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Review Article

Microencapsulation of Essential Oils by Spray-Drying and Influencing Factors

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Essential oils (EOs) are known as any aromatic oily organic substances which are naturally synthesized in plants. Exhibiting a broad range of biological activities, EOs have played a key role in numerous industries for ages, including pharmaceutical, textile, and food. However, the volatility and high sensitivity to environmental influences pose challenges to the application of EOs on industrial scale. Microencapsulation via the spray-drying method is one of the promising techniques to overcome these challenges, thanks to the presence of wall materials that properly protect the core EOs from oxidation and evaporation. By optimization of key factors related to the infeed emulsion properties and spray-drying process, the encapsulation efficiency and retention of encapsulated EOs could be significantly improved, thus allowing a wide range of EO applications. This review attempts to discuss on different determining factors of the spray-drying process to develop an effective encapsulation formula for EOs. Furthermore, recent applications of encapsulated EOs in the fields of foods, pharmaceuticals, and textile industries are also thoroughly addressed.

1. Introduction

Essential volatile oils, which are commonly known as essential oils (EOs), are one of the secondary compounds naturally synthesized in various organs of aromatic plants, including fruits, leaves, flowers, bark, stalks, and roots [1, 2]. Among over 300 different compounds identified in EOs, terpenoids (e.g., isoprenoids, monoterpenoids, diterpenoids, and sesquiterpenoids) are the major constituents, followed by oxygenated derivatives (e.g., alcohols, esters, aldehydes, phenols, and ketones) as well as phenylpropanoids (e.g., eugenol and cinnamaldehyde) [1, 3, 4]. Despite accounting for less than 5% of the dry basis of raw materials, EOs are essentially involved in (1) protection against herbivores and pathogen attack, (2) signaling regulation, and (3) responses to environmental stimulation. It is believed that most aromatic EOs were firstly extracted by ancient Egyptians dated back to 4500 B.C. [5]. Down the centuries, the demand for EOs has extended across the Middle East, North Africa, and Europe [6]. A tremendous amount of EOs was utilized by the ancient civilizations in Egypt, China, and India for traditional remedies, perfume production, and home cooking [7].

To date, the international market of EOs has reached over $17 billion (2019), mainly distributed in food (as natural flavors, preservatives, and edible packages), pharmaceutical (functional properties), and home (as fragrance and cleaning products) segments [8, 9]. These industries have recognized EOs as a promising alternative for synthetic chemicals, providing valuable health benefits to global consumers as well as minimizing environmental impacts.

The health-promoting benefits of EOs are given by their biological activities, which have been intensively exploited over the past few decades. Specifically, antioxidant, antimicrobial, anti-inflammation, anticancer, insecticidal [9], antipigmentation [10] and antiviral [11] properties have been recognized as highlighted properties of EOs. Firstly, the antioxidant activity of EOs is derived from their content of phenolic compounds (e.g., β-pinene, sabinene, γ-terpinene, limonene, and myrcene), targeting to free-radical scavenging and lipid peroxidation which could either prevent or cure various cell damage and diseases [12–15]. These phenolic compounds also take the main responsibility in the antimicrobial activity of EOs against a wide array of Gram-negative and Gram-positive bacteria such as *E. coli* [10], *P. aeruginosa* [14], *Streptococcus* spp. [16], and *Lactobacillus* spp. as well as many yeast and mould [17]. Due to natural lipophilicity, EOs molecules were able to penetrate through the lipids of the bacterial cytoplasmic membrane, disrupting the membrane structure and causing the loss of cell constituents [12]. EOs have also been employed in clinical treatment to treat inflammatory diseases [18]. The biologically active compounds of EOs contribute to restricting the histamine formation or diminishing the production of inflammation mediators [1]. Recently, these biological activities of EOs have even been taken to the next level by combination therapy and nanotechnology development [19, 20]. Combinations of EOs with other EOs or with synthetic chemical agents (e.g., commercial antibiotics and antimicrobial peptide) have exerted synergistic effects towards multiple targets at a lowered dose of the individual compounds and less administration times, thus minimizing possible adverse effects as well as resistance development [21–24]. Meanwhile, the use of numerous nanomaterials as a delivery system for EOs has been shown to significantly improve EOs stability and controlled release and bioavailability while extending their efficacy and shelf-life over a prolonged period of time, thereby successfully counteracting antibiotic-resistant microbial biofilm formation and their associated chronic diseases in human, animals, and plants [25–27].

As the yield and quality of EOs are highly prone to environmental conditions, an effective extraction method is in high demand [28–30]. For this reason, an encapsulation method has been proposed to enhance the EOs stability and controlled release [31]. The process involves emulsifying core materials (e.g., EOs) with coating materials (e.g., carbohydrates or proteins) and then cooling or drying the resulted emulsion [32]. Encapsulation provides a firm protection against moisture, oxygen, light, and undesirable interactions with insects, thus minimizing their effects on the EOs shelf-life and activities [33–36]. The final products of the microencapsulation process are microcapsules with various sizes ranged from a few micrometers to a few millimeters, in the form of particles, capsules, droplets, or complexes [37].

Currently, encapsulation is performed via emulsification, coacervation, spray-drying, complexation, and nanoprecipitation methods [38, 39]. Selection for an appropriate encapsulation method is entirely dependent on the wall material. An emulsion-based encapsulating method, which is commonly used to pack poorly water-soluble ingredients, results in discrete droplets or capsules after removing the organic solvent [40]. The coacervation technique is applied to encapsulate hydrophobic molecules, based on the electrostatic interaction between colloids that gives rise to spontaneous liquid-liquid phase separation [41]. The nanoprecipitation method, which is commonly known as solvent displacement, is relatively similar to the emulsification method. The nanoprecipitation results in the formation of nanoparticles suspension in water after solvent evaporation. Complexation is the process of adding EOs to an aqueous solution and then applying certain heat to obtain desired complexes. All four mentioned techniques present disadvantages with regards to high cost, high solvent consumption, and highly complex industrial scale [42].

