antimicrobial essential oils from active packaging material. CRP uses package to deliver the

antimicrobials to food product, in a controlled manner, in order to extend the quality and shelf-life and protect food from deterioration.¹⁵⁵ This innovative technology is relatively new and intensive research needs to be carried out to fill the knowledge gap. However the use of volatile antimicrobials for CRP systems, has been widely reported.^{156,157,158}

8. CONCLUSION

The spoilage and poisoning of foods by microorganisms is a worldwide problem, which has not yet been completely solved. An appropriate solution for food storage would be beneficial both from an economic point of view and for human health. Natural Essential oils could represent the revolution of food industry because they could completely replace chemical preservatives, obtaining an excellent consent from consumers. The limitation in their use depend on their instability and their unpleasant alteration of the taste of food. Encapsulation of essential oils could be a solution to all these problems. For this reason, it is necessary to find a good method for the EOs encapsulation using suitable edible polymers, safe for human health, which allows the protection of EOs and increase their antimicrobial activity. An innovative EOs delivery system with these characteristics could represent the revolution for food preservation.

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SUPPORTING INFORMATION

Table 1. Classes of food preservatives and their potential toxic effect on human health. a= antimicrobials; b= Antioxidants; *GRAS substances.

PRESERVATIVES	FOOD USE	TOXIC EFFECTS	REFERENCES
Propionic acid, Na, Ca and K salts	To prevent mould and fungal contamination	Reduction of the immune system activity and occurrence of allergic diseases.	(12)
^a Sorbic acid, K salt	Inhibitors of mould and yeast growth.	Genotoxic and mutagenic agent for human lymphocytes.	(13) (14)
Acetic acid	Antimicrobials	Corrosive; it produces vapors which are irritant for the eyes and the respiratory system.	(15)
^a Benzoic acid, Na and K salts	Antimicrobials	Reduction of the immune system activity and possible occurrence of allergic diseases.	(12,16,17) (17)
^a p-Hydroxybenzoate Me, Et, Pr, Bu esters*	Antimicrobials	Genotoxic in human lymphocytes (<i>in vitro</i>). Slight estrogenic activity.	(18,19)
Nitrite, K and Na salts	For curing and preserving meat and fish	Methemoglobinaemia	(20)
Nitrate, Na and K salts		Carcinogenesis (at high doses).	(21, 22)
Sulphites	Antimicrobial in food and beverage	Genotoxic	(23)
Sulphur dioxide		DNA damage	(24)
^b Ascorbic acid	Antioxidant and antibrowning	-	(25)
Na and Ca salts Palmitate*			(10)
Isoascorbic acid Na salt	Antioxidant and antibrowning	-	(26)
BHA (butylated hydroxyanisole)	Antioxidant	Genotoxicity	(27)
BHT (butylated hydroxytoluene)	Antioxidant	Genotoxicity	(27)
Gallates (propyl, octyl, dodecyl)	Antioxidant	Carcinogenicity	(28)
^b ter/-Butylhydroquinone (TBHQ)	Antioxidant	DNA damage	(29)

EDTA (ethylenediamine tetraacetic acid, and Na salts)*	Metal agents	chelating	-	(10)
^b Citric acid	Metal agents	chelating	Chromosomal aberrations	(30)
			Gingival cell death	(31)

Table 2. Essential oils with different antimicrobial activity.

