PRESENT STATUS OF MEMBRANE EMULSIFICATION AND ENCAPSULATION BY SPRAYDRYING OF VEGETABLE OILS WITH INCREASED ACTIVE INGREDIENT CONTENT – A REVIEW

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Abstract

Vegetable oils such as flaxseed oil, olive oil and soybean oil are a common source of bioactive ingredients. In addition to basic nutrition, they are consumed for their various functional properties. Consequently, they are being used in the preparation of safe products with a positive impact on consumer health. Nevertheless, they are chemically unstable and susceptible to degradation.

Microencapsulation is an effective and important tool to prepare oil-based high-quality and health-beneficial products in various industries in order to enhance their chemical, oxidative, and thermal stability. Emulsification technology is a key step in the microencapsulation of oils. It is generally applied for the encapsulation of bioactive ingredients in aqueous solutions, which can either be used directly in the liquid state or can be dried to form powders after emulsification.

Our research is directed toward the use of microencapsulation technology to encapsulate different oils by varying techniques. In this way, the oils' off-flavor can be masked and safety, quality, and the nutritional value of the product can also be improved. In addition, more than one microencapsulation technique can be utilized in the process. Future suggestions include the purification and use of factory waste as a wall material, thereby reducing the economic cost of the encapsulation process.

Keywords: Membrane emulsification, vegetable oil, bioactive ingredient, wall materials, microencapsulation, Spray drying.

1. Introduction

Vegetable oils such as flaxseed oil, olive oil and soybean oil are a common source of bioactive ingredients such as plyohenols, omega-fatty acids, phytosterols and tocopherols. However, besides their basic nutrional values, they are consumed for various functional properties such as antioxidant activity, anti-inflammatory, antivasoconstrictive, anti-arrhythmic, antithrombotic, antimicrobial, antihypertension, antiulcer, anti-aging, anticancer,

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antidiabetic, antidepressant, antipyretic antitussive, antidandruff, antinociceptive, and insect repellent activities (Bakry et al., 2016).

Therefore, because of their high instauration degree, these vegetable oils are subject to oxidation during processing, distribution and handling. Oxidation leads to the formation of unpleasant tastes and odors and, consequently, to the reduction of oil's shelf life, besides promoting the generation of free radical, which may have negative physiological effects on the organism. Thus, one of the solutions to preserve these bioactive is the microencapsulation: Microencapsulation is a method in which tiny particles or droplets are surrounded by a coating wall, or are embedded in a homogeneous or heterogeneous matrix, to form small capsules. The key step to release capsules is the emulsification technology. It is generally applied for the encapsulation of bioactives in aqueous solutions (Đorđević et al., 2014)

Basically, an emulsion consists of at least 2 immiscible liquids, usually oil and water, with one of the liquids being dispersed as small spherical droplets in the other. A system that consists of oil droplets dispersed in an aqueous phase is called an oil-in-water (O/W) emulsion, whereas a system that consists of water droplets dispersed in an oil phase is called water-in-oil (W/O) emulsion. Multiple emulsions, such as oil-in-water-in-oil (O/W/O) or water-in-oil-in-water (W/O/W) emulsions have also been developed. Dispersion by artificial membranes, or as widely known Membrane Emulsification, is a novel unit operation for producing single and multiple emulsions or complex suspension/emulsion fluids. The dispersed phase is forced through the pores of microporous membrane directly into the continuous phase by using low pressure. Emulsified droplets are formed and detached at the end of the pores with drop by drop mechanism (Joscelyne & Trägårdh, 2000).

The purpose of this article is to provide a state of the art review on the membrane emulsification process for food applications. We first recall the main aspects of the membrane emulsification process, and then we present the main studies on the preparation of encapsulated vegetable oils by membrane emulsification and their applications.