Therefore, due to cost-effective operation, high retention of core materials, and effortlessly convenient equipment, spray-drying is commonly used in food as well as flavoring industries [35, 36]. This technique physically converts EOs from a hydrophobic liquid into a more convenient low-moist powder form, which contributes to promote the alternative use of EOs [43, 44].

The present review aims to discuss about the spray-drying process applied in EO encapsulation, focusing on (1) the factors which influence the spray-drying encapsulation efficiency as well as (2) encapsulated EOs uses in food, pharmaceutical, and textile industries.

2. Encapsulation of EOs via the Spray-Drying Process

2.1. Material Preparation and Process Mechanism. Spray-drying is a known a common encapsulation technique and has been well developed in pharmaceutical, chemical, food, and flavor industries [45]. Since its discovery in 1937 by A. Boake Roberts, the technique of spray-drying has played a significant role in producing various powdered flavors and encapsulating aromatic EOs [46, 47], vitamins [48], minerals [49], colorants [50], fat [51], aroma compounds [34], oleoresins [52], and enzymes [53–56]. In general, spray-drying is a flexible and low-cost method for protecting core biomaterials using spray-drying readily available equipments [44].

Materials for spray-drying are prepared by firstly dissolving the wall or coating material made from maltodextrin, modified starch, gum, or their combination and then allowing them to completely saturate in a refrigerator overnight [43]. The addition of emulsifiers may depend on emulsifying properties of the wall materials. The wall matrix: core material ratio is varied according to different

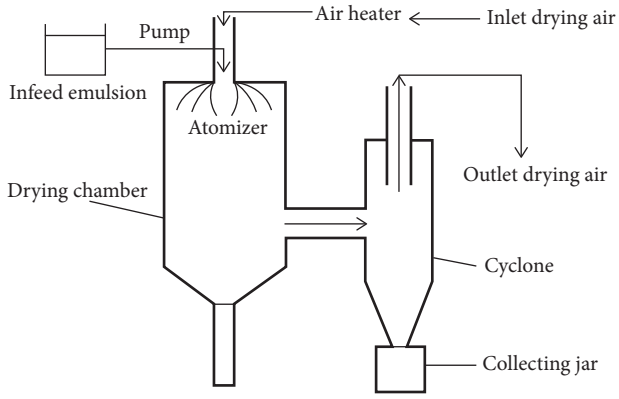

Figure 1: Schematic diagram of the spray dryer.

experimental conditions. The prepared oil-in-water emulsion is then speedily homogenized to produce emulsions with smaller oil globules, thus reducing the attractive forces between the globules and stabilizing infeed emulsions [54]. The mixing time at high speed is maintained. The emulsion once formed when contacted with heated air under high pressure would form a mist spread in the drying chamber (Figure 1). Three basic types of atomizers which have been broadly utilized are the high pressure nozzles, the double piston, and the spinning wheel [46]. The atomizers are responsible for producing uniform-sized, homogeneous, and smooth spray-dried particles. Next, inside the drying chamber, the hot air steam encounters the atomized droplets and dehydrates the solvent, producing dried particles which are small droplets of the core ingredient covered in a wall matrix [43]. Finally, the dried particles travel through the cyclone to the collecting jar where they are finally collected.

As a result, the spray-drying process produces highly fine discrete particles with a variety of sizes depending on the type of wall matrix material. Each particle contains a complex matrix in which EO globules are freely dispersed. As previously mentioned, the low-cost processing, available industrial-scale production, diverse encapsulating matrices, high volatile compounds retention, readily available equipment, and good quality of spray-dried particles are some significant advantages of microencapsulation by spray-drying [36, 43]. Apart from these advantages, the main limitation is that the obtained microcapsules are partially present in an agglomerated form, which is due to fiber formation between polymeric chains [36]. Hence, this phenomenon requires the wall materials to be carefully selected in the case of liquid application [42, 43]. Overall, spray-drying encapsulation is a low-cost and energy-efficient technology that can be applied to various food ingredients, enabling encapsulation of thermolabile compounds with low degradation risk, high encapsulation efficiency, and prolonged shelf-life.

2.2. Factors Which Affect the Efficiency of Spray-Drying Encapsulation of EOs. Successful EO encapsulation is expected to bring about high efficiency, minimum amount of unencapsulated material, and maximum retention of encapsulated powder. For this reason, a significant number of research studies have been conducted over the past few years to optimize the retention of volatile compound content in encapsulated EOs [33, 37, 51]. According to King and Hecht [57, 58], the volatile compounds can be lost through (1) flowing of atomized emulsion during atomization; (2) water diffusion of an unstable selective membrane, and (3) water interference towards morphological development. It is at the third stage that a greater amount of volatile compounds is lost, as compared to the others.

For a highly efficient spray-drying process, at least four criteria should be taken into consideration: (a) the type and concentration of wall materials; (b) the concentration of core materials and their composition retention; (c) the characteristics of infeed emulsion; and (d) the spray-drying conditions (Figure 2).

2.2.1. The Attributes of Wall Materials. Currently, wall materials derived from numerous natural and synthetic polymers have been employed as encapsulating agents of volatile compounds and oils. Selecting an appropriate material is an important step in infeed emulsion specification prior to spray-drying, as well as chemical interactions and volatile compounds retention throughout the process [42]. The choice of wall material is based on a number of factors regarding to the physicochemical properties of the core (e.g., solubility and porosity), the wall (e.g., molecular weight, viscosity, mechanic properties, film-forming, and emulsifying properties), compatibility, control mechanism, and economic factors [46]. Therefore, the favorable wall materials are carbohydrates such as modified and hydrolyzed starches, cellulose derivatives, gums, and cyclodextrins [32]. Among the available ingredients, maltodextrin, gum Arabic, and their combination have been commonly used due to excellent emulsifying capacities and volatile compounds retention [33, 34, 37] (see Table 1).

