Microencapsulation of Essential Oils by Coacervation

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Contributor: Alicja Napiórkowska, Marcin Kurek

Essential oils (EO) are called essential in the sense that it contains the essence of the aroma of the plant it is derived from, whereas the term "oil" is used because it contains the oil-soluble chemicals in the plant, not only because it feels oily. EOs are also known as volatile oils, ethereal oils, or aetheroleum. The wider use of essential oils in the food industry is severely limited. This is because these substances are highly sensitive to light, oxygen, and temperature. This creates problems with their processing and storage. In addition, they have a strong smell and taste, which makes them unacceptable when added to the product. The solution to this situation seems to be microencapsulation through complex coacervation. To reduce the loss of essential oils and the undesirable chemical changes that may occur during their spray drying—the most commonly used method—complex coacervation seems to be an interesting alternative.

Keywords: essential oils ; proteins ; microencapsulation

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1. Essential Oils

Essential oils (EO) are multi-component, hydrophobic mixtures containing up to several hundred volatile compounds (usually 100 to 200 chemicals per essential oil) in different concentrations. Essential oils can be characterized by two or three major components at relatively high concentrations (20–70%), which determines the biological properties of EOs. The main ingredients chemically are terpenes, aldehydes, ketones, phenols, alcohols, and others [1][2][3]. This complex chemistry gives them their therapeutic properties and explains why different essential oils may have overlapping effects [4][5][6][7]. Some examples of major components can be cited—carvacrol and thymol represent, respectively, 30% and 27% of the composition of EO from oregano (*Origanum compactum*). EOs from coriander (*Coriandrum sativum*) have in their constitution 65% linalool. Menthol (59%) and menthone (19%) are found in EOs from peppermint (*Mentha piperita*) [8][9].

Essential oils are characterized by having many pharmacological properties, including anti-inflammatory, antispasmodic, sedative, analgesic, and digestive-supporting properties. Due to their very rich and diverse chemical composition, one essential oil can have several positive effects. For example, rosemary essential oil has the effect of improving digestion, enhancing appetite, and being anti-flatulence. In addition, they have well-documented antimicrobial activity against bacteria, yeasts, and molds [4][5][10]. Again, thanks to its complex composition, one essential oil can effectively inhibit the growth of both bacteria and fungi. The same rosemary EO inhibits the growth of Gram-positive (*Enterococcus* spp.) and Gram-negative (*Salmonella* spp.) bacteria, yeasts (*Candida* spp.), and molds (*Penicillium* spp.) [11][12][13]. For this reason, EOs can be an alternative to the commonly used food-preservation agents.

This is indicated by studies conducted, inter alia, by Coimbra et al. [14]. The team tested the applicability of thyme essential oil (*Thymus zygis*) to *Listeria monocytogenes* in four food matrices (chicken juice, lettuce leaf model, ultra-high-temperature (UHT)-treated skim, and whole milk). EO inhibited the growth of *L. monocytogenes* 13305 in a model medium with chicken juice and lettuce. A significant reduction in the number was observed for the two highest concentrations of EO tested from 4 to 14 days for chicken juice and from 2 to 14 days for the model medium with lettuce. Research on the possibility of using essential oils for preserving food products was also conducted by Shah et al. [15]. Thymol concentrations used in apple cider resulted in complete bacterial inhibition or were bacteriostatic at 35 °C for *E. coli* and 32 °C for *L. monocytogenes*. Attempts have been made to use essential oils also for preserving such products as romaine lettuce, iceberg lettuce, mature bunched spinach, and baby spinach [16], snacks based on meat and seafood [17][18][19], juices [20], milk, yogurts, and other milk products [21][22][23] or chocolates [24], but also fruits or vegetables coated with edible coatings with the addition of EO [25][26].

Nanodispersion of eugenol (the basic ingredient of clove oil) in whey protein isolate and maltodextrin did not change its antimicrobial properties against *E. coli* O157: H7 and *Listeria monocytogenes*. However, nanodispersion allowed eugenol to be evenly distributed in the milk at concentrations above the solubility limit of the antimicrobial agent, which improved the antimicrobial efficacy in milk. Thus, nano-delivery systems hope to reduce the amount of antimicrobials without altering the turbidity of food products [21].

The stability of meat products during storage is a primary factor that is compromised by lipid oxidation and microbial growth. Hemmatkhak et al. [27] researched the use of active papers soaked in a nanoemulsion or Pickering emulsion containing cumin seed essential oil (CSEO). The effect of active papers on the quality and shelf-life of beef hamburgers stored at 4 °C for 7 days and at −18 °C for 60 days was investigated. Research results indicate good antioxidant and antimicrobial activity of cellulose papers impregnated in CSEO capsules. Packing beef burgers in contact with the produced active papers had a significant effect on extending the shelf life of hamburger samples by significantly reducing TBARS, the total number of mesophilic bacteria and psychrophilic. Furthermore, the sensory characteristics of the hamburgers did not changed.

It also seems important that many essential oils are on the Generally Recognized As Safe (GRAS) list published by the US Food and Drug Agency (FDA). Among the EOs that are approved for use in food are clove, rosemary, oregano, basil, mint, lavender, sage, cinnamon, and laurel [28][29][30]. Considering the above, essential oils can be not only a natural replacement for artificial preservatives but also a functional additive.

