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REVIEW

Polymeric Encapsulates of Essential Oils and Their Constituents: A Review of Preparation Techniques, Characterization, and Sustainable Release Mechanisms

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ABSTRACT

Natural polymer based encapsulation of essential oil (EO) is one of the emerging and challenging area of research in perfumery, cosmetics, flavoring agents, preservatives, therapeutics, etc. The knowledge of formulation techniques and physico-chemical properties of the polymers are the basic requirements for the successful encapsulations of essential oils (EOs). This current review article is focused on a comparative account of various formulation techniques based on their applicability. For the first time, it also reviews various physico-chemical techniques used in the analysis of EO encapsulates to determine their stability, structure, surface morphology, and encapsulation efficiency. Further, the mechanisms involved in the release of EOs from encapsulates, along with various factors affecting their release, have also been discussed.

KEYWORDS

Essential oils; natural polymers; encapsulation; physico-chemical characterization

1. Introduction

Essential Oils (EOs) are secondary metabolites of plant origin. Chemically, EOs are a mixture of hydrogenated and oxygenated monoterpenes, sesquiterpenes, phenols, simple alcohols, ketones, coumarins, etc. EOs are lipophilic, volatile, and aromatic in nature and the aroma depends on individual constituents present in EOs. Because of their sweet aroma, EOs are well known since ancient times for their uses in perfumery, cosmetics, flavoring agents, preservatives, therapeutics, etc. Research in the last few decades has established that EOs have excellent antimicrobial,[1–3] fungicidal,[4–6] herbicidal,[7–10] insecticidal,[11–14] medicinal,[15–17] and antioxidant[18–22] properties. Although EOs have many useful properties, yet their use is limited due to their water insolubility, high volatility, rapid oxidation, and degradation on exposure to air.[23,24] Further due to these factors, the chances of success and scale up of the EO products at industrial level becomes difficult. In order to overcome these problems, research in the past two decades was oriented towards the exploration of techniques

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that can provide physical stability and water solubility to EOs without masking their valuable properties. Various techniques explored include emulsification, aerosol spray, fumigation, and encapsulation of EOs.[25–28] Among these techniques, encapsulation of EOs using biomaterials has emerged as one of the most promising techniques.

Encapsulation is a method of providing a protective layer by coating of single or mixture of polymers over the active ingredient to improve the ingredients stability, sustainability, and ease out the handling. The last ten years of research work on encapsulation of EOs was mainly focused on the synthesis of nanoemulsions, nanocapsules, nano/micro particles, packing films, and capsules for their application in cosmetics, drugs, food packaging, and preservatives.[1,29–36] Encapsulations of EOs have been reported for controlled release of flavoring agents in the manufacturing of chewing gums,[37] as an antimicrobial agent and in the food packaging plastic films[38] at an industrial level. A summary of different polymers explored for encapsulation of EOs extracted from various plants or their constituents, the size of encapsulates, and formulation type are enlisted in Table 1. Further, Table 1 also highlights that the applicability of the encapsulated EOs ranges from food industries as preservative to therapeutics. The polymers used in various EOs encapsulation studies are of natural origin and biodegradable in nature.

Literature survey of scientific database search engines like PubMed, Web of Science, Scopus and Google Scholar using the words "Essential Oils and Encapsulation" reveals that nearly 120–125 research papers have been published in the field and majority of them (nearly 60%) in the last 5 years (Fig. 1). During the past ten years, few review articles have also been published in reputed journals.[29,38,87,88] Martín et al. reported a detailed account of co-precipitation processes/ technologies for encapsulation of EOs and its applications.[87] Marques has extensively reviewed β-Cyclodextrin (β-CD) based encapsulation process for EOs and other volatile components of plant origin.[38] In another study, Matalanis et al. have provided a comprehensive review of biopolymer-based delivery systems for encapsulation, protection, and release of lipophilic compounds.[88] Recently, Bilia et al. have discussed the strategies for development of EOs loaded nano-systems for therapeutic approaches.[29] The available literature mainly provides information about the selected polymers for the encapsulation studies, techniques for the development of different carrier systems, and transformation of these into nano/micro sizes for improving targeted and sustainable delivery of EOs.

In this present manuscript, an attempt has been made to comprehensively review all the carrier polymers and their derivatives used until now for encapsulation of EOs. This manuscript provides an in-depth description of various technologies used in encapsulation along with their comparative account of advantages and disadvantages. The present manuscript reviews various physico-chemical techniques used in the analysis of encapsulates to determine their stability, structure, surface morphology, and encapsulation efficiency for the first time. It also enlists the mechanisms involved in the release of EOs from the encapsulates along with various factors affecting their release. The authors are confident that the manuscript will be highly useful for the beginners who wish to work in this field.

2. Biomaterials for EOs encapsulations

A broad range of biomaterials have been used for the encapsulation of EOs. Among these, polymers are most widely used followed by oligomers and associated colloids. However, the

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Table 1. A comprehensive account of EOs-Polymers encapsulate, size, formulation type, and their applications.

S. No.	Polymer used	Source Plant of Essential Oils/ Constituents	Plant Part	Size of Formulation	Formulation type	Application
1.	Cassava starch	Limonene	Commercial	0.2 – 15 μm	Microparticles	Food industry[39]
2.	Wheat starch	Thymol *Syzygium aromaticum, Origanum* and Camphor	Commercial	5–300 μm	Porous microspheres	Entrapping of the EO in the pores due to very less aggromeration even at high loading rate[40]
3.	Modified Starches	*Mentha piperita* L.	Leaves	11.4–137.6 μm	Microparticles	Flavor retention[41]
4.	n-octenyl succinic (OSA)-modified starches	*Lavandula hybrid Rev. Clone super*	Commercial	500–1400nm	Particles	For making physically stable agrochemical formulations of the EOs[42]
5.	Ethyl cellulose, hydroxypropyl methylcellulose and poly (vinyl alcohol)	Citronellal, camphor, eucalyptol, limonene, mentol and *Eucalyptus*	Commercial	<400 nm-2μm	Polymer-blend	Improvement in release profile and stability of fragrance[43]
6.	Maltodextrin	*Cannarium commune* and *Mentha piperita*	Leaves	nd	Microparticles	Increased stability due to encapsulation[44]
7.	Maltodextrin and modified starch	*Rosmarius officinalis*	Leaves	12.0 μm (mean diameter)	Microparticles	Lesser hygroscopic, larger oil and moisture content on increasing the EO concentration in the emulsion formed by spray drying process[45]
8.	Maltodextrin, trehalose and sucrose	Orange	Orange peel	nd	Micro/Nanoparticles	Flavor retention in food industry[46]
9.	Maltodextrins combined with different wall materials	*Linum usitatissimum*	Seed	1.73–2.11 μm	Microparticles	Oxidative stability[47]
10.	Maltodextrin and hydroxypropylmethyl cellulose	*Salvia hispanica* L.	Seeds	10–100μm.	Microparticles	Oxidative stability[48]
11.	Inulin	*Origanum vulgare* L.	Leaves	(3–4.5 μm)	Microcapsules	Different release rate depending on structural changes in matrix wall due to interactions between inulin and the EO[49]
12.	Calcium alginate	*Melaleuca alternifolia* (tea-tree)	nd	0.43 μm	Microcapsules	For maintaining uniform release behavior of oil[50]
13.	Calcium alginate	*Cinnamomum zeylanicum*	Commercial	nd	Beads	For maintaining the quality of northern snakehead fish fillets at refrigeration temperature (4 $\pm$ 1°C)[51]
14.	Calcium alginate	*Eucalyptus*	Leaves	0.5–2.5mm	Capsule	Controlled release[52]
15.	Sodium alginate, glutaraldehyde and PEG	*Azadirachta indica*	Commercial	1.28–1.49 mm	Beads	Controlled release[53]
16.	Chitosan	*Origanum vulgare* L.	Oregano Leaves	40–80 nm	Nanoparticles	*In-vitro* release showing initial burst effect and followed by a slow drug release [54]

(*Continued*)

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Table 1. (*Continued*)