Maltodextrin (MD) $[(C_6H_{12}O_5).nH_2O]$ is a nonsweet polysaccharide which is made of multiple D-glucose units linked by α-1,4-glycosidic bonds. It normally presents as a white hygroscopic spray-dried powder or concentrated solution resulted from partial hydrolysis of corn starch with acids and/or enzymes [38]. MD is rapidly digested and absorbed as glucose and has been used as a food additive for a long time. The hydrolysis level of starch which is termed dextrose equivalent (DE) is directly associated with the degree of core material protection against oxygen penetration [39]. Using MD as the wall material is considered as inexpensive (one-third of the cost of modified starch), almost flavorless, less viscous, less hygroscopic, highly soluble in cold water, and excellently protectable for EOs, milk fat, soy oil, and fish oil [63]. MD is commonly combined with Arabic gum, modified starches, or proteins in a fixed ratio to improve the emulsifying stabilization. Thuong Nhan et al. [34] used MD and MD-gum mixture to carry out microencapsulation for lemongrass EO by spray-drying. The tested concentrations of wall material ranged from 15 to 30%, and the mixture ratio was 4 : 1. Tween 80 acted as an emulsifier to improve the stabilization of infeed emulsion.

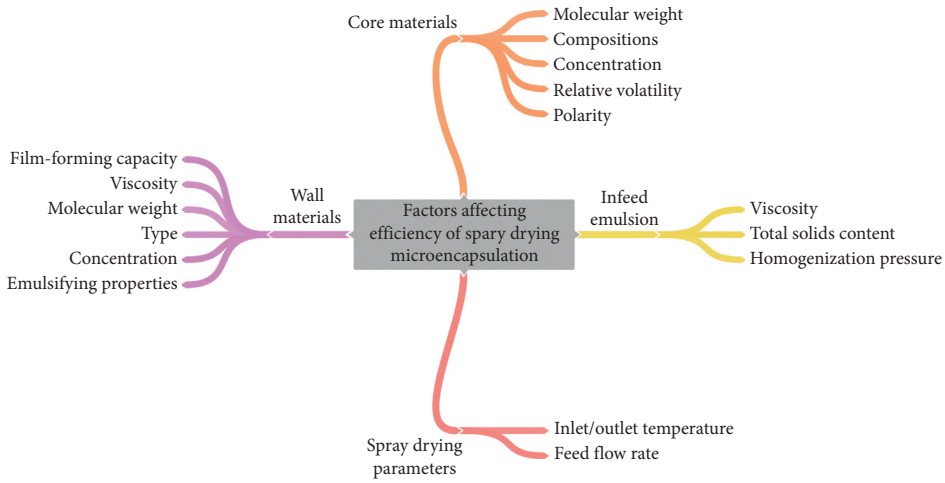

Figure 2: Factors affecting the encapsulation efficiency of spray-drying encapsulation.

Table 1: Encapsulation of EOs in different materials by spray-drying.

Wall materials	Core materials	Concentration	Results	Reference
Individual MD and MD-GA combination (4 : 1)	Lemongrass EO	15–30%	MD in individual form at 30% of concentration effectively encapsulated lemongrass EO at high encapsulation efficiency (84.75%) and oil retention (89.31%)	Thuong Nhan et al. [34]
MD and GA combination	Lavender EO	MD (25–58.33 of total solid) and GA (25–50% of total solid)	The highest encapsulation efficiency was obtained and minimum oil loading	Burhan et al. [33]
HPMC-MD-CSD mixtures (1 : 1:0 and 2 : 1:1)	Oregano EO	NA	The relation 2 : 1 (WM : C) without CSD enhanced the antioxidant activity and retention of volatile compounds after 90 days of storage	Asensio et al. [59]
Mesquite gum (MG), nopal mucilage (NM), and MG-NM mixtures (1 : 3, 1 : 1, and 3 : 1)	Lemon EO	30% of total solid content (w/w)	MG-NM combination exhibited a synergic effect in giving rise to desirable retention and shelf-life extension for lemon EO	Cortés-Camargo et al. [60]
MD-IP, MD-OSA, and MD-OSA-IP	Avocado EO	OSA equals to 3% MD base and IP : MD ratio equals to 1 : 9	The OSA modification of MD reduced by about 80% the oxidation of the EO as compared to its nonmodified counterpart	Sotelo-Bautista et al. [61]
GA, MD, MS, GA-MD mixtures (3 : 1, 1 : 1, 1 : 3), GA-MS mixtures (3 : 1, 1 : 1, 1 : 3), and GA-MD-MS mixtures (1 : 1: 1, 4: 1: 1, 1: 4: 1, and 1: 1: 4)	Lemon EO	30%	GA produced the highest encapsulation efficiency, followed by MS and MD.	Kausadikar, Gadhave, and Waghmare [62]

EO- essential oil, MD- maltodextrin, GA- gum Arabic, HPMC- hydroxypropyl methyl cellulose, CSD- colloidal silicon dioxide, IP- isolated protein, OSA-octenylsuccinic anhydride, MS- modified starch

Results have shown that lemongrass EOs (1.5%) had been encapsulated by MD (30%) at significant efficiency (84.75%) without interfering most of the Eos' chemical composition and product quality. MD has also been blended with whey protein isolate (MD-PI), octenylsuccinic anhydride (MD-OSA), and their combination (MD-OSA-PI) for avocado EO encapsulation. The MD-OSA blend produced homogenous-sized microcapsules which maintained EO stability for three weeks after the experiment, yet the encapsulation efficiency was the lowest, due to the sensitiveness of OSA to light and oxidation [61, 64]. Another work conducted by Asensio et al. [59] had successfully microencapsulated oregano EO by spray-drying using combinations of MD with hydroxypropyl methyl cellulose (HPMC) and colloidal silicon dioxide at 1 : 1:0 and 2 : 1:1 ratios as wall materials. Interestingly, the oregano EO microcapsules without CSD exhibited high antioxidant activity and lowered release of volatile compounds after 90 days of storage.