2. Microencapsulation as a Solution for Essential Oils Application Limitations

There are limitations to the use of pure essential oils in food products. First of all, their characteristic strong aroma and taste can cause undesirable organoleptic changes. In addition, EOs are very sensitive to the influence of the external environment —light, oxygen, and temperature. Other limitations are their lipophilic nature and hence low water solubility, low bio-accessibility, and bio-availability [3][31]. These problems can be solved by microencapsulation—an effective method of preserving the quality of sensitive substances. Microencapsulation is defined as a method of coating or encapsulating a given material or mixture of materials within the shell of a specific material or system. The substance that is encapsulated is called “active”, “encapsulate”, “payload”, or “core” and constitutes 30–99% of the total weight of the capsule. The core material can be a single substance or a mixture of solid, liquid, and gaseous forms. The enclosing polymer is called “shell”, “wall”, “matrix”, or “coating” [31][32][33]. The wall material is usually insoluble and non-reactive with the core material. The wall can be made of gums, proteins, lipids, and synthetic polymers. The wall material is generally applied as a liquid (solution, suspension, or molten material) to permit enrobing of the core material. Because the task of the shell is to protect the encapsulated substance, it should have excellent film-forming and barrier properties against oxygen, water, pressure, heat, and/or light [33][34]. A single microcapsule may have a round or irregular shape, depending on the method of producing the microcapsule, the type of active, and wall materials (**Figure 1**). The average size of the microcapsules is 100–500 µm [35]. Therefore, this process can provide many benefits to the use of EOs in food recipes, including protecting them from harsh conditions (light, shear, oxygen, moisture, heat, and others), improving their solubility and bioavailability, increasing their controlled release, and preventing their interaction with other food ingredients. This also allows for the reduction of volatilization of volatile substances, slowing mass transfer or modifying the physical properties of the core material. It reduces the evaporative loss of liquids and the reactivity of the core material, and extends the duration of its activity [32]. Encapsulation facilitates application by transforming the liquid into a solid phase, ensuring precise dosing, improving stability, and masking the encapsulated substance’s taste and/or smell. In addition, microencapsulation of essential oils can be a viable and effective approach to liquid food matrices with high water content by increasing their dispersibility. Facilitating the distribution of EOs in food areas where microorganisms thrive (water phase) and minimizing their particle diameter may also contribute to improving their antimicrobial properties. The smaller size of the molecules favors the migration and attachment to the bacterial cell walls [3][36]. Commonly used microencapsulation techniques are emulsification, spray-drying, coaxial electro-spray system, freeze-drying, coacervation, in situ polymerization, extrusion, fluidized-bed-coating, and supercritical fluid technology [31][33].

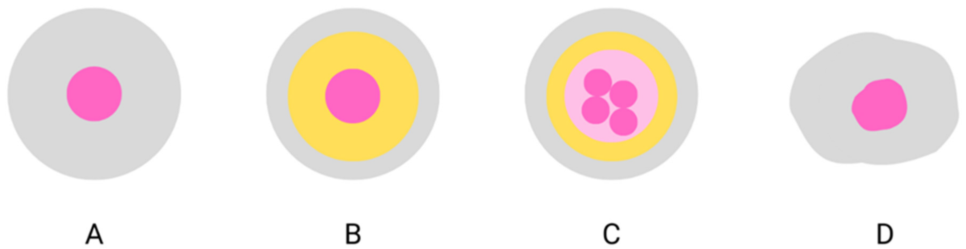

Figure 1. Schematic illustration of various morphologies formed by microencapsulation: monolayer and mononuclear microcapsule (**A**), multilayer and mononuclear microcapsule (**B**), multilayer and multinuclear microcapsule (**C**), and microparticle (**D**), own elaboration [35].

3. Complex Coacervation of Essential Oils

The process of microencapsulating essential oils by complex coacervation occurs in stages. The first one is hydration and preparation of wall materials solution. The essential oils and an emulsifier (e.g., Tween 20, 60, 80) are then added to these solutions, and the emulsification process is carried out. This step is essential because the quality of the emulsion and the interaction between its droplets directly affect the stability of the microcapsules produced later. The final step is curing by spray drying or freeze-drying [37][38].

Microencapsulation of essential oils using complex coacervation allows for their controlled release. It is the process of delivering EO delayed after administration or incorporation into the food matrix for an extended period [37][39]. This process is influenced by environmental conditions (type of food matrix), type of EO, the composition of microcapsules (proteins, polysaccharides), and microcapsule architecture. Essential oils can be released from their microcapsules through a variety of mechanisms. The swelling mechanism involves increasing the pore size as the matrix swells, which promotes the release of the encapsulated Eos. The mechanism of erosion is the dissolution of the outer part of the support (surface erosion) or all of the support (bulk erosion), often due to enzymatic or chemical hydrolysis. The mechanism of fragmentation is the rupture or breakage of the support matrix, which often occurs due to mechanical forces. The resulting increase in surface area and shorter diffusion paths mean that the bioactive agents are released from the fragments faster than the original carrier. The diffusion mechanism involves the diffusion of the bioactive component through the carrier matrix into the surrounding environment. EO microcapsules should be designed to be used in a specific product because food is exposed to a variable temperature, ionic strength, pH and mechanical conditions and stress during processing and storage [40][41][42][43]. The preparation of microcapsules of essential oils produced by complex coacervation can be an effective method to preserve their physicochemical properties. At the same time, it can contribute to increasing the applicability of essential oils in food as natural additives with a preservative effect and increasing the nutritional value of the final product. EO microcapsules can be an effective way to reduce the use of synthetic food additives while enabling the creation of interesting products placed in the functional food segment.

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