S. No.	Polymer used	Source Plant of Essential Oils/ Constituents	Plant Part	Size of Formulation	Formulation type	Application
17.	Chitosan	*Zataria multiflora*	Aerial part	125–175nm	Nanoparticles	Enhanced antifungal activity for controlling *Botrytis cinerea* the causal agent of gray mould disease[55]
18.	Chitosan	*Cuminum cyminum*	Seeds	30–250 nm	Nanogel	Management of beetle[36]
19.	Chitosan (CH) and Cashew gum (CG)	*Lippia sidoides*	Commercial	1.27 and 1.53 mm of CH and CH-AG beads respectively (mean diameter)	Beads	*A. aegypti* larval control[56]
20.	Chitosan and Cashew gum	*Lippia sidoides*	Commercial	335–558 nm	Nanogel	Oil loaded samples presenting more effective larvicidal efficacies than the free *L. sidoides* oil[1]
21.	Chitosan and benzoic acid	*Thymus vulgaris*	Commercial	<100 nm	Nanogel	Enhanced antimicrobial activity against *Aspergillus flavus*[57]
22. acid	Chitosan and caffeic *Cuminum cyminum*	Commercial	<100 nm	Nanogel	Enhanced antimicrobial activity[35]	
23.	Chitosan and *k*-carrageenan	*Pimenta dioica* L.	Dried fruit	1.172–1.224mm	Microcapsules	Antimicrobial activity against *Candida utilis Bacillus cereus* and *B. subtilis*[58]
24.	Gum arabic and maltodextrin	*Schinu smolle*	Leaf and fruit	10 – 40 μm.	Microparticles	Insecticidal potential[59]
25.	Gum arabic	*Cymbopogon citrates*	Leaves	1.03–2780 nm	Microcapsules	Effect of modifications on encapsulation efficiency of the EO[60]
26.	Gum arabic	Limonene	Commercial	13 and 27 μm	Microparticles	Food industry[39]
27.	Guar gum and gum arabic	*Mentha*	Commercial	12–14 μm.	Emulsion	Better flavor retention by irradiated guar gum and gum arabic microencapsulates[61]
28.	Cyclodextrins	Estragole, *Ocimum basilicum* var. *basilicum* and *Artemesia dracunculus*	Commercial	nd	Inclusion complex	Improvement in stability and antioxidant activity of EOs [62]
29.	Cyclodextrins and cross linked cyclodextrins	Linalool and camphor from *Lavandula angustifolia*	Flowers	<125 μm	Inclusions	Controlled release of the fragrant compounds by β-CD[63]
30.	β-CD	*Rosmarinus officinalis* and *Thymus vulgaris*	Dry plant materials	>1 μm	Microcapsules	Food preservation and packaging (fungicide)[64]
			Microcapsules	Food preservation and packaging (fungicide)[64]		

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31.	β-CD	Carvacrol	Commercial	0.441–0.899 μm	Inclusion complexes	Delivery systems for antibacterial and antioxidant application[65]
32.	β-CD	trans-cinnamaldehyde, eugenol, cinnamon bark, and *Syzygium aromaticum*	Commercial	0.860–2.006 μm	Inclusion complexes	Antimicrobial activity[66]
33.	Hydrogenated Phospholipon®80H, phosphatidylethanolamine, non-hydrogenated soy phosphatidylcholine and Lipoid S100	β-caryophyllene and eugenyl acetate and Syzygium *aromaticum*	Commercial	223–260nm	Liposomes	Liposomes protected eugenol from UV-induced degradation and increased stability of the EO[67]
34.	Hydrogenated Palm oil, Softisan154, sorbitol and polysorbate 80	*Nigella sativa* L.	Seeds	66–142 nm	Nanoparticles	Nanoparticles prepared from mixed lipids show low crystallanity which make these advantageous in comparison to solid lipid and liquid lipid mixture[68]
35.	L-α-phoshatidylcholine and cholesterol	*Origanum dictamnus* L.	Aerial parts	nd	Liposomes	Antimicrobial activity[69]
36.	Dipalmitoyl-phosphatidylcholine (DPPC)	*Zanthoxylum tinguassuiba*	Leaves	9.37 $\pm$ 4.69 μm (mean diameter)	Liposomes	Incomplete release of EO from loaded in liposomes leading to targeted release for application in pharmaceuticals[70]
37.	Egg yolk phosphatidylcholine, dipalmitoylphosphatidyl choline, dimiristroylphosphatidyl choline, dioleoylphosphatidyl choline and dimiristroylphosphatidyl glycerol	*Anethum graveolens*	nd	70 – 457 nm	Liposomes	To improve stability and their bioavailability of EO[71]
38.	Enriched and hydrogenated soy phosphatidylcholine, cholesterol, stearylamine and polyoxyethylene lauryl ether	*Artemisia arborescens* L.	Aerial parts	78–457 nm	Liposomal vesicles	Antiviral activity[72]
39.	Lipids	*Atractylodes macrocephala* Koidz	Flowers and leaves	173 nm	Liposomes	Entrapping of EO by modified RESS method[73]
40.	Soy lecithin, modified starch	Terpenes mixture and d-limonene (from *Melaleuca alternifolia*)	Commercial	75–175 nm	Nanoemulsions	Enhance antimicrobial activity[74]
41.	Soybean lecithin, n-octenyl succinic anhydride (OSA) modified starch and poly-caprolactone	*Lavandula hybrida* super	Commercial	14–113μm	Microparticles	Antimicrobial activity of against three pathogenic food-borne bacteria[3]
42.	Compritol 888ATO (glyceride mixture)	*Artemisia arborescens* L.	Leaves	199–294 nm	Nanoparticles	For improving physical stability[75]

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Table 1. (*Continued*)

S. No.	Polymer used	Source Plant of Essential Oils/ Constituents	Plant Part	Size of Formulation	Formulation type	Application
43.	Gelatin, sucrose and inulin	*Origanum vulgare* L.	Flowers and leaves	nd	microparticles	Retention, Protection and *in vitro* delivery[76]
44.	Gelatin and gum arabic	*Chrysopogon*	Commercial	19–142μm	Microparticle	Cross-linking dependent controlled release of EO [77]
45.	Polyoxyethylesorbitan mono-laurate	*trans*-cinnamaldehyde	Commercial	10–200nm	Nanoemulsions	For enhancing Antimicrobial Activity[78]
46.	Sodium caseinate and lactose	*Salvia hispanica* L.	Commercial	0.24–0.54 μm.	Microencapsule	Physical Stability[79]
47.	Calcium caseinate and whey protein	*Origanum, Pimenta*	Commercial	nd	Edible films	Preservation of whole beef muscle[80]
48.	Whey protein concentrate, skimmed milk powder and their mixture with maltodextrins	*Carum carvi* L.	Commercial	nd	Matrices	Flavor retention[81]
49.	Whey protein concentrate	Limonene	Commercial	170 and 0.4 μm	Microparticles	Food industry[39]
50.	Poly ethylene glycol (PEG 6000)	*Mentha piperata*	Commercial	226–331 nm	Nanoparticles	Housefly control[33]
51.	Poly ethylene glycol (PEG)	Geranium and bergamot	Commercial	184–618 nm	Nanoparticles	Pest control[82]
52.	Poly vinyl alcohol (PVA) crosslinked with glutaraldehyde	*Cymbopogon citrates*	Leaves	10–250 μm	Microparticles	Antimicrobial activity[83]
53.	Span 80 and Tween 80	*Eucalyptus*, Linalool and Marjoram	Commercial	3–8 μm	Emulsions	Antifungal[84]
54.	Poly (DL-lactide-co-glycolide) (PLGA)	trans-cinnamaldehyde and eugenol	Commercial	14.2–317 nm	Nanoparticles	Antimicrobial[85]
55.	Polyurea	*Melissa officinalis* L. *Lavandula angustifolia* Miller *Salvia officinalis* L and *Thymus vulgaris* L.	Commercial	1–100 μm	Microcapsules	Antigerminative activity[86]

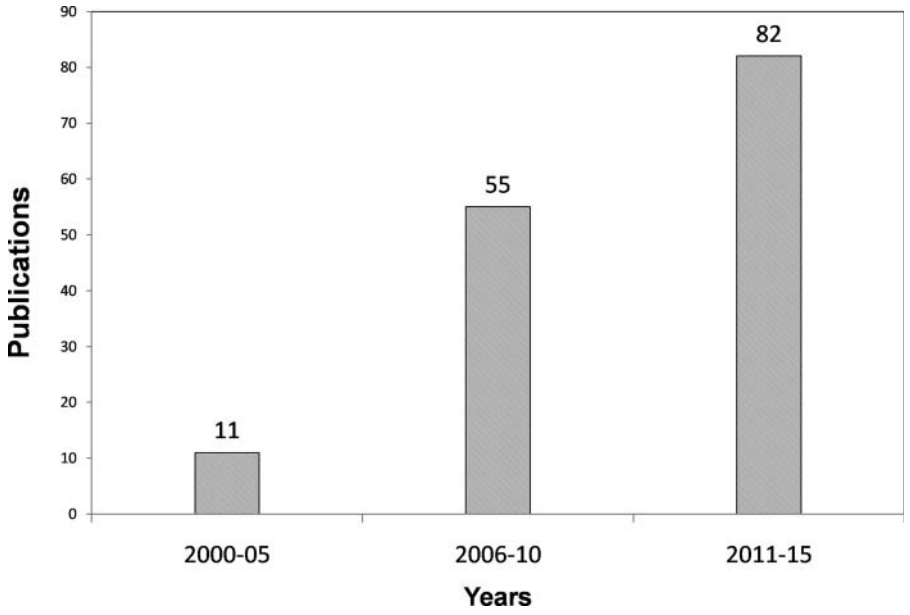

Figure 1. Publications in last 15 years in the field of EOs encapsulation.

term "polymer" has been used as a synonym for all biomaterials in this article. These have been classified on the basis of their origin (Fig. 2) and chemical composition (Table 2). Some of these are used directly in the formulations while the others require some chemical modification or processing to alter the polarity, stability, porosity, permeability, etc. The selection of the polymers depends on various parameters like their applicability, safety, biocompatibility, cost and availability. The major salient features considered for the selection of polymers for encapsulation have been compiled in Table 3. In addition to the polymers enlisted in Table 2, the chemical compositions of some polymers also vary on the basis of their sources, especially in case of gums and proteins.[89]

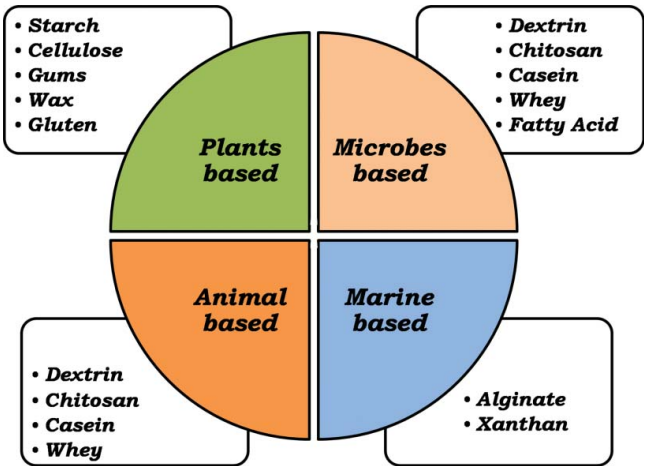

Figure 2. Classification of Biomaterials/Polymers used in EO encapsulate formulations.