Plant from which EO is extracted	Species of bacteria	EOs or components	References
<i>Cymbopogon</i> spp.	<i>Bacillus ceretis</i> <i>Bacillus subtilis</i>		(43)
	<i>Escherichia coli</i> <i>Pseudomonas aeruginosa</i> <i>Salmonella enteritidis</i> <i>Staphylococcus auerus</i>		(44)
		Neral	(45)
	<i>Fusarium proliferatum</i> <i>Fusarium graminearum</i> <i>Salmonella typhimurium</i> and <i>Staphylococcus aureus</i>	Geranial	(46)
<i>Rosmarinus officinalis</i> L.	<i>Escherichia coli</i> <i>Staphylococcus aureus</i> <i>V. parahaemolyticus</i> <i>Aeromonas hydrophila</i> <i>Listeria monocytogenes</i> <i>Pseudomonas fluorescens</i> <i>Salmonella enteritidi</i>	Camphor/1,8-cineole/borneol	(47)
	<i>Staphylococcus aureus</i> , <i>Bacillus subtilis</i> , <i>Escherichia coli</i> , <i>Klebsiella pneumoniae</i>	α -pinene, β -pinene, myrcene 1,8-cineole, borneol, camphor, and verbenone	(48)
	<i>Fusarium proliferatum</i> <i>Fusarium graminearum</i>	Neral	(45)
		Geranial	(46)
	<i>Salmonella Typhimurium</i> , <i>Y. enterocolitica</i> , <i>Escherichia coli</i> <i>Staphylococcus aureus</i> , <i>Lactobacillus plantarum</i> <i>Aspergillus niger</i> , <i>Geotrichum</i> and <i>Rhodotorula</i>	Carvacrol Eugenol	(49)
<i>Listeria monocytogenes</i> <i>Staphylococcus aureus</i>	Thymol Carvacrol	(50)	

<i>Thymus vulgaris</i> L.	<i>Salmonella typhimurium</i> <i>Staphylococcus aureus</i>	Camphor α -Pinene β -Pinene 1,8-cineole	(51)
<i>Syzygium aromaticum</i> L.	<i>Listeria monocytogenes</i> <i>Escherichia coli</i>	Essential oils	(52)
	<i>Fusarium proliferatum</i> <i>Fusarium</i> <i>graminearum</i>	Carvacrol Eugenol	(45) (46)
	<i>Pseudomonas fluorescens</i>	Carvacrol, thymol, γ - terpinene, p-Cymene	(53)
<i>Thymus vulgaris</i> L.	<i>S. typhimurium</i> <i>Listeria monocytogenes</i> <i>Escherichia coli</i>	Eugenol Thymol	(52)
	<i>Salmonella typhimurium</i> <i>Staphylococcus aureus</i>	Cinnamaldehyde	(54)
<i>Curcuma longa</i> L.	<i>Bacillus cereus</i> <i>Escherichia coli</i>	Turmerone	(55)
<i>Lippia chevalieri</i> and <i>Lippia multiflora</i>	<i>Shigella dysenteria</i> <i>Bacillus cereus</i> <i>Staphylococcus aureus</i> <i>Escherichia coli</i>	p-Cymene Thymol 2-Phenyl ethyl propionate	(56)
<i>Ocimum basilicum</i> L.	<i>Staphylococcus aureus</i> , <i>Yersinia enterocolitica</i> , <i>Aspergillus niger</i> <i>Rhodotorula</i>	Linalool	(50)
	<i>Lactobacillus plantarum</i> , <i>Listeria monocytogenes</i> , <i>Staphylococcus aureus</i> <i>Escherichia coli</i> , <i>Pseudomonas aeruginos</i> <i>Salmonella typhimurium</i> <i>Yersinia</i> <i>enterocolitica</i> , <i>Rhodotorula</i>	Linalool	(57)
	<i>Staphylococcus aureus</i> <i>Escherichia coli</i> <i>Aspergillus niger</i>	Linalool	(58)
<i>Cinnamomum osmophloeum</i>	<i>Coriolus versicolor</i> <i>Lenzites betulina</i> <i>Pycnoporus</i> <i>coccineus</i> <i>Trichaptum abietinum</i> <i>Oligoporus lowei</i> <i>Antrodia taxa</i> <i>Fomitopsis pinicola</i> <i>Laetiporus</i>	Trans-cinnamaldehyde	(59)