2. Emulsions and emulsification technology

Emulsions are liquid systems composed of two immiscible phases referred to as continuous and disperse phase, respectively. In O/W emulsions, the oil is the dispersed phase and water is the continuous phase. Alternatively, in water-in-oil emulsions (W/O), the water is dispersed phase and the continuous phase is oil. Multiple emulsions can also be produced, like water-in-oil-in-water (W/O/W) or oil in-water-in-oil (O/W/O) emulsions. However, one characteristic that all emulsions have in common is that they are thermodynamically unstable. To enhance

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the stability of the emulsions a specific type of ingredients, generally referred as stabilizers, can be used. The stabilizers can be divided into two categories, named emulsifiers or texture modifiers, depending on its stabilization mode. Emulsifiers are surface-active molecules that absorb to the surface of freshly formed droplets, while texture modifiers enhance emulsion stability by increasing the viscosity of the continuous phase. Nevertheless, there are some physicochemical mechanisms that are responsible for emulsion instability, being the most important sedimentation, flocculation, coalescence and phase inversion (Dickinson, 2009; DJ McClements 2015).

2.1.Membrane emulsification ME

The process of membrane emulsification (ME) is defined as forcing the dispersed phase, e.g. a vegetable oil, to permeate through the pores of a microporous membrane using a low pressure.The continuous phase flows along the membrane surface providing the shear stress that will cause the droplets detachment. To maintain the stability of the emulsion and avoid the coalescence of the droplets, one or more surface active agents are generally used (Zanatta et al., 2017)

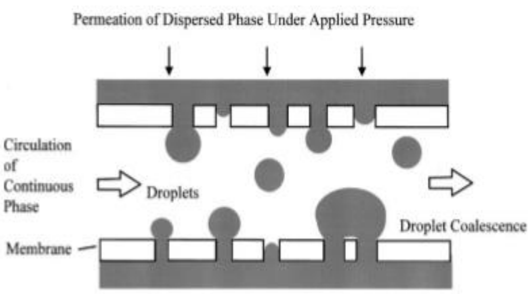

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Figure 1: Membrane emulsification process.

This method is known as the simplest technique; demands lower energy and needs less surfactant. It is applicable for both oil in water and water in oil emulsions. Besides its narrow resulting droplet size distributions, it is known by its lower surfactant need (Charcosset, 2009; Joscelyne & Trägårdh, 2000).

2.2.ME process controlling parameters

ME is influenced by several parameters including membrane pore size and distribution, membrane porosity, membrane surface type, emulsifier type and concentration, dispersed phase flux, velocity of the continuous phase and transmembrane pressure(Joscelyne & Trägårdh, 2000). This parameters are classified by Charcosset, 2009 into membrane parameters, phase parameters and process parameters. Membrane parameters include mean

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pore size, pore size distribution, pore shape, number of active pores, porosity, wettability, permeability K and thickness L. Phase parameters include interfacial tension, emulsifier type and concentration, viscosity and density of continuous and dispersed phase. Process parameters include wall shear stress, transmembrane pressure , temperature, membrane module configuration. Their influence of the emulsifying process (droplet size distribution, dispersed phase flux, dispersed phase percentage).

3. Technologies for production of microcapsules

The food industry is more and more focused in developing healthier foods by re-designing the production processes and enriching the foods with bio compounds.Microcapsules or microspheres are widely used in many different applications in chemical, food, cosmetic and pharmaceutical industries to encapsulate protect and deliver bioactive compounds.

3.1.Reasons of application of microencapsulation

There are many reasons for applying microencapsulation in food industry(Desai & Jin Park, 2005;Gharsallaoui, Roudaut, Chambin, Voilley, & Saurel, 2007):

-to reduce the core reactivity with environmental factors such as heat, air and moisture;

-to decrease the transfer rate of the core material to the outside environment;

-to promote easier handling;

-to control the release of the core material;

-to mask the core taste;

-to dilute the core material when it should be used in only very small amounts;

-to separate components within a mixture that would otherwise react with one another.

3.2.Encapsulation processes

Many encapsulation processes such as: spray-drying, spray-cooling, spray-chilling, air suspension coating, extrusion, centrifugal extrusion, freeze-drying, coacervation, rotational suspension separation, co-crystallization, liposome entrapment, interfacial polymerization, molecular inclusion(Gharsallaoui et al., 2007).In our research, we will be focusing in spray drying method.