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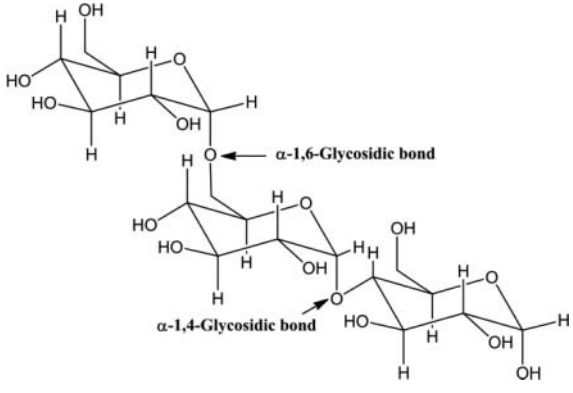
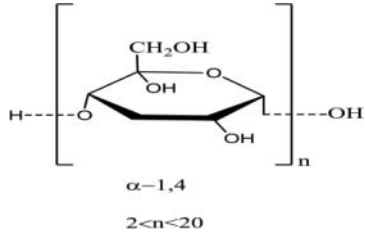
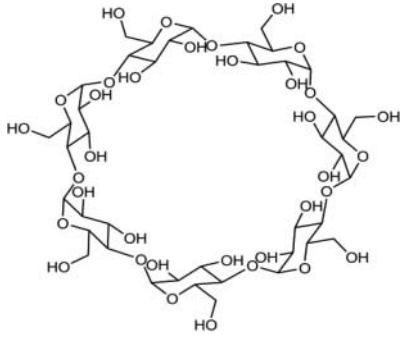
Table 2. Chemical and structural features of various polymers used in EO encapsulation.[111,112,153–155]

	Class	Name of Biomaterial	Monomeric unit	Structural formula
1.	**Carbohydrate**	Starch	D-glucose linked by α-(1→4) or α-(1→6) glycosidic bonds	
2.		Maltodextrins	D-glucose units linked by α-(1→4)	
3.		β-CD	7- glucopyranoside unit	

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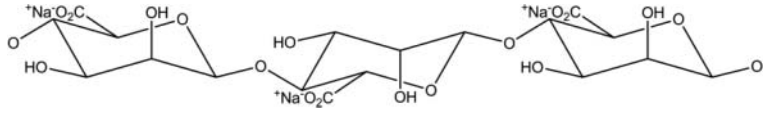
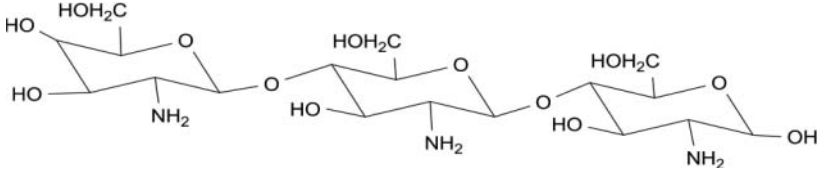
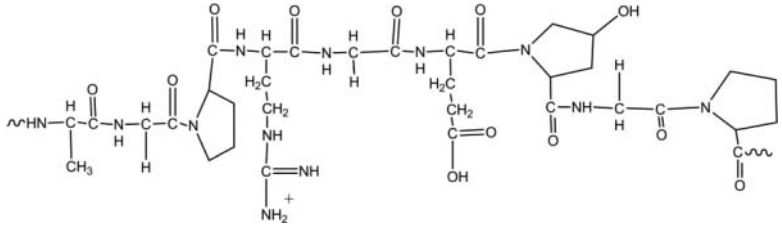
5.		Alginate	(1–4)-linked β-D-mannuronate (M) and its C-5 epimer α-L-guluronate (G) residues	
6.		Chitosan	Modified Chitin	
7.	**Protein**	Gelatin	amide condensation of α-amino acids (Glycine, proline and 4-hydroxyproline)	

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Table 2. (*Continued*)

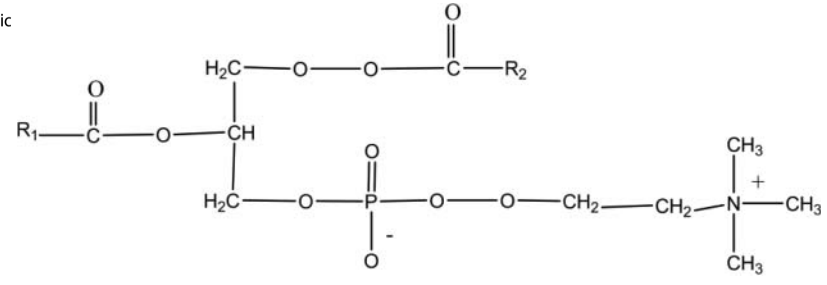
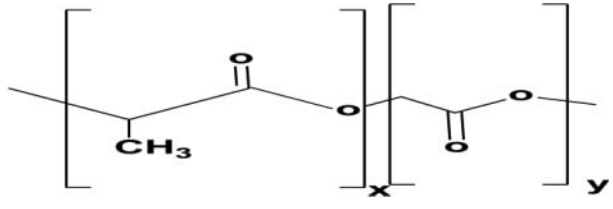
	Class	Name of Biomaterial	Monomeric unit	Structural formula
8.	**Lipids**	Phosphatidyl choline	Choline and glycerophosphoric acid	
		PLGA	Poly(D,L-lactide-co-glycolide)	

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Table 3. Salient features of different polymers for EOs encapsulations.

S. No	Biomaterial	Starch and Its Derivatives			Gums Gum arabic	Chitosan	Alginate Sodium/ calcium alginate	Lipids Phosphatidyl choline, PLGA and lecithin	Proteins Soybean, whey protein and gelatin
		Starch	Maltodexrins	β-CD					
1.	Chemical class	Polysaccharide	Polysaccharide	Cyclic			oligosaccharide	Complex mixture of glycoproteins and polysaccharides	Amine derivative of polysaccharide
	Carboxylic acid derivative of polysaccharide	Lipids	Proteins						
2.	Biodegradability	√	√	√	√	√	√	√	√
3.	Cost	Low	Low	High	Moderate	Moderate	Moderate	High	Moderate
4.	Water Solubility	Above 50°C	Low	Above 50°C	√	Low pH	At 40–50°C	×	√
5.	Porosity	Vary with source	Less	√	√	√	Regulated by crosslinking	—	√
6.	Presence of Permeability	Vary with source	Less	√	Easy permeable	—	Regulated by crosslinking	√	√
7.	Inclusion/gel/film Forming Capacity	After modification	After modification	√	Gel can be formed	Film/gel	Beads/gel	Liposomes/ micelles	√ Gel/film
8.	Pre-organized and Complementary cavity for EOs encapsulation	Generated by mixing and modification	Generated by mixing and modification	√ (Inclusion complex)	Generated by mixing and modification	Modification required	Generated by crosslinking	Generated by micelles formation	Generated by crosslinking/ mixing

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2.1 *Starch and its derivatives*

Starch is a readily available polymer of plant origin commonly used for encapsulation process. Schreiber et al. used starch for the first time for the encapsulation of pesticides.[90] Krishnan et al. used starch in the encapsulation of flavoring agents and EOs.[91] Currently, modified forms of starch are extensively used in the encapsulation. The aim of starch modification is to alter the structure and affect the hydrogen bonding in order to enhance its applicability in the encapsulation.[92] The modification also alters the DE (Dextrose Equivalent value) of products, which represent the degree of hydrolysis, this in-turn regulates the retention capacity of volatile compounds. Maltodextrins, cyclodextrins, and dextrin whites (DE value 20 – 100) are the major hydrolysates derived using acid treatment or enzyme modification of the starch and have been widely used for the encapsulation of EOs and other flavoring agents.[93,94] Among these, β-cyclodextrins (β-CD) have seven glucopyranose units which form a trapezoidal cone with internal cavity of 0.6 nm (diameter).[38] This cavity is hydrophobic and tends to form inclusion complexes with aromatic moieties of compounds like benzene, tyrosine, phenylalanine, etc.[38] These salient features of β-CD have been reviewed extensively for the application of inclusion complex formation with EOs/its constituents by Marques.[38] The process involves the attraction of hydrophobic EO particles towards the lipophilic cavity of β-CD and removal of water molecule towards the surrounding aqueous medium. The lipophilic cavity of β-CD provides microenvironment to the non-polar EO constituents which facilitate the formation of inclusion complex (Fig. 3). Maltodextrin alone cannot be used as a protective matrix for EOs encapsulation due to low solubility and rapid solidifying/lack of emulsifying properties, hence used in combination with other polymers like gum arabic, chitosan, proteins, etc.[95,47]