	<i>sulphureus Phaeolus schweinitzi</i>	Carvacrol	(45)
	<i>Fusarium proliferatum</i>	Eugenol	(46)
	<i>Fusarium graminearum</i>		
	<i>Escherichia coli</i> ,		
	<i>Klebsiella pneumoniae</i> ,		
	<i>Pseudomonas aeruginosa</i> , <i>Proteus</i>	Eugenol	(50)
	<i>vulgaris</i>		
	<i>Bacillus subtilis</i> , <i>Staphylococcus</i>		
	<i>aureus</i>		
	<i>Campylobacter jejuni</i> , <i>Salmonella</i>		
	<i>enteritidis</i> , <i>Escherichia coli</i> ,		
	<i>Staphylococcus aureus</i> and <i>Listeria</i>		
	<i>monocytogenes</i>		
<i>Coriandrum</i>	<i>Escherichia coli</i>	Linalool	(60)
<i>sativum</i>	<i>Listeria monocytogenes</i>	α -pinene	
L.	<i>Staphylococcus aureus</i>		
	<i>Saccharomyces cerevisiae</i>		
	<i>Aspergillus niger</i> ,	Linalool	(50)
	<i>Geotrichum</i> ,		
	<i>Rhodotorula</i>		
	<i>Staphylococcus aureus</i>		
<i>Coriandrum</i>	<i>Pseudomonas fragi</i>	Linalool	(60)
<i>sativum</i>	<i>Escherichia coli</i>	E-2-decanal	
L.	<i>Listeria monocytogenes</i>		
	<i>Staphylococcus aureus</i>		
	<i>Saccharomyces cerevisiae</i>		
<i>Eucalyptus</i>	<i>Pseudomonas fragi</i>	Piperitone	(60)
<i>dives</i>	<i>Escherichia coli</i>	Terpinen-4-ol	
	<i>Salmonella typhimurium</i>		
	<i>Listeria monocytogenes</i>		
	<i>Staphylococcus aureus</i>		
	<i>Saccharomyces cerevisiae</i>		
<i>Anethum</i>	<i>Escherichia coli</i>	D-limonene	(60)
<i>graveolens</i>	<i>Staphylococcus aureus</i>	Carvone	
L.	<i>Saccharomyces cerevisiae</i>		
			(50)
	<i>Staphylococcus aureus</i>	Essential oils	
	<i>Escherichia coli</i>		
	<i>Yersinia enterocolitica</i>		
	<i>Geotrichum candidum</i>		
	<i>Rhodotorula</i>		
<i>Citrus bergamia</i>	<i>Listeria monocytogenes</i>	Limonene	(61)
	<i>Staphylococcus aureus</i>	Linalool	
	<i>Bacillus cereus</i>		
	<i>Campylobacter jejuni</i> ,		(62)
	<i>E. coli</i> O157, <i>L. monocytogenes</i> ,	Linalool and citral	
	<i>Bacillus cereus</i> and <i>S. Aureus</i>		
<i>Artemisia</i> spp.	<i>Botrytis</i> sp	(Z)-anethole	(63)

	<i>Fusarium spp</i>	(Z)- β -ocimene	
	<i>Verticillium sp</i>	(E)- β -ocimene	
	<i>Drehslera sp</i>	Limonene	
	<i>Nigrospora sp</i>	methyleugenol	
	<i>Phoma sp.</i>		
	<i>Phytophthora capsisi</i>		
	<i>Rhizoctonia solani</i>		
	<i>Bacillus subtilis</i> <i>Brevibacillus brevis</i>		
	<i>Brevibacterium casei</i> <i>Brevundimonas</i>		
	<i>diminuta</i> <i>Burkholderia pyriocinia</i>		
	<i>Chromobacterium violaceum</i>		
	<i>Chryseobacterium indologenes</i>		
	<i>Citrobacter freundii</i> <i>Erwinia</i>		
	<i>chrysanthemi</i> <i>Kocuria rosea</i> ,		
	<i>Kocuria varians</i>		
	<i>Leclercia adecarboxylata</i>		
	<i>Micobacterium saperdae</i> <i>Ralstonia</i>		
	<i>pickettii</i> , <i>Serratia grimesii</i>		
<i>Pimpinella anisum</i>	<i>Aspergillus niger</i>	Essentials oils	(50)
	<i>Geotricum candidum</i>		
	<i>Lactobacillus plantarum</i>		
	<i>Staphylococcus aureus</i>		
	<i>Yersinia enterocolitica</i>		
	<i>Salmonella Typhimurium</i>		
<i>Citrus sinensis</i>	<i>Bacillus subtilis</i>	Limonene	(62)
	<i>Enterococcus faecalis</i>		
	<i>Campylobacter jejuni</i>		
	<i>Escherichia coli</i>		
	<i>Listeria monocytogenes</i>		
	<i>Bacillus cereus</i>		
	<i>Staphylococcus aureus</i>	Linalool and citral	
<i>Citrus lemon</i>	<i>Bacillus subtilis</i>	Limonene	(62)
	<i>Enterococcus faecalis</i>	Linalool and citral	