Gum Arabic (GA) obtained from the *Acacia* stems and branches mainly consists of complex polysaccharides (approximately 500 kDa), along with their potassium, calcium, and magnesium salts. Up to the present, GA is commercially

available in various forms, ranging from white to yellowish-white flakes, granules, powder, roller-dried, or spray-dried material [39]. GA is commonly employed as an emulsifier, stabilizer, and thickener in the production of confectionery and beverage industries due to high solubility, high oxidative stability, and excellent emulsification in a wide range of pH [65, 66]. Regarding EO encapsulation by spray-drying, GA is a common wall material due to high retention of many volatile compounds during the process. However, GA application faces difficulties in high cost, limited availability, and contamination. Therefore, researchers have made an attempt to either combine GA with different materials or to exploit for other alternatives [32].

Rajabi et al. [67] described the use of three types of wall materials (maltodextrin, gum Arabic, and gelatin) alone or in combinations for the microencapsulation of saffron extract by the spray-drying technique. As seen from the concerned blends, the stability of saffron bioactive compounds increased as the quantity of gelatin decreased in its blend with maltodextrin and gum Arabic up to 1% (w/w). The microencapsulation process was optimized at 40% total solids, consisting of maltodextrin, gum Arabic, and gelatin in the w/w ratio of 0.94 : 0.05 : 0.01.

Guergoletto et al. [68] invested a study on, in vitro, the antioxidant capacity and prooxidant effects of juçara pulp fermented with two bacterial strains before and after spray-drying with maltodextrin, gum Arabic, or gelatin and storage at 25°C for 90 days. The reduction in total phenolic content and antioxidant capacity was recorded when the samples were dried with maltodextrin (24% decrease of total phenolic content and 19% of DPPH scavenging), followed by gum Arabic (decrease of 50.7 and 69%) and gelatin (decrease of 61 and 78%).

Flores-Belmont et al. [69] evaluated the survival of *Lactobacillus acidophilus* in model gastrointestinal conditions through microencapsulation by simple or double spray-drying using chitosan-inulin or chitosan-maltodextrin (1 : 15 or 1 : 25) solutions. A microbial reduction of 7 log cycles was obtained for the simple microencapsulated probiotic and only 3 log reductions for the double microencapsulated probiotic. The double microencapsulated probiotic, thus, demonstrated greater resistance to simulated gastrointestinal conditions.

Kausadikar, Gadhave, and Waghmare [62] have used native MD, native GA, native modified starch (MS), GA mixed with MD (the ratios of 3 : 1, 1 : 1, 1 : 3), and GA mixed with MS (the ratios of 3 : 1, 1 : 1; 1 : 3) as the wall materials for the encapsulation of lemon oil by spray-drying. The result showed that the encapsulation efficiency ranged from 50.52% to 83.60% with the highest value obtained from 1 : 1 GA-MS and the lowest value obtained from native MD. The encapsulation efficiency of GA was the highest, followed by MS and MD. Moreover, GA was recommended to blend with MD and MS to reduce production costs.

Wall materials made of GA-MD combination were used to encapsulate the lavender EO liquid into a solid powdered form using the spray-drying method. The optimal ratio of GA, solid concentration, and oil loading was found as 25 w/w%, 30 w/w%, and 16.67 w/w%, respectively, producing lavender oil microencapsulates at high encapsulation efficiency (77.89%) and remaining stable for up to 1 month of storage. However, once the content of either solid concentration or GA outweighed each other, such protective effect would suffer from high viscosity, delayed particle formation, and reduced solubility, which lead to low powder yield as well as EO volatilization and loss during the drying process. In general, these studies have proposed that a suitable wall material plays a key role in protecting the loaded active compounds and maintaining the microencapsulates' stability for use and storage in a long run. Beside the favorable use of MD and GA, the disadvantages of each individual compound should be improved via new formulation and strategies to enable clinical and industrial applications. In addition, the search for alternative materials and combinations, as well as optimization work, should be further extended to construct a specifically effective spray-drying encapsulation process for each EO.

2.2.2. The Properties of Core Material. Over the years, EOs from lemongrass [34], thyme [33], lavender [37], oregano [59], avocado [61], lemon [62], and cinnamon [70] have been successfully encapsulated by spray-drying. These studies reported in general that the spray-drying encapsulation acted as a protective barrier for the EO during processing, storage, and transport, thereby actively preventing the loss, volatilization, and degradation of EO as well as their chemical modification [71].

Despite the fact that spray-drying microencapsulation would mainly attempt to restrain the volatile compounds loss, the retention is also affected by the molecular weight, polarity, and relative volatility of the volatile compounds [32]. Firstly, the molecular weight of volatile compounds is the primary factor which determines their diffusion rate. A high molecular weight allows the compounds to have more interaction time with the atomized droplet surface during the drying process, thus increasing the volatile retention. The second factor that determines volatile retention is the relative volatility [72]. For instance, the retention of octanol, octenol, octanon, and octanal was correlated with their relative volatility when they were encapsulated with maltodextrin before spray-drying [72]. As a result, the higher the relative volatility was, the lower the retention was. The third factor is the polarity of volatile compounds. Studies have shown that the volatile compounds would become more polar when dissolving in water for a long time. Water fractions when passing through the selective membrane even at the late stage of the drying process enhance the volatile polarity, thus promoting volatile compounds loss [73]. Next, volatile compounds retention can also be varied according to their chemical composition. Goubet et al. [74] proved the retention of aromatic compounds increased proportionally with the functional group polarity. Besides, the retention of volatile compounds may indirectly depend on the physicochemical properties of the volatiles [75].