2.2 *Gums*

Gums are the natural products obtained from *Acacia* and guar beans. The gum powders have good emulsifying properties, low viscosity, high retention capacity, and solubility in cold and hot water.[60,91] They are excellent material for the encapsulation of different natural

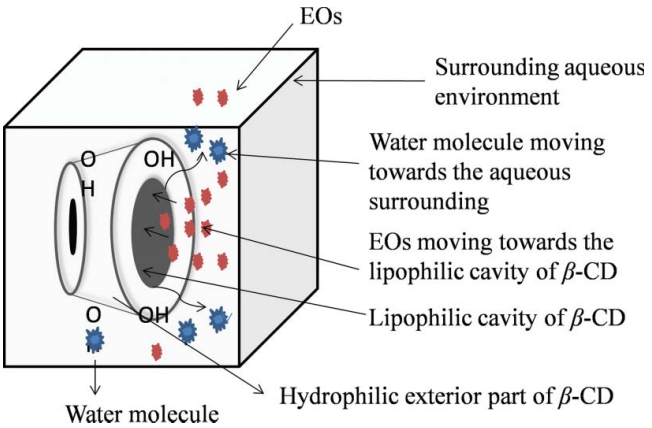

Figure 3. Diagrammatic representation of EO-β-CD inclusion complex formation. Ability of β-CD to form an inclusion complex with EOs, the process takes place (1) By movement of polar water molecules towards surrounding aqueous environment and nonpolar EOs molecule in the lipophilic cavity, and (2) mixing the hydrophilic exteriors with surrounding aqueous environment.

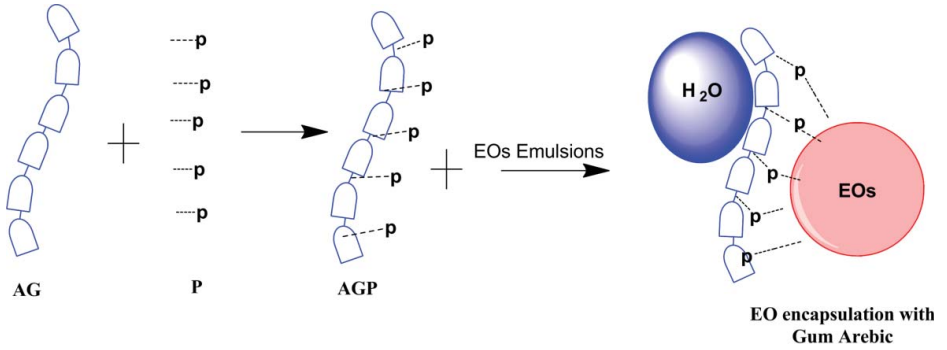

Figure 4. Schematic representation of EOs emulsions with gum arabic. The major fraction (88-90%) of Gum Arabic consists of highly of branched polysaccharides containing the units of rhamnose, arabinose and glucuronic acid plus low protein content, it also associated with minor fraction (10%) of the protein collectively called Arabinogalactan-Protein (AGP). During the encapsulation of EO the ambiphilic protein part attached with the EO and the hydrophilic carbohydrate part phases towards the aqueous phase.

products like phenolics, alkaloids, fish oils, vitamins, EOs, etc.[61,91,96–100] The semipermeable nature of gums make them porous and unstable. Hence, to prolong their stability, these are mixed with others polymers (mainly maltodextrins and chitosan) during the encapsulation process. Consequently, the mixture of gum and other polymers form a thin film around the EO droplet and protect it from rapid evaporation and particle aggregation due to humidity (Fig. 4).[77,95,98,101]

2.3 *Chitin*

Chitin is the second most abundant polymer in nature after cellulose. It is the principal part of exoskeleton of crustaceans such as crabs, shrimps, prawns, lobsters, and cell walls of some fungi such as *Aspergillus, Zygomicetes* and *Mucor*.[1,102] For the encapsulation of EOs, chitosan (a deacetylated derivative of chitin) (Fig. 5) was used with the help of cross-linking agents. Hsieh et al. developed chitosan encapsulated volatile citronella oil microcapsules for their

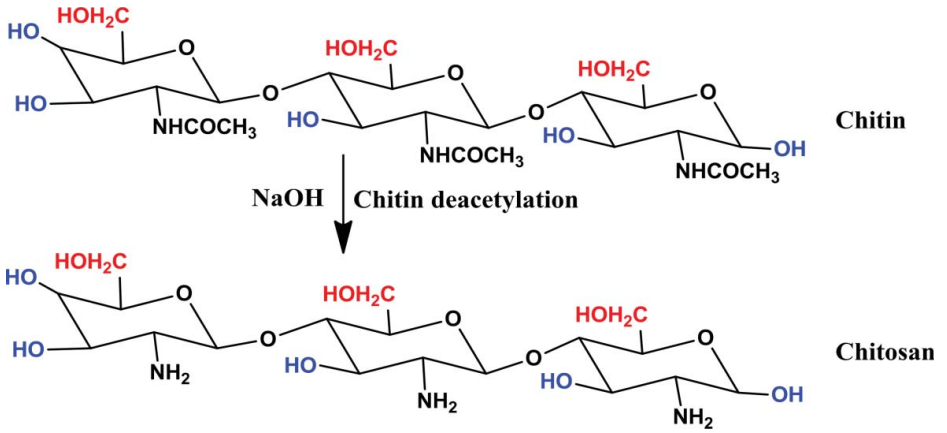

Figure 5. Conversion of chitin into chitosan via alkaline deacetylation.

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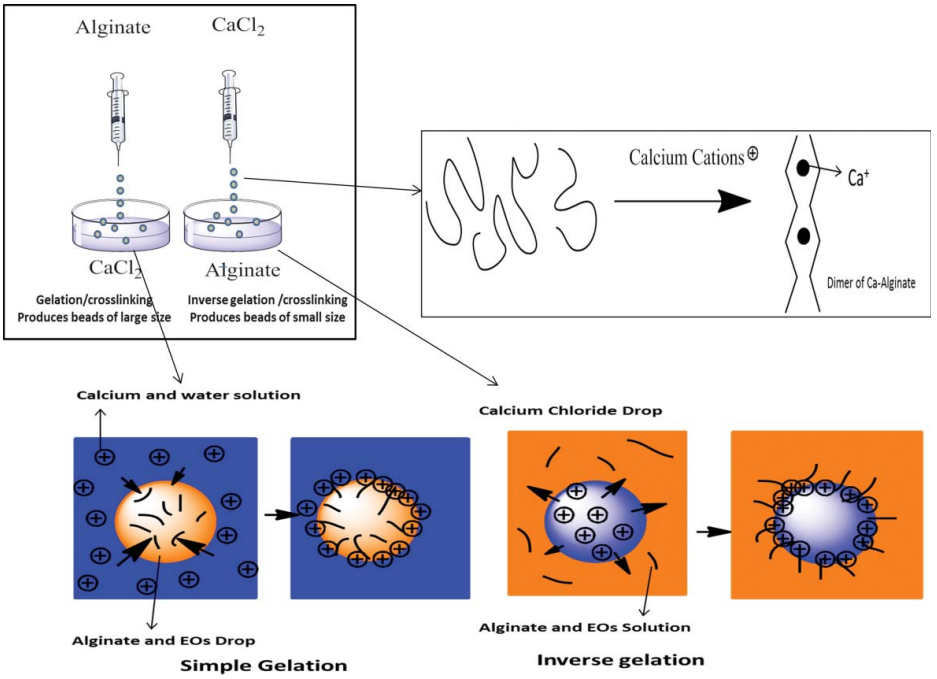

Figure 6. Gelation and inverse gelation process for EO encapsulated Ca-Alginate beads.

sustained release.[103] Similarly, Hosseini et al. used chitosan in the development of EO containing nanoparticles for the *in vitro* release of EO.[54] Further, the use of chitosan was also reported in the EO encapsulation with the combination of various substances like cashew gum, alginate, cinnamic acid, carrageenan, and benzoic acid.[36,56–58,104,105]

2.4 *Lipid colloids*

Lipids are ambiphilic compounds with hydrophilic head (phosphate and diglyceride group) and a hydrophobic tail (long fatty acid), which spontaneously assemble to form colloids of different structures like liposomes, micelles, bilayer, or n-lamellar sheets. On the basis of their lamellarity and size, liposomes are classified as small uni-lamellar vesicles (SUV) (size between 20–100 nm), large uni-lamellar vesicles (LUV) (size > 100 nm) and large multi-lamellar vesicles (MLV) (size > 0.5 mm).[106–108] In case of EOs encapsulation in lipid colloids, it was observed that EO components present inside the liposomes decrease the size of the colloid due to the intermolecular Van der Waals interaction between the non-polar chains of lipids and EO in the membrane vesicles.[109] Ortan et al. compared the influence of lipid composition on entrapment efficiency using number of phospholipids Phosphatidylcholine (PC) from fresh egg yolk, dipalmitoylphosphatidyl choline (DPPC), dimiristroylphosphatidyl choline (DMPC), dioleoylphosphatidyl choline (DOPC), and dimiristroylphosphatidyl glycerol (DMPG)) having different physical and chemical characteristics for encapsulation of *Anethi fructus* EO and concluded that the encapsulation efficiency, stability, and release rate of active ingredient is governed by the type of lipids used and their molar ratios with EOs in