Table 3. Particles loaded with essential oils with antimicrobial activity on food products.

Pharmaceutical Form	EOs or components	Methods	References
Nanoparticles	Oregano	Ionic gelation	(78)
Nanocapsules	Turmeric oil	Gelation	(79)
	thymol carvacrol	Liquid-liquid dispersion	(80)
	α -tocopherol	Nanoprecipitation	(81)

Microparticles	Oregano	Supercritical fluid Technology	(82)
	Cinnamon leaf and garlic oils	Molecular-complex with β -cyclodextrin	(83)
Microcapsules	Oregano	Spray-drying	(84)
	Orange essential oil	Spray-drying	(85)
Nanoemulsion	Lemongrass Terpenes mixture of <i>Melaleuca alternifolia</i> and D-limonene	Coacervation	(86) (77)
	Eugenol	High Pressure Homogenization (HPH)	(87)
Microemulsion	Eugenol Carvacrol	Mix of lipophilic phase and surfactants	(87)
	Eugenol	Mix of lipophilic phase and surfactants	(88)
Liposomes	Carvacrol and Thymol	Thin film hydration	(89)

Table 4: Particles loaded with essential oil (adapted from El Asbahani et al.⁶⁸).

Essentials oils/EOs compound	Polymer	Pharmaceutical Form	Solvent/non-solvent	Stabilizer agent	Application	Size (nm)	Zeta Potential (mV)	Method	References
<i>Lippia sidoides</i>	Alginate/cashew gum	Nanoparticles	Water	Tween 80	Larvicide	223–399 nm	-30/-36	Spray-drying	(93)
<i>Zanthoxylum limonella</i>	Gelatin	Microparticles	Water/glutaraldehyde solution	Tween 80, glutaraldehyde solution	Mosquito repellent	-	-	Coacervation	(94)
<i>Citronella oil</i>	Chitosan	Microparticles	-	Natural coconut oil	Mood lifting, depression, restless reducing, deodorizing, sterilizing, bug repelling properties	11-225 μ m	-	Modified orifice method	(95)
<i>Lavandin essential oil</i>	PEG n-octenyl succinic (OSA)-modified starches	Microcapsules	Water	(OSA)-modified starches	Natural biocides	30-100 μ m	-	PGSS PGSS-drying	(96)
<i>Croton zehntneri Pax et Hoffm</i>	Alginate/cashew gum	Beads (spherical particles)	Water	Tween 80	Larvicide	0,71-1,46 mm	-	Emulsion preparation	(97)
<i>Lippia sidoides</i>	Chitosan/angico gum	Nanoparticles	Acetic acid/water	Tween80	Larvicide	18.7-271 nm	-21/ -14.7	-	(98)
<i>Lippia sidoides</i>	Chitosan/cashew gum	Beads	Acetic acid/water	Tween 80	Larvicide	1.27±0.2 nm	-	Emulsion preparation	(99)