Particularly, the concentration of core materials or EOs significantly influences the volatiles retention as well as encapsulation efficiency. Optimally, the concentration

should provide high core retaining content and drying yield at a lowered cost and energy consumption. For example, upon encapsulation into GA wall material via the spray-drying method, increasing concentrations of lemon EO from 10% to 20% reduced the encapsulation efficiency of approximately over 10% [62]. Similarly, Frascareli et al. [76] and Thuong Nhan et al. [34] found that the total retention of coffee and lemongrass EO was dramatically dropped when an excessive concentration of these oils was loaded into the encapsulating particle. A possible explanation could be that the high oil concentration would enhance the emulsion viscosity which limits the oil diffusion to the drying particle surface.

2.2.3. The Characteristics of Infeed Emulsion. Encapsulation efficiency of EO and volatile retention by using the spray-drying method are solely dependent on the infeed emulsion. In order to prepare an effective infeed emulsion, the following parameters are taken into consideration: (1) total solid content, (2) emulsion stability, viscosity, size, and method, and (3) homogenization pressure.

The total content of solids determines the EOs encapsulation efficiency during spray-drying by regulating the emulsion viscosity and modulating the droplets circulation, leading to rapid semipermeable crust formation [32]. Estimating an optimum total solid content could prevent wasting wall materials and maintain the emulsion viscosity as well as stability at an appropriate level for better retention. Homogenization pressure could also affect the emulsion properties and encapsulation efficiency of spray-drying. In a study conducted by Frascareli et al. [76], the effect of various homogenization pressure (0–100 MPa) on the droplet size, emulsion viscosity, and the oil retention of microencapsulated basil EO was evaluated. Results have shown that increasing the pressure up to 50 MPa could break down the emulsion droplet to smaller sizes. In contrast, as the pressure continued to rise beyond 85 MPa, Brownian motion of the droplets and performance of emulsifiers are increased [77], causing the droplets to become thermodynamically unstable and coagulate with other droplets [72]. Homogenization pressure had no significant effect on viscosity, yet positively affected the oil retention.

Jafari et al. [73] completely discussed about the influence of infeed solid concentration on the rapid formation of crust on the surface of the droplet. As infeed solids concentration as well as emulsion viscosity increased up to an optimum point, the internal circulations and oscillations inside the droplets will be reduced. At the optimum viscosity, the atomization process will be easy to perform; thus, rapid a semipermeable membrane and reasonably spherical particles will be formed. In parallel, the selective diffusion will be put into action earlier, the volatile compounds will be rapidly trapped, and the retention will be improved. However, the viscosity beyond its optimum limit will produce larger droplet sizes. During atomization, larger droplet sizes cause the larger exposures and the slow formation of discrete particles. The more thermal exposure leads to a rational decrease of the retention.

2.2.4. Temperature and Feed Flow Rate. Apart from initial emulsion, selection for an appropriate feed flow rate and the inlet and outlet drying air temperatures also contributes to maximize the encapsulation efficiency.

Numerous studies have reported the influences of drying temperature on coffee EO [76], fish EO [77], mandarin EO [78], and lemon EO [60, 62]. These studies proposed that the optimal range of inlet temperature for effective spray-drying encapsulation was between 160°C and 220°C, at which rapid semipermeable membrane formation and high flavor retention occurred [32].

The feed flow rate also significantly affects the physical properties of microencapsulates. Fernandes et al. [79] investigated the influence of various feed flow rates (0.5–1.0 L h^{-1}) and inlet air temperatures (135–195°C) on rosemary EO microencapsulation. Results showed that high temperature combined with slow rate of feed flow resulted in lowered particle moisture content, hygroscopicity, and wettability of the powders. This study suggested that the best spray-drying conditions included the wall material concentration of 24%, the inlet temperature of 135°C, and the feed flow rate of 0.7 L h^{-1}. Another work has been carried out by Maskat et al. [80] to determine the effects of inlet air temperature (150, 160, and 170°C) and feed rate (280, 350, and 420 mL h^{-1}) on encapsulating roselle extract by spray-drying. These two factors significantly affected yield, color, moisture, and anthocyanin content of spray-dried roselle, whereby increasing feed flow rate would reduce the moisture content and promote anthocyanin degradation.

3. Applications of Microencapsulated EOs

As compared to other microencapsulation methods, spray-drying has shown several dominant advantages such as equipment availability, materials diversity, and large-scale production of microencapsulates with high retention and stability and low processing cost, hence their favorable use for numerous EOs [81]. EOs encapsulated by spray-drying is applied in foods, pharmaceuticals, textiles, and footwear products (see Table 2).

3.1. Food Sector. Food spoilage and decay caused by bacteria and fungi during postharvest, processing, and storage stages results in tremendous economic losses and consumer dissatisfaction [109–111]. As applications of food preservatives and fungicides may unexpectedly increase the toxicity level of the food, as well as trigger resistance development [42, 112]. Consumer preferences have shifted towards natural-based alternatives (i.e., biopreservatives or "green" preservatives) which appear to be healthier and more eco-friendly and cost effective [113]. In this regards, EOs have been favorably employed.

Kringel et al. [84] carried out a work to demonstrate the strong antifungal activity of orange EO in bakery products. The EO was loaded into a zein and β-cyclodextrin combination and inhibited the growth of *A. terreus*, *A. Niger,* and *A. fumigatus*. In the presence of encapsulated orange EO, the

Table 2: Microencapsulated EOs applied in various fields.