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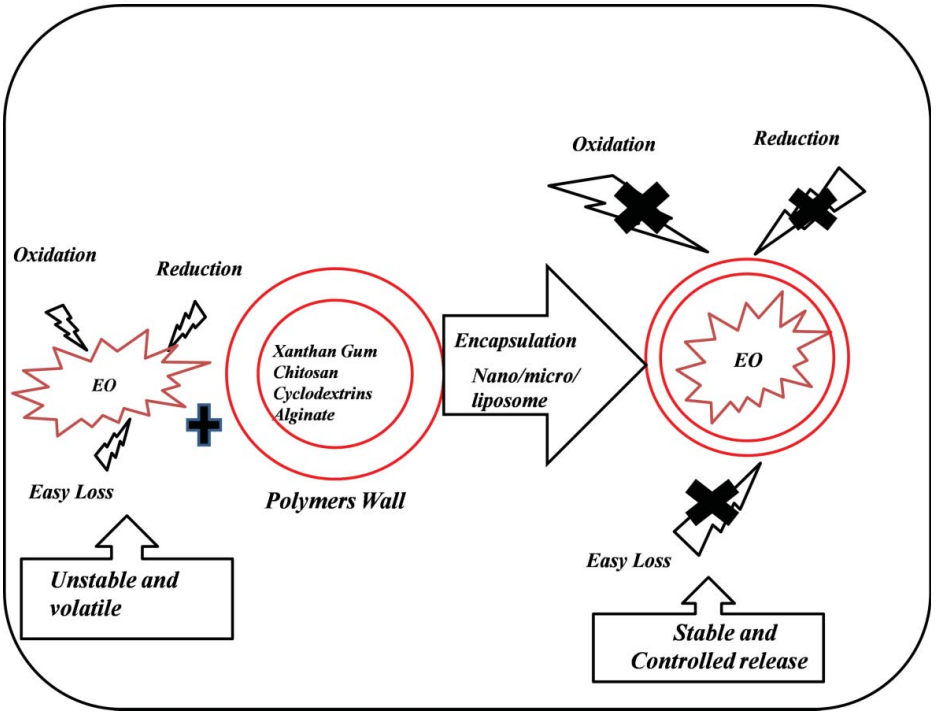

Figure 7. Protection of EOs encapsulation prevents its easy loss, oxidation and reduction in open conditions.

the preparation of the liposome.[71] Similarly, ALHaj et al. reported the increase in stability of *N. sativa* L. EOs when incorporated in Solid Lipid Nanoparticle (SLN) formulation using hydrogenated palm oil Softisan 154 and EOs as lipid matrix, sorbitol and water as surfactants.[68]

2.5 *Alginate*

Alginate is an anionic biopolymer produced by marine algae and is used in the preparation of nanocapsules, beads, and nanoparticles due to its sensible biocompatibility, biodegradability, non-toxicity, mucoadhesion, gelation, and film formation properties.[110] Alginate consists of linear chains of α-L-guluronic acid (G) and β-D-mannuronic acid (M) residues joined by 1, 4-glycosidic linkages.[111] The alginate polymer features a phenomenon of cross linking of the polymer in presence of divalent metal ions. Thus, soluble sodium alginate upon treatment with Ca^{2+} ion leads to the formation of cross-linked polymeric calcium alginate beads (Fig. 6). The size distribution of the beads depends on the concentrations of sodium alginate and calcium chloride used during the cross-linking.[112] The process of inverse crosslinking/gelation lead to formation of smaller sized beads as compared to simple gelation.[87]

2.1.6 *Other polymers*

Some other polymers like inuline, gelatin, k-carrageenan, polylactide acide (PLD) poly(lactic-co-glycolic acid) (PLGA), etc., have also been reported for EOs encapsulation.[113] Among

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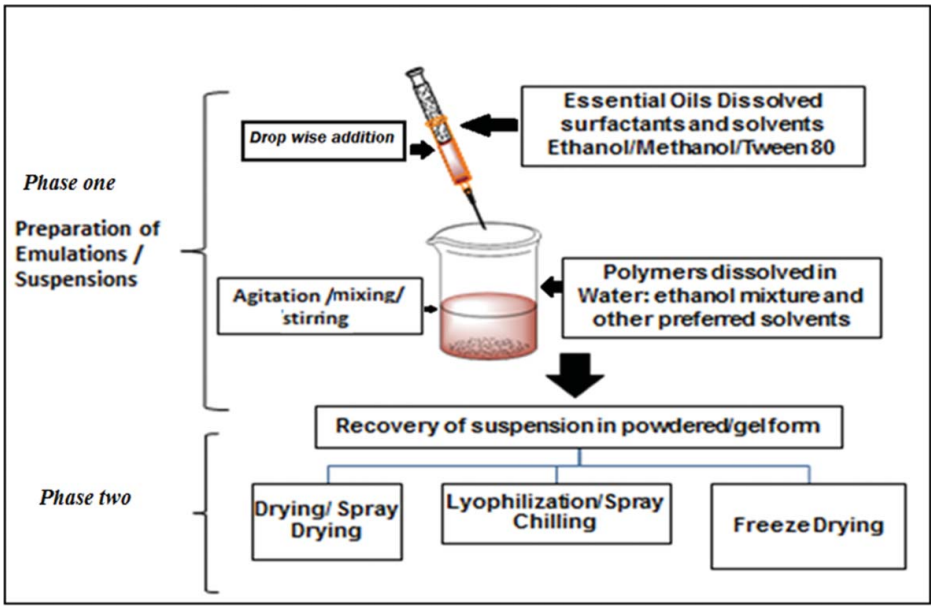

Figure 8. Schematic representation of the EO encapsulation process.

these, the inuline, gelatin, and k-carrageenan have been used in combination with gums, chitosan, starch, and gelatin, while the PLGA and PLD were used solely for the encapsulation of EOs and their constituents.[113–115]

3. Techniques used for encapsulation of EOs

In general, encapsulation is a method by which one material or a mixture of materials is coated with or entrapped within another material or system. The coated material is called active or core material, and the coating material is called a shell or wall material, and the combination of both is known as carrier or encapsulate. Sometimes, EOs-polymer complex are known as host-guest complex where polymers act as host and EOs as guest. Historically, encapsulation is traced back to the 1950s wherein pressure-sensitive coatings for the manufacture of carbonless copying paper were introduced by Green and Lowell, 1960.[116] Encapsulation is now a well-developed and accepted technology with application in pharmaceutical, chemical, cosmetic, food, and printing industries.[29,41,77,94,95,99,117–120] Technologies used in EO encapsulation are based on three basic principles:
(1) Formation of a wall around the material to be encapsulated; (2) ensuring sustained release of EO; (3) the enhanced life of the encapsulates.[99]

Figure 7 represents how the encapsulation process protects the EOs from release under natural conditions by covering it in a polymer wall. The encapsulation process proceeds in two phases: the first phase involves the formulation of EO-polymer suspensions and second phase involves product recovery as represented in Fig. 8. The common approaches used in EO-polymer suspension formation are inclusion complexation, cross-linking, coacervation and melt dispersion. The stepwise process is described in Table 4. Further, these approaches require different set of techniques for the recovery of encapsulates in the powdered form. To

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Table 4. General steps involved in different formulation techniques for EO encapsulations.

S.No	Techniques and preferred polymers	Basic Parameter	General Steps	Reference
1.	Inclusion formation or co- precipitations with β-CD	***Inclusion formation or co-precipitations conditions*** Temperature for solubilization of β-CD is 55±2°C. Temperature for Inclusion formation or co-precipitations is 25–30°C with continuous stirring.	1. β-CD is dissolved in ethanol/water (1:2) mixture at 50–55± 2°C. 2. A predetermined quantity of EOs dissolved in ethanol then slowly added to the warm β-CD solution. 3. The mixture is continuously stirred on the magnetic stirrer at 55°C. 4. The mixture is stirred for another 4 h without heating till temp. decrease to 25°C. 5. The final solution is refrigerated overnight at 7°C. 6. The cold precipitated material is recovered using vacuum filtration. 7. The precipitate is dried in a convection oven at 50°C for 24 h. 8. The powder is then allowed to air-dry at 25°C for an additional 24 h.	123, 127
2.	**Coacervation/ Crosslinking Polymer: Gelatin, Chitosan, Cashew, Alginate**	***Coacervation*** Variation in coacervation process arises due to separating or crosslinking or solidifying agents [Glutaraldehyde, calcium chloride, poly ethyl glycol (PEG) etc] and slight modification in pH, ionic strength and poly ion concentration of medium.	7. Preparation of Polymer Solution in Water (Temp 40–60°C depends on the polymers) with continuous stirring 2. EOs dispersion on the polymer solution 3. Mixing of phase separating or crosslinking or solidifying agents 4. Then, filtration and drying the materials.	23,36,60,77,101,128,130
3.	**Melt Dispersion** Poly ethyl glycol (PEG)	***Melt Dispersion*** Melting temp may be differ according to polymer used	PEG is heated separately at 65°C for melting after EOs were mixed with melted PEG and stirred lightly with a glass rod to ensure even distribution of the mixture. The mixture was ground completely in pestle mortar after being cooled naturally at 25°C and then sieved using a sieve mesh 200. The powders were placed in airtight, pouches and in desiccators.	131

Table 5. Techniques of product recovery after encapsulation.