<i>Oregano, red thyme, and cassia</i>	Zein	Nanospheres	Water/ethanol	-	Feed applications	25-250 nm	-	Phase separation	(100)
<i>Oregano and thyme oil</i>	Zein	Nanoparticles	Water/ethanol	-	Antimicrobials / antioxidant	51.9-328.1	-	Liquid-liquid dispersion method	(80)
<i>Tea tree</i>	Lecithin	Nanoemulsion	Sunflower and palm oil/water	Tween 20, glycerol monooleate, soy lecithin, Cleargum	Antimicrobial	74.4-365.7 nm	-	High Pressure Homogenization	(77)
<i>Cinnamon and thyme essential oils</i>	β -cyclodextrin	Inclusion complexes	Water	-	-	-	-	Co-precipitation	(101)
<i>Elicriso italic, chamomile blue, cannella corteccia (cinnamon), levanda vera (lavender), tea tree, peppermint, eucalyptus globulus juvenile, lemongrass and lemon essential oils</i>	Sodium alginate	Films	Water	Igepal	Anti-microbial and anti-fungal properties (wound dressings but also in food packaging, medical device protection and disinfection, and indoor air quality improvement materials,)	-	-	-	(102)
<i>Lippia sidoides</i>	Chitosan / cashew gum	Nanoparticles	Acetic acid solution/water	Tween 80	Larvicide	335-558 nm	4-49.6	Complex coacervation	(103)

<i>Origanum vulgare</i>	Starch	Microparticles	Carbon dioxide	-	Food preservative	<10 μ m	-	Supercritical fluid technology	(82)
<i>Mentha piperita</i>	Chitosan and cinnamic acid	Nanoparticles	Acetic acid/water	Tween 80	Antimicrobial	<100 nm	-	Ionic gelation	(104)
<i>Rosemary essential oil</i>	Gum Arabic, maltodextrin and modified starch	Microparticles	Water	-	-	12.1–13.5 μ m	-	Spray drying	(105)
<i>Pimenta dioica</i>	Chitosan and k-carrageenan	Microparticles	Acetic acid/water	Tween 40	Antioxidant and antimicrobial in food industry	1172–1224 μ m	-	Complex coacervation	(106)
<i>Oregano</i>	Chitosan	nanoparticles	Acetic acid/water	Tween80	Antimicrobial	281.5-402.2	-	Ionic gelation	(78)
<i>Satureja hortensis</i>	Alginate	Microparticles	Water	Tween 80, Span 80	Antibacterial	47–117 μ m	-	Ionic gelation	(107)
<i>Schinus molle</i>	Maltodextrin and gum Arabic	Microparticles	Water	-	Insecticidal	0.2–40 μ m	-	Spray drying	(108)
<i>Jasmine</i>	Gelatin and gum Arabic	Nanoparticles	Water	-	-	74–384nm	-8.67/-1.92	Complex coacervation	(109)
<i>Salvia hispanica</i>	Whey protein concentrate and gum Arabic or whey protein concentrate and mesquite	Microparticles	Water	Mesquite gum and arabic gum	-	13.17–28.20 μ m	-	Spray drying	(110)

<i>Zanthoxylum limonella</i>	gum Alginate gelatine	et	Microparticles	Water/methylene chloride	Tween 80	Mosquito repellent	209.4 1– 223.1 7µm	-	Emulsion solvent evaporation	(111)
<i>Carvone and anethole</i>	Poly(lactide-co-glycolide)		nanoparticles	Dichloromethane-acetone/ aqueous polyvinyl alcohol solution		Antimicrobial	112– 472 nm	-	Emulsion solvent evaporation, Nanoprecipitation	(112)
<i>Eugenol</i>	Chitosan		Nanoparticles	DMSO/water Acetone/water	Tween 60	Antioxidant for thermal processing	80– 100 nm	16.2–33.5	Ionic gelation	(113)
<i>Ocimum sanctum Linn essential oil</i>	Gelatin		Microparticles	Water	Tween 80	Natural antioxidant	392.3 0 µm	-	Simple coacervation	(114)
<i>Carvacrol and thymol</i>	Egg phosphatidylcholine and cholesterol	L-α-	Liposomes	Chloroform-methanol/Water	-	Antimicrobial	-	-	Film hydration	(89)
<i>Carvacrol</i>	PLGA		Nanoparticles	Acetone/	Tween 80/Pluronic F68	Antimicrobial biofilm	209.8 nm	-18.99	Nanoprecipitation	(115)
<i>Lemon</i>	Silver-modified titanium dioxide		Nanonoparticles	Water/AOT/cyclohexane	-	Antifungal	5-10 nm	-	Microemulsion system	(116)
<i>Nigella sativa</i>	Gum Arabic/maltodextrin			Water	Tween 80	Fortification of processed food and nutraceuticals.	2.33 µm	-	Spray drying	(117)