Fields of application	Type of EO	Purpose	Result	Reference
Food	Thyme EO	Antimicrobial	Thyme EO encapsulated by casein-maltodextrin by the spray-drying method exhibited antioxidant, as well as *in vitro* and *in situ* antibacterial against *L. monocytogenes, E. coli,* and *S. typhimurium* were also inhibited	Radünz et al. [82]
	Lemongrass EO	Insecticidal	The release of citral from the encapsulated lemongrass EO in a slow and controlled manner has exerted an insecticidal effect against potato tuber moth up to one week of experiment	Jovanović et al. [83]
	Orange EO	Antifungal	Encapsulation into zein and β-cyclodextrin complexes has enhanced the capability of orange EO in decelerating the cake spoilage caused by *A. terreus, A. Niger,* and *A. fumigatus* growth from 30 to 150 days	Kringel et al. [84]
	Lamiaceae EO and oregano EO	Antifungal	The use of niosome-based encapsulated EOs has reduced the fungal growth and aflatoxin accumulation in maize grains for 75 days	García-Díaz et al. [85]
	Oregano EO	Antifungal	Nanoencapsulated oregano EO inhibited the growth of *Cladosporium* sp., *Fusarium* sp., and *Penicillium* sp. isolated from Minas Padrão cheese	Bedoya-Serna et al. [86]
	Rosemary EO	Antibacterial	Encapsulation of rosemary EO onto chitosan-benzoic acid nanogel reduced the growth of *S. typhimurium* which helped in extending meat shelf-life	Hadian et al. [87]
	Lemon EO	Stability	Stability of encapsulated lemon oil was maintained up to 6 months	Kausadikar et al. [62]
	Rosemary EO	Antimicrobial	The mucilage-coated rosemary EO maintained the ascorbic acid content and color, reduced peroxidase enzyme, and inhibited microbial growth	Alikhani [46]
Food Packaging	Cinnamon EO	Antimicrobial	The Ag^{+}/Zn^{2+}-permutite containing cinnamon EO showed sustained release and strong inhibitory activity against *Aspergillus Niger* and *Penicillium* sp., thus controlling the degradation of fresh Chinese bayberry	Niu et al. [88]
	Lemongrass EO	Antimicrobial	Lemongrass EOs was sustainably released from alginate films and exhibited antimicrobial activities against *E. coli* and *B. cinerea.* Such activity was affected by the droplet size and storage conditions (4°C)	Riquelme et al. [89]

Table 2: Continued.

Fields of application	Type of EO	Purpose	Result	Reference
Pharmaceutical	Thyme EO	Antimicrobial and stability	Upon encapsulation to lipid matrix microparticles, thyme EOs quality was stable during feed pelleting and storage processes. In addition, the oil was slowly released in a controlled manner in weaned pigs	Choi et al. [90]
	Jasmine EO	Antioxidant and anticancer	By loading onto pectin/chitosan nanoparticles, the anticancer activity of jasmine EO against MCF-7 breast cancer cells was 13-folded improved, possibly due to enhanced thermal stability	Attallah et al. [91]
	Lime EO	*Propionibacterium acne* treatment	Results from disk difusion assay have shown that lime EO encapsulated into chitosan nanoparticles has improved its inhibitory activity against *Propionibacterium acnes,* as compared to the nonencapsulated formula	Julianti and Rusliati [92]
	Rosemary EO	Antimicrobial and wound healing	Rosemary EO loaded into the lipid nanocarriers (NLCs) inhibited *Streptococcus epidermidis*, *Streptococcus aureus*, *E. coli, Listeria monocytogenes*, and *Pseudomonas aeruginosa*. It also accelerated wound healing and reduced the rate of tissue bacterial colonization	Khezri et al. [93]
	Vetiver EO	Sedative activity and hypnotic activity	Vetiver oil cross-linked polymeric microcapsules were able to provide a sustained release of encapsulated oil	Ali et al. [94]
	Peppermint EO	Antibacterial activity and wound healing	Peppermint-EO-loaded nanostructured lipid carriers (NLC) showed antibacterial activity against *S. epidermidis*, *L. monocytogenes*, *E. coli*, *P. aeruginosa,* and *S. aureus*. The animals administered with NLC showed increasing wound contraction rate	Ghodrati et al. [95]
	Babchi EO	Dermatological treatment	Encapsulation of Babchi EO in microsponges exhibited antimicrobial activity against *P. aeruginosa*, *S. aureus,* and *E. coli*.	Wadhwa et al. [96]
	Rosemary EO	Anti-inflammatory	Nanoemulsion containing rosemary EO was confirmed to inhibit the inflammatory process in zebrafish	Borges et al. [97]
	Citronella EO	Mosquito repellent	The efficiency of applying β-cyclodextrin containing the citronella oil system as repellent against *Aedes aegypti* was 84.67% after 5 minutes	Pujiastuti et al. [98]
	Cardamom EO	Antimicrobial	Cardamom-oil-loaded chitosan nanoparticles combatted β-lactamase-producing *E. coli* and MRSA	Jamil et al. [99]

Table 2: Continued.