Features	Spray drying	Freeze drying	Spray Chilling
Process	The mixture of EOs is atomized with a nozzle or spinning wheel. The contact between the drop and the hot air takes place during atomization. Rapid evaporation of solvent (usually water) maintains droplet temperature at low level and allows entrapping the active compound after entrapping microcapsules are then collected to the bottom of the drier.[92]	For this EOs and polymer suspensions agitated for 24 h (300 rpm at 25°C) and filtered to eliminate non-dissolved compounds. Then filtrates were first frozen then lyophilized at −85°C in freeze dryer for approximately 48 h or until all moisture had been sublimated.[62]	In Spray chilling and Spray Cooling methods, the coating materials is melted and atomized through a pneumatic nozzle. The melting material is sprayed into the vessel containing chilled CO_2.[132,133]
Temp of operation	100–190°C	−85°C	Chill temperature
Instrument	Spray Drier	Lyophilizer	Spray Chiller with Chilled CO_2 Vessels
Time(for 100 mL)	40–50min	40–50 min	1–2 days

achieve the solid product recovery of desired size (micro to nano dimension), techniques like spray drying, spray chilling and freeze drying are frequently used.[62,91,121,122] Table 5 provides insight of the methodology and the conditions involved in product recovery. Further, a comparative account of all the techniques involved in encapsulation and product recovery in terms of their operating cost, time duration, and suitability for EOs are mentioned in Table 6.

4. Physico-chemical methods for characterization of EO encapsulates

Physical, structural, and chemical characterizations of EOs encapsulated formulations are essential for the assessment of variables that affect the process optimization and product functionality. Optimization of the process depends on the involved methodology and ratio of polymers (wall material) to EOs. The functionality of developed EO formulation depends on its physical stability, morphology, EO loading, and its release characteristics. A better understanding of these parameters help in the reduction of formulation cost and

Table 6. Comparative analysis of different formulation technologies.

S.No	Name of the method	Operating Cost and maintenance of conditions	Product morphology	Time duration	Suitability for EOs
			Chemical Methods of Encapsulation		
1.	Coacervation	Moderate	Powder, gels, Capsules	Depends upon solubility of polymer	Less
2.	Co-crystallization	Low	Crystalline powder	Moderate	Moderate
3.	Molecular Inclusion	Low	Suspensions, droplets, powder, gel	Less	Suitable with non-polar oils
4.	Liposome	Depends upon the polymer used	Suspensions, droplets, gel	Depends upon solubility of polymer	Suitable
			Product Recovery Methods of Encapsulation Process		
4.	Spray Drying	Low as compared to freeze drying	Powder	Less	Less
5.	Freeze Drying	high	Powder	More	Less
6.	Spray Chilling and Spray Cooling	low	Powder	Less	Suitable

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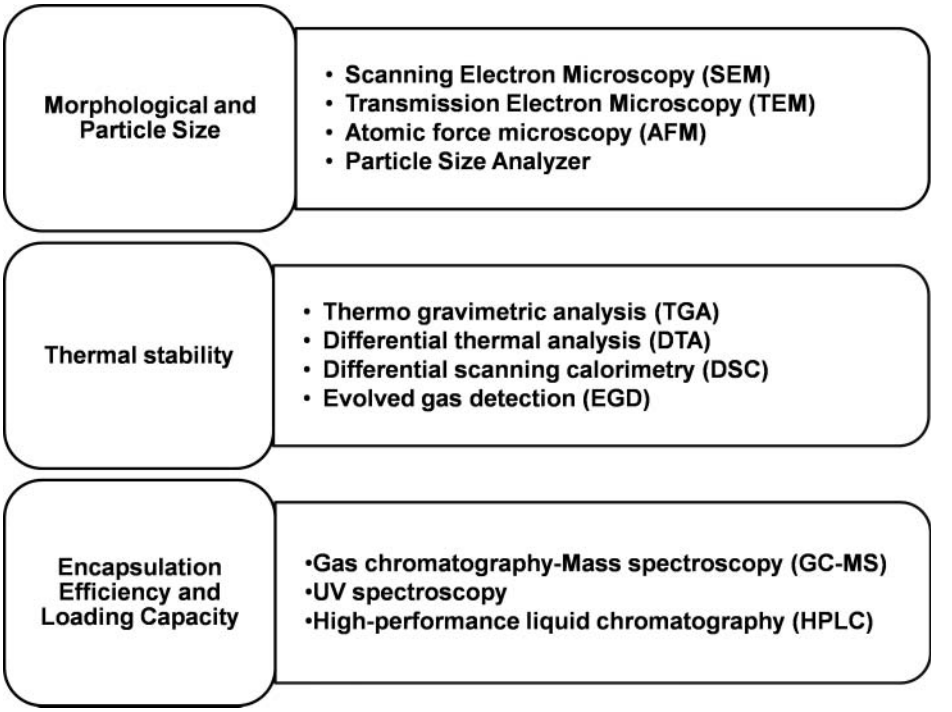

Figure 9. Techniques used in Physico chemical analysis of EO encapsulates.

development of durable and functionally active EO encapsulate. Therefore, a number of analytical and instrumental techniques are used in the physical, chemical, and structural characterization of EOs formulations (Fig. 9).

4.1 *Microscopic imaging*

In order to analyze physical appearance and morphology of EO encapsulates, microscopic techniques like SEM (Scanning Electron Microscopy), TEM (Transmission Electron Microscopy), OM (Optical Microscopy), and AFM (Atomic Force Microscopy) are widely used.[55,65,72,134] However, analysis of the particle size and surface charge of formulation usually carried out by dynamic laser scattering technique coupled with Zeta Potential analyzer.[30,33,55,134]

SEM analysis facilitates the study of surface morphology of the encapsulated material while TEM helps in the exploration of micro-structural features and thickness of wall material. The study of external surface provides information about the presence of cracks and pores that decide the permeability of the materials. A large number of pores and cracks on the surface of material lead to enhanced permeability which allows the easy loss and degradation of EOs.[46,131] SEM and TEM not only help in visualizing the porosity and cracks but also assess the shape, structural uniformity and aggregation/dispersion of encapsulated materials. The round shape, uniform size, and less aggregated encapsulates are reported to possess better stability, dispersion and less permeability of volatiles.[59,60,135] Yang et al. visualized Polyethylene glycol (PEG 6000) nanoparticles loaded with *Allium sativum* L. EOs using TEM reported well dispersed spherical particles of mean size of 233 ± 108 nm with good

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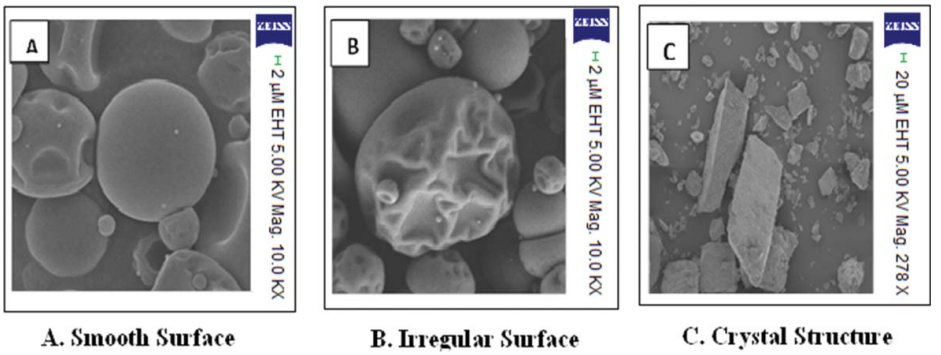

Figure 10. SEM images of EO encapsulate made using (A) Gum Arabic and Maltodextrin (B) Maltodextrins (C) β-cyclodextrins.

stability.[131] The external morphology of the EOs loaded nanoparticles presented no apparent cracks or porosity, indicating strong protection of the core material.[131] SEM imaging analysis of holy basil EO loaded gelatin microencapsulate, was spherical in appearance with folds and rough external surfaces at low magnification and sponge-like structure with a large number of micron-sized pores randomly distributed on the outer surface at higher magnifications.[23] Further, the internal cross-section of the EO holy basil loaded gelatin microencapsulates revealed honeycomb like porous structure. Similarly, SEM imaging of β-CD loaded with EOs of lamiaceae plants showed rhombohedral-parallelepipedic shaped crystalline nanoparticles.[32] Sinico et al. incorporated *Artemisia arborescens* L. EO in the liposomes and investigated its morphology by TEM, light polarization, and optical microscopy and observed multilamellar niosomes containing oil droplets.[72] The TEM images of carvacrol β-CD inclusion complexes showed agglomeration where larger particles appeared to attract smaller particles.[65] Another study of TEM imaging of empty and *Zataria multiflora* EO loaded chitosan

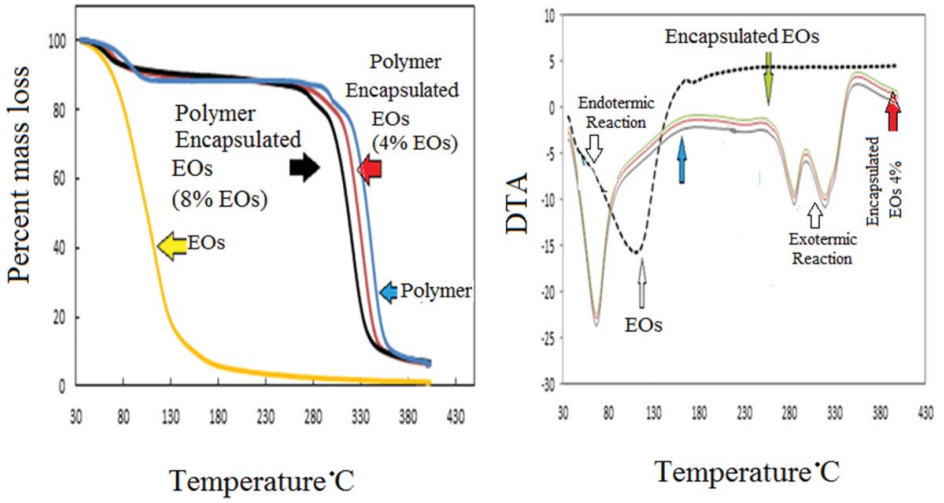

Figure 11. TGA/DTA Curves of EO and its encapsulate over the temp range of 30–450°C.