<i>Orange essential oil</i>	Gum Arabic/maltodextrin	Microparticles	Water	-	-	-	-	Spray drying	(118)
<i>Origanum vulgare</i> L., <i>Cymbopogon nardus</i> G., <i>Majorana hortensis</i> L.	Skimmed milk powder (SMP) Whey protein concentrate (WPC)	Microparticles	Water	-	Antimicrobial	6 -280 μm 2-556 μm	-	Spray-drying	(119)
<i>Rosmarinus officinalis</i> , <i>Larrea sp.</i> , <i>Acantholippia seriphioides</i> , <i>Origanum vulgare</i> , <i>Artemisia sp.</i> , <i>Lavandula sp.</i> , <i>Eucalyptus sp.</i>	Arabic gum; Maltodextrin, and Cellulose microcrystalline	Microparticles	Water	-	Antioxidant	-	-	Spray-drying	(120)
<i>Anethum graveolens</i> , <i>Artemisia arborescens</i> L., <i>Eugenia uniflora</i> L., <i>Santolina insularis</i> , <i>Tea tree</i> , <i>Zanthoxylum tingoassuiba</i>	Egg yolk phosphatidylcholine, dimiristroylphosp-hatidyl choline, Cholesterol	Liposomes	-	-	Antimicrobial	232-360 nm	-	Thin film hydration	(121)

Eucalyptus

Freeze-thaw

camaldulensis

*Eucalyptus
camaldulensis
l*

Modified of
rapid
expansion of
supercritical
solution

*Atractylodes
macrocephala,
Rose*

<i>Thymol</i>	Zein	Nanoparticles	Water-ethanol	Sodium caseinate-Chitosan hydrochloride	Antimicrobial	118.3 0- 204.7 5 nm	- 38.95/28.1 0	liquid-liquid dispersion method	(122)
<i>Artemisia arborescens</i>	Compritol 888 ATO	Solid Lipid Nanoparticles	-	Pluronic F68-Miranol Ultra C32	Antiviral agent	219- 242 nm	-11.3/-39	Hot-pressure homogenization technique	(123)
<i>Camomile trans- Cinnamaldehyde, eugenol, cinnamon bark, and clove bud extracts</i>	β -cyclodextrin β -cyclodextrin	- -	Water/ethanol Water	- -	- Natural antimicrobials	- 0.860- 2.006 μ m	- -	Complexation Freeze-drying method	(124)
<i>Lavandula angustifolia</i>	β -cyclodextrin	-	Water/Ethanol	-	-	-	-	Precipitation	(125)

<i>Vetiver</i>	Gelatin-gum Arabic	Microparticles		Tween 80	-	20–60 μm	-	Complex coacervation	(126)
<i>Oregano essential oil</i>	Inulin	Microparticles	Water	-	-	34.5 μm	-	Spray drying	(127)

Table 5. Edible polymers used in food field with their main characteristics.^{148,128}

Polymer category	Edible polymers	Characteristic	References
Hydrocolloids	Starch	Hydrophilic, poor water vapor and gas barrier properties (O ₂ , CO ₂); good mechanical properties.	(130, 131, 132)
	Chitosan		
	Carrageenan		(133)
	Alginate		(134)
	Pectin		
Polypeptides	Gums	Predominantly hydrophilic, good oxygen barriers, poor moisture-barrier	(135)
	Casein		(101)
	Cellulose		(136)
	Collagen		(137)
	Zein		(138)
	Soy proteins		(139)
Lipids	Milk proteins	(140)	
	Gelatin	(141)	
	Wheat gluten	Hydrophobic, water vapor and gas barrier. Often used in association with others.	(142)
	Wax		
	Fatty acid		(143)
Surfactants	(144)		
Galactomannans and collagen	(145)		
Composite Edible Polymer	Starch	Combination of the functional characteristics of the different polymers	(146)
	Methylcellulose		
	Soybean oil		
	Soy protein and gelatin		(147)