Fields of application	Type of EO	Purpose	Result	Reference
Textile	Limonene and permethrin	Mosquito repellent	Cotton fabrics containing limonene and permethrin capsules showed repellency after 20 washing cycles	Türkoğlu et al. [100]
	Tea tree EO	Antibacterial	The capsules made of tea tree EO and Arabic gum, polyvinyl alcohol, and β-cyclodextrin were applied to viscose fabric, showing antibacterial activity against *E. coli* and *S. aureus*	Beşen et al. [101]
	Lime EO	Antibacterial	The growth of *E. coli*, *B. cereus*, *S. typhimurium,* and *S. aureus* on the surface of cotton fabrics containing lime EO microcapsules was inhibited before and after washing	Wijesirigunawardana et al. [24]
	Thyme EO	Antimicrobial	Microcapsules of thyme EO and gelatin/gum Arabic were applied on nonwoven fabric and showed inhibitory activity against *S. aureus, E. coli*, and *C. albicans*	Karagonlu et al. [102]
	Vanillin and limonene	Antibacterial agent and fragrance	Application of limonene and vanillin encapsulated in chitosan/gum Arabic inhibited *E. coli* and *S. aureus* growth	Sharkawy et al. [103]
	Lemongrass EO	Mosquito repellency	Cotton fabrics containing lemongrass encapsulates had resistance against mosquito bites and durability after 30 wash cycles	Vinayagamoorthy et al. [104]
	Citronella EO	Fragrance	Gelatin-gum Arabic citronella EO microcapsules formed hydrogen bonds with cotton fiber, thereby releasing EO in a controlled manner from the cellulose matrix	Bezerra et al. [105]
	Thyme EO, cypress EO, and grapefruit EO	UV protection and mosquito repellency	Applying EOs onto bamboo/tencel fabric resulted in UV protection and mosquito prevention for a long duration	Geethadevi et al. [106]
	Rosemary EO, Bergamot EO, and lemon EO	Fragrance	EOs were encapsulated in methyl methacrylate-styrene, resulting in nanocapsules with small size and uniform distribution. These nanocapsules were applied to produce fragrant fabrics with good thermal stability and washing durability	Liu et al. [107]
	Peppermint EO	Antimicrobial agent and fragrance	Encapsulated peppermint EO in alginate was applied on cotton fabric. The EO was slowly released and exhibited inhibitory activity against *S. aureus* and *E. coli*	Ghayempour et al. [108]

microbiological spoilage was restrained up to 5 months, indicating its potential to extend the shelf-life of bakery products. In another study conducted by Alikhani [46], rosemary EO coated by mucilage was able to maintain the ascorbic acid content and customer satisfaction, while decreasing the rate of peroxidase activity and microbial growth. The results indicated that application of mucilage encapsulated rosemary EO effectively prolonged the quality and extended the storage period of sliced mango. Overall, the strong inhibitory activity against fungi and bacteria of encapsulated EOs applied in food products has been well documented in several reports [85–87, 114–116].

In addition to antimicrobial activity, the insecticidal effect of encapsulated lemongrass EO against *Phthorimaea operculella* was investigated by Jovanović et al. [83]. The release of the citral compound present in the encapsulated EO has caused a lethal effect to the potato tuber moth for seven days as compared to the nonencapsulated one. Recently, the use of encapsulated EOs has emerged in food industry as a safe and high-quality packaging for food products [117]. Nanoencapsulation is reported to improve EOs' solubility, stability, and volatilization, thereby maintaining their biological activities during transport and storage. For example, according to Hadian et al. [87], the

chitosan-benzoic acid nanogel has improved the antioxidant and antimicrobial activities, as well as stability of rosemary EOs. The nanoencapsulated rosemary EOs could be used as an effective tool to reduce *Salmonella* spp. and extend the shelf-life of meat.

3.2. Pharmaceutical Sector. Regarding the pharmaceutical field, EOs are firstly considered as an effective alternative for conventional antibiotics to combat multidrug-resistant pathogens, whose development is derived from indiscriminate use of antibiotics [118]. Indeed, Khezri et al. [93] have investigated the antimicrobial activity and wound-healing capacity of rosemary EO loaded into lipid nanocarriers (REO-NLCs). Results have shown that, in addition to *in vitro* antibacterial activity against several Gram-positive and Gram-negative bacteria, REO-NLCs also promoted wound healing in mice by reducing bacterial density, vascularization, and re-epithelialization. A similar observation was reported by Ghodrati et al. [95]. By loading onto NLCs, peppermint EO exhibited antibacterial activity and *in vivo* wound-healing effect. Meanwhile, according to Jamil et al. [99], cardamom EO encapsulated into chitosan nanoparticles was able to combat β-lactamase-producing multidrug-resistant *E. coli*.

Secondly, the applications of encapsulated EO have been branched towards mosquito-borne diseases and anti-inflammation. For instance, Pujiastuti et al. [98] have reported the mosquito-repellent action of citronella EO contained in β-cyclodextrin. The liquid microcapsules upon application onto the mice skin showed repellency percentage of 84.67% after 5 min. Along with the mosquito-repellent activity, the encapsulated oil had a lowered volatility rate. In terms of anti-inflammatory activity, Wijayadi and Rusli [119] revealed that encapsulating lime EO into chitosan nanoparticles improved the inhibitory activity of the EO against *Propionibacterium acnes* with minimal side effects. Wadhwa et al. [96] has formulated Babchi EO encapsulated in ethyl cellulose microsponges for dermatological treatment. The formulation effectively inhibited *P. aeruginosa, S. aureus,* and *E. coli*. The safety and stability of the prepared microsponges on dermal cells were also enhanced, as compared to the nonencapsulates. Furthermore, Ali et al. [94] developed a polymeric encapsulation system for vetiver EO. The presence of SA and gellan gum as wall material allowed EO release in a sustained manner and exhibited a sedative-hypnotic activity in an animal model. Notably, thyme EOs and lauric acid were loaded into alginate microparticles to maintain the oil stability during processing, showing a controlled release profile in a simulated intestinal tract environment. Encapsulation of EO has also been applied in fabrication of anticancer delivery systems. For instance, jasmine EO encapsulated into pectin/chitosan composite nanoparticles has been shown to exhibit anticancer activity approximately 13 times higher than the nonencapsulated EO and enhanced normal cell viability [91].