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nanoencapsules showed nearly spherical shape and smooth surface.[55] Further, the study concludes that loading of EO on chitosan nanoencapsules leads to increased particle size of the formulations.[55] Menezes et al.[124] studied the SEM images of the physical mixture, paste, and slurry of β-CD -() -linalool inclusion complex. The physical mixture showed no change in the particle shape; however, the paste and slurry complex showed a significant change in the particle shape with agglomeration due to inclusion complexation. Further, the particle size of the inclusion complex was observed to be smaller than that of the physical mixture, which could be attributed to the technique used in formation. Mehyar et al. studied the SEM images of microencapsules of cardamom EO with Whey protein isolate (WPI).[129] The significant increase in surface porosity and irregular shape was observed in the WPI-Cardamom EO microencapsule in the presence of Guar Gum (GG) or Carrageenan (CG). Similar study of morphological imaging using SEM variations in the EO loaded gum arabic maltodextrins microparticles and β-CD inclusions synthesized in our lab is shown in Fig. 10.

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4.2 *Thermal analytical characterization*

Thermal behavior is a critical parameter to define changes in the physical attributes like melting, evaporation, sublimation, decomposition, etc., of EOs, polymer used for encapsulation, and loaded encapsulates before and after the inclusion complex formation process. It also determines the retention/release properties and thermal stability of the EOs encapsulates. Thermal behavior of encapsulates is characterized using Thermo Gravimetric Analysis (TGA), Differential Thermal Analysis (DTA), and Differential Scanning Colorimetric (DSC) techniques. Thermo analytical methods are commonly preferred over the other techniques like X-ray powder diffraction, FT-IR spectroscopy, and solid-state NMR due to their relatively lower cost and ease of analysis.[125,136]

In the TGA/DTA analysis of the EO encapsulates, TGA curves enumerate the changes in thermal stability of guest and host molecules before and after encapsulation by the assessment of percentage weight loss over pre-defined temperature range. TGA also helps in quantitative determination of the EO in the loaded encapsulates. The DTA curve helps in the assessment of exothermic and endothermic processes taking place in a pre-defined temperature range. The weight loss and thermal degradation pattern of EO before and after encapsulation using TGA/DTA curves (temperature range 30–450°C) are shown in Fig. 11.

DSC is another technique to determine the nature of reactions (endothermic and exothermic) taking place during the thermal decomposition of materials and helps to investigate the nature of inclusion complex.[137] Fernandes et al. used another thermal technique named Evolved Gas Detection (EDG) to measure the release rate of EO vapors from the encapsulates at different temperature and time intervals.[138] Further, these thermo analytical techniques become more informative when coupled with Fourier transform infrared spectroscopy (FTIR) or mass spectrometry (MS) techniques.[127] Fernandes et al., in their study of *Lippia sidoides* EO and gum arabic/maltodextrin encapsulates, used Thermo Gravimetry coupled Mass Spectrometry (TG-MS) to directly confirm the formation of inclusion complex.[125,127] The studies established increased thermal stability of the micro encapsulates with an increase in the amount of gum arabic in carrier mixture. Further, TGA study of β-CD EO inclusion complex showed an increase in the temperature of volatilization of the EO constituents in the inclusion complex.[125,136]

Table 7. Physico-chemical methods for characterization of EO encapsulates.

S. No	Analytical Techniques	Use	Major steps involved in sample preparation Conditions /methodology		References
1.	SEM	Determination of morphology and cracks on surface	Specimen Coating	Most preferable Gold coating	36, 55, 65, 72, 131, 134
			Magnification	100X-200kX	
			EHT	5–20kV	
2.	TEM	For external pores and their adsorption characteristic	Specimen Coating	Platinum	
			EHT	80kV	
3.	DTA/TGA	Quantification of the loading of EO by evaporation based thermal release	Temp. Range	5–10°C/min 30°C to 400–600°C. depends on the Polymers	30, 54, 77, 124, 130, 142, 143
			Pan	Aluminum	
			Atmosphere	N_2	
4.	FTIR	For interaction at molecular level based on functional group	Spectra range 4000–400 cm^{-1} Using KBr pelletas standard.	1, 104, 127, 128, 144	
6.	GCMS	Estimation of individual constituents of the EO adsorbed	Colum	DB5, Fused Silica	31, 46, 127, 138
			Temp	60 – 280°C	
			Detector	Flame Ionization Detector	
			Split Ratio	1:5,1:50,1:100	
			Carrier Gas	Helium (He)	

4.3 *Techniques for encapsulation efficiency and loading capacity*

Techniques for chemical characterization of the formulation involve determination of encapsulation efficiency and loading capacity which in turn help in confirming the amount of loaded EO inside the encapsulates. The percent encapsulation efficiency is defined as the ratio of mass of loaded EO encapsulate to the initial mass of EO used in the encapsulation process and the loading capacity is calculated as the ratio of weight of loaded EO and mass of prepared formulation.[55] Simple techniques involved in the gravimetric measurement of loaded EOs are distillation process using Clevenger type apparatus,[60] Soxhlet extraction system,[135] inverse dialysis technique,[139] etc. However, these methods have disadvantages like requirement of large amount of formulation, improper extraction, and chances of loss of EOs due to volatilization.

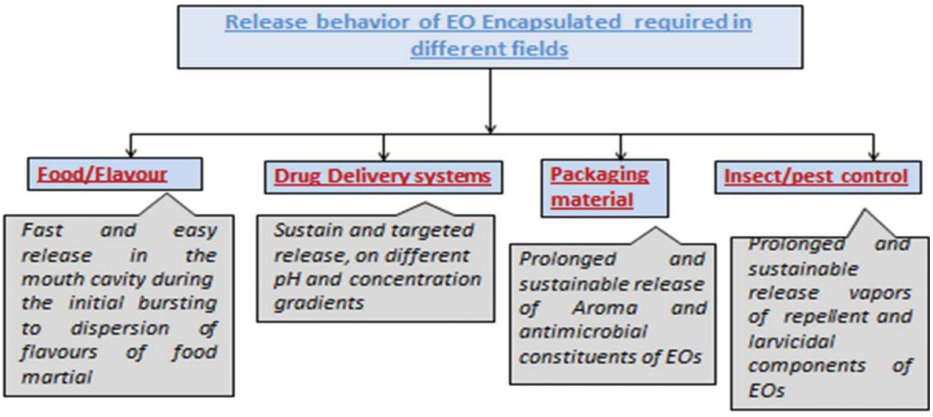

Figure 12. Flow chart for application based desirable EOs release characteristics.

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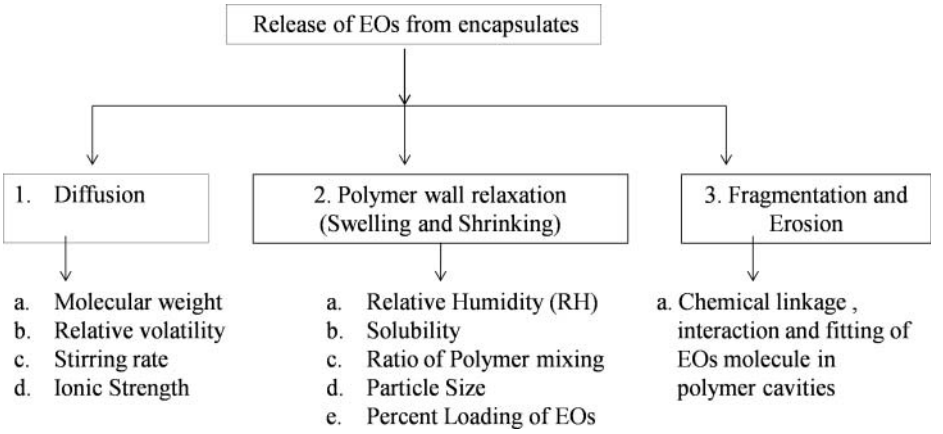

Figure 13. Flow diagram representing release of EOs from encapsulates: Mechanism and factors affecting the process.

Hence, sophisticated techniques like UV spectroscopy and GC-MS are used extensively to determine the amount of loaded EOs in formulation. For both the methods, the two main steps required are
(i) extraction of EO from the encapsulated sample by its lysis/partitioning using solvents like ethanol, methanol, n-hexane, dichloromethane, acetonitrile, etc., (ii) preparation of calibration curve of known amount of oil within expected range.[23,55,59,124,130,134]

In case of UV spectrophotometric analysis, the wavelength for EO is decided through the spectral scanning which usually falls in the range of 260–320 nm.[33,72,123,131] Similarly, the EO quantification with GC-MS also requires the preparation of calibration curve, by taking the retention time, peak area, and intensity of its major components.[41,140] This method can be hyphenated with the time-efficient method like head space–solid phase microextraction technique (HS-SPME).[46,124,141]

4.4 *Other techniques*

On the basis of the chemical nature of the polymers and core material used (EOs or its constituents), some other analytical techniques like FTIR, NMR, XRD, etc., have also been reported for the physical and structural analysis of encapsulates. FTIR facilitates the investigation of possible interactions between oil and polymers by analysis of broadened and shifted peaks due to the changes arising during the complex formation.[23,57,104] The changes in the crystallographic arrangement of EO loaded nanoencapsules are studied by the XRD analysis.[54,127,128]

Physico-chemical characterization of encapsulates require standardized protocols and conditions that depend on the nature and chemical properties of the polymers and core material use. The standardized conditions and protocols used by different researchers are described in Table 7.

5. Release characteristics of EOs encapsulates

The desired release rate of EOs from the encapsulates varies according to its field of application. EO constituents, which are used as flavoring agents, require fast release at the time of

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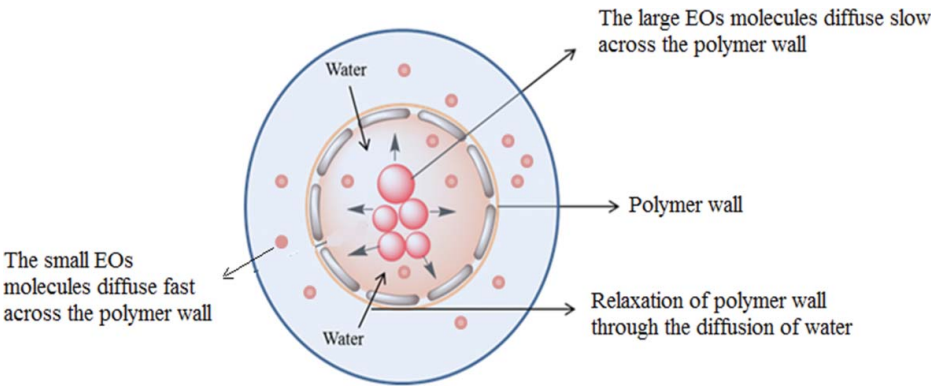

Figure 14. Diffusion and polymer relaxation based mechanism of EOs release.

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contact with the saliva or mouth microenvironment[92,94] while the EOs used in the drug delivery systems may need to be designed for sustained and targeted release.[29] On the other hand, the EO related formulations used in the food packaging and insect pest control require prolonged and sustainable release to attain higher stability under natural conditions (Fig. 12). The efficacy and sustainability of EO encapsulates depend on the releasing rate of EOs from the encapsulating systems.[41,49,54,122,140] The release of EO from various EOs encapsulates are regulated by different alternate mechanisms like diffusion, polymer relaxation (swelling/shrinking), erosion, fragmentation, etc.[88] Further, these mechanisms depend on the properties of encapsulated EO, polymer used, physico-chemical characteristics of encapsulates (size of particle and its internal cavity, interaction between EOs and cavity sites), and surrounding environmental conditions (Relative humidity and Temperature). Nuchuchua et al. correlated the computational and spectroscopic studies of inclusion complex of eugenol with α-CD, β-CD and HP-β-CD (2-hydroxypropyl-β-cyclodextrin) to find the relation between size of EOs constituent and size of internal cavity of encapsulate.[145] It was observed that eugenol released rapidly from α-CD complex due to its small size of internal cavity whereas the release was slow in case of β-CD and HP-β-CD complex due to the proper fitting and bonding of eugenol with the sites present in their internal cavities.[145] Similarly, Marques et al. suggested that the encapsulation and release of volatile may depend on the size and the interaction sites present in the cavity of the different CDs.[38] The various processes involved in release of EOs are discussed in further sections and a general mechanism of release is shown in Fig. 13.

5.1 *Diffusion*

Diffusion is a process that allows the movement of the molecules with concentration gradient, i.e., from high concentration to lower concentration. In case of EO encapsulates, diffusion rate depends on the pore size of polymer, thickness of wall and size of the EO constituents.[146] The study of Voilley on isoamyl butyrate (MW 158) and ethyl butyrate (MW 116) suggests that the molecules with high molecular weight were released slowly and retained for longer time inside the polymer wall of glucose, maltose, and corn syrup.[146] Similarly, the study of Jafari et al. also reveals that the EO constituents with low molecular weight and smaller size diffuse faster as compared to the molecules having high molecular weight and large size.[147]

Fig. 14 depicts the general mechanism of EOs diffusion from the encapsulates. Further, the diffusion rate also depends on the relative volatility of EOs. The volatility of the EO constituents with respect to water is considered the relative volatility of that constituents.[132,148] The study by Krishnan et al. suggested an inverse relation between the relative volatility and retention of the EOs molecule, i.e., higher the relative volatility lower the retention.[98]

The diffusion rate of EO through the polymer wall of encapsulate can be regulated by altering the encapsulate preparation parameters like stirring speed, polymer to EO ratio and ionic strength. Leimann et al. studied the effect of stirring rate (500–900 rpm) during formulation preparation on microcapsule morphology and EO release and suggested that the increase in the stirring rate leads to the formation of particles with smaller size and uneven distribution but it does not significantly affect the EOs release.[83] Heating of the encapsulated formulations has been evaluated to positively affect the release of the EOs during their application, but it depends on the type of method and polymer used, size distribution characteristics of the encapsulate. Yeo et al. suggested that the conditions for forming homogenized microparticles need to be optimized for sustained release upon heating.[116] In their study, the heating release rate of EOs from microencapsulates prepared using mixture of gum arabic and gelatin was observed to depend on the distribution characteristics of the encapsulates. The microencapsules with homogeneous morphology and smaller size had a greater EOs release in response to heating as compared to large size agglomerated microencapsules.[149] The possible reason for the same is that smaller microencapsules have single large cavity with thin coating for EOs while large micro encapsules have numerous small cavities with comparatively thick coating for EOs.

5.2 *Swelling and shrinking properties of polymers*

Release of EO is also regulated by polymer relaxation or bursting. During the release of EO from the encapsulate, the initial rate of release is high which becomes constant after sometime.[54] Initial higher rate of release may be attributed to diffusion and dispersion of water inside polymer wall which leads to relaxation due to swelling and finally bursting of the encapsulates. The swelling and relaxation of polymers is governed by the relative humidity, temperature of surrounding/storage, solubility, and size of encapsulates, etc. Whorton and Reneccius, in their study proved that the water adsorption on the wall material accelerates the hydration process which leads to the cracks on the wall material which subsequently triggered the release of volatiles.[150] However, according to Choi et al. the hydration mediated swelling and shrinking is comparatively rare phenomena in encapsulates prepared from rigid and water insoluble polymers.[151] The release rate of encapsulates formulated using polymers which absorb moisture can be better understood from the studies of Ponce Cevallos et al.,[152] Ayala-Zavala et al.,[153] and Del-Toro et al.[154] Ponce Cevallos et al. in their study examined the effect of relative humidity (RH) on release of EOs from encapsulates formulated using β-CD, and observed abrupt release of EOs constituents at RH higher than 84% due to higher water sorption by encapsulate.[152] Similarly, Del-Toro et al. reported 76% release of EOs at 97% RH while working on thyme EOs and β-CD capsules.[154] From the studies, it can be concluded that different types of encapsulates formed respond differently on increasing the RH% which depends on the characteristics of EOs, polymer and EOs and polymer interaction. The bursting of encapsulates also depends on its size. The studies by Hosseini et al.[54] and Luo et al.[105] involving EOs and chitosan encapsulates concluded that the release of EOs was higher in smaller

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encapsulates. This may be attributed to the fact that smaller particles have greater surface area-to-volume ratio which accelerates the release of surface absorbed EOs.[54,105]

5.3 *Erosion and fragmentation*

Another mechanism of EOs release from the encapsulates involve biodegradability of the polymer. Various mechanisms like surface erosion, disintegration, and fragmentation are responsible for the degradation of the polymer (biodegradability) and release of the active ingredients.[88] These processes allow nearly complete breakdown of the polymeric encapsulates and help in the release of EO which remains chemically linked with the wall of the polymer material after the initial bursting. Since this is the last phase of EO release and only a small amount of EOs are left bounded to the polymer, the release rate is very slow, hence it is known as the lag phase. Very limited literature is available for these studies and requires further investigation.

6. Conclusions

For the commercial application, the stability and long lasting release of EOs has to be enhanced. For this purpose, number of techniques has been developed in recent past, out of which encapsulation using various biomaterials is one of the most viable process. The EOs encapsulates have been explored for the various applications like cosmetics, pharmaceuticals, perfumery, and therapeutics. Although, the techniques for the formulations are well developed but lack consistency in EOs loading to the polymers. Nano-encapsulation has provided few alternate methods for efficient and consistent loading of EOs; still the opportunities are many to be explored. Apart from these the better quantitative, reliable, and time efficient techniques are required for the physico-chemical characterization of encapsulates. The release characteristics are required to be tested in natural conditions in a time efficient manner with the study of multifactorial effects of environment on the EOs encapsulates.

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