3.3. Textile Sector. During the past few decades, mosquito-borne diseases such as dengue fever and malaria have vastly caused severe harm to millions of people worldwide [120]. Hygiene has, therefore, become one of the major concerns among the world population. To date, the use of EOs has emerged in textile industry as a revolutionary alternative for synthetic repellents, taking advantage of their natural antimicrobial ability, nontoxicity, nonirritation, and environment friendliness [121, 122]. With the advances in microencapsulation techniques, the loss and deterioration of EO-based mosquito repellents due to oxidation and volatilization can be prevented throughout a long period of time [123–125]. According to Özyildiz et al. [126], the disposable nonwoven fabrics containing ozonated red pepper seed EO microcapsules actively reduced up to 5 log of antibiotic-resistant micro-organisms in 1 h. In another work conducted by Kim and Sharma [75], the application of microcapsules made of red thyme EOs combined with clove bud into fabrics has helped in getting rid of 94% of harmful household dust mites. Meanwhile, Türkoğlu et al. [100] have used limonene and permethrin capsules to provide mosquito-repellent activity for cotton fabrics. The treated fabrics were protected from mosquitoes, with mortality rates of 41% and 54% for limonene and permethrin, respectively. The repellent efficiency of fabrics remained after 20 washing cycles. Antimicroorganism textile has also been exploited to prevent colonization of skin pathogenic as well as odor-generating bacteria and fungi [127]. Beşen et al. [101] attempted to produce disposable fabric (100% viscose nonwoven) containing tea tree EO microcapsules. The fabric exhibited antibacterial activity against *S. aureus* and *E. coli*. Meanwhile, Wijesirigunawardana et al. [24] demonstrated that cotton fabric carrying lime EO microcapsules effectively inhibited *E. coli*, *B. cereus*, *S. typhimurium,* and *S. aureus* even after washing. Similarly, antimicrobial effects against *S. aureus, E. coli*, and *C. albicans* were observed when applying gelatin-gum Arabic thyme EO microcapsules at different concentrations onto fabrics [102].

The encapsulated EO also produced scent and UV protection. Bezerra et al. [105] carried out production of fragrant textile using encapsulated citronella EO. The cotton fiber formed a hydrogen bond with the microcapsules, aiding in controlled release of EO from the cellulose matrix. Geethadevi and Maheshwari [106] reported the UV protection of bamboo/tencel fabric containing encapsulated thyme EO, cypress EO, and grapefruit EO. These EOs formed a firm layer on the fabric surface to prevent sunray penetration. The fabric durability of UV protection was enhanced up to 30 washes.

4. Conclusions and Future Perspectives

For centuries, the diversity of composition and biological activities have made EOs a crucial provider of health benefits to the global population. Up to the present time, EOs have been employed in various industries as a natural and healthy alternative for synthetic chemicals. However, the stability of EOs against adverse conditions during manufacturing, storage, and transport raises a major concern about their quality degradation and concentration loss as well as non-target delivery. Therefore, encapsulation via the spray-drying method has been widely proposed to effectively

overcome these limitations, producing EO powder with extended longevity at a relatively low cost. Furthermore, the biological activity, controlled release, and quality of EOs can also be maintained.

Regarding the spray-drying encapsulation method, four parameters related to the process itself and emulsion properties would be taken into consideration: the attributes of wall materials, the profiles of core materials, the characteristic of infeed emulsion, the inlet/outlet temperature, and the feed flow rate of the pump. Optimization work for each of these parameters is required to continue to provide optimal conditions in order to (1) achieve maximum encapsulation efficiency and oil retention and (2) maintain controlled release and efficacy of encapsulated EOs, thereby extending their use and applications. Numerous studies have shown that food, pharmaceutical, and textile are among the segments in which EO encapsulation is most commonly applied. In the food industry, the presence of microencapsulates plays a crucial role in preventing food degradation by taking advantage of antimicrobial and insecticidal activities over a prolonged storage time. In the pharmaceutical industry, release of encapsulated EOs in a sustained manner effectively targeted multidrug-resistant bacteria, skin inflammation, and cancer cells in animal models. In the textile industry, the fabrics containing encapsulated EOs exhibited remarkable repellent activity against mosquitos and micro-organisms and UV damage after multiple washing times. This evidence has again highlighted the essential role of EO encapsulation and provided helpful insights for further exploitation in the future.

The potential application of EO microencapsulation has been investigated under various environmental conditions. Based on the current insights, it is recommended to investigate the effects of storage time and handling practices on the encapsulated EO retention. Further studies on sensorial analysis of encapsulated EOs could be conducted to improve consumer satisfaction and economic profits. In addition, evaluation of the product hygroscopicity under different humid conditions is required to support the storage time prediction as well as selection for compatible packaging materials. Finally, the exploration for new wall materials should also be in demand to extend the use of encapsulated EOs toward environmental sustainability and cost efficiency.

Data Availability

No data were used to support this study.

Conflicts of Interest

The authors declare no conflicts of interest.

Authors' Contributions

Nguyen Thi Thu Trang, Le Thi Van Anh, Dang Nhu Ngoc, Nguyen Dan Chi, Nguyen Phu Thuong Nhan, Tran Thanh Truc, Nguyen Quang Vinh, Long Giang Bach, and Pham Nguyen Thuy Dung conducted investigation; Le Thi Van Anh and Long Giang Bach supervised the work; Nguyen Thi Thu Trang, Dang Nhu Ngoc, Nguyen Phu Thuong Nhan, Tran Thanh Truc, and Nguyen Quang Vinh wrote the original draft of the manuscript; Nguyen Thi Thu Trang and Pham Nguyen Thuy Dung reviewed and edited the manuscript.

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