Spray drying

Microencapsulation by spray-drying has been successfully used in the food industry for several decades, and this process is one of the oldest encapsulation methods used since the 1930s to prepare the first encapsulated flavors using gum acacia as wall material (Calvo, Hernández, Lozano, & González-Gómez, 2010).

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Various oils in the food industry where encapsulated successfully using this technique. Recently, several studies have been under- taken to encapsulate the oils by spray-drying in food(Bakry et al., 2016; Carneiro, Tonon, Grosso, & Hubinger, 2013; Fernandes, Borges, & Botrel, 2014; Quang & Le, 2017; Rubilar et al., 2012; Zhu, 2017), cosmetics (Carvalho, Silva, & Hubinger, 2014), pharmaceutical (Liu and Yang 2011), and pesticide industries(López et al., 2014).

Food industry refers to spray drying in order to ensure a microbiological stability of products, avoid the risk of chemical and/or biological degradations, reduce the storage and transport costs, and finally obtain a product with specific properties like instantaneous solubility for example. It is also used due to its low cost and available equipment..

The application of spray-drying process in microencapsulation involves three basic steps:

1- Preparation of the dispersion or emulsion to be processed;
2- homogenization of the dispersion;
3- Atomization of the mass into the drying chamber.

The mechanism of this technique includes the homogenization of the material for encapsulation with the carrier material at a different ratio. The mixture is then fed into a spray dryer and atomized with a nozzle or spinning wheel. Water is evaporated by the hot air contacting the atomized material. The microcapsules are then collected after they fall to the bottom of the drier.

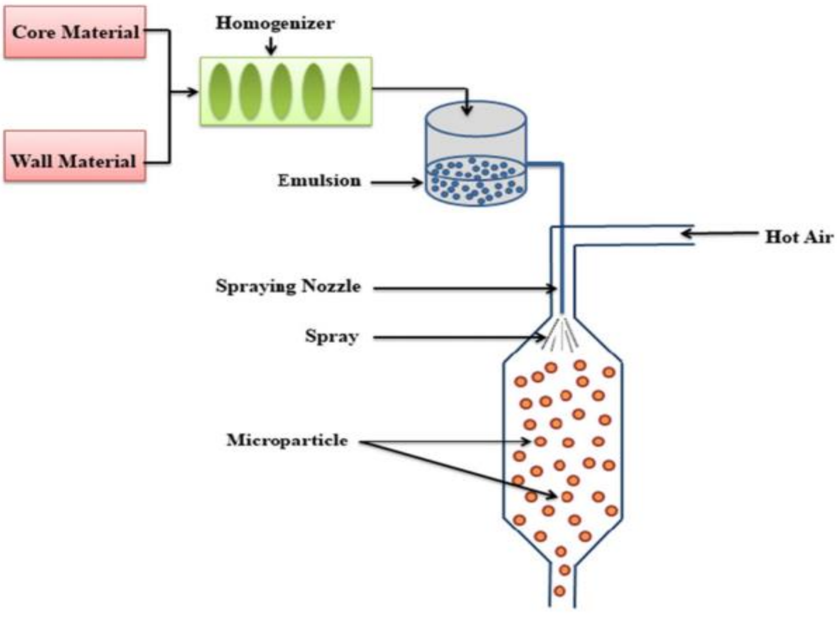

Figure 2: Schematic representation of the microencapsulation process by spray-drying (Bakry et al., 2016).

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Spray drying limitations

One limitation of the spray-drying technology is the limited number of wall materials available which are expected to have good water solubility. Typical wall materials include gum acacia, maltodextrins, hydrophobically modified starch, and mixtures of them. Other polysaccharides (alginate, carboxymethylcellulose, guar gum) and proteins (whey proteins, soy proteins, sodium caseinate) can be used as the wall material in spray-drying, but their usage becomes very tedious and expensive because of their low solubility in water. Another limitation is that spray drying provides fine microcapsules powder which needs further processing such as agglomeration. In addition, Encapsulated materials havelower oxidative stability due to the high temperatures used during the atomization process(Bakry et al., 2016; Gharsallaoui et al., 2007).

4. Applications of emulsified and microencapsulated vegetable oils

Various applications of microencapsulated vegetable oils have been found in the fields of foods, pesticides, textiles, and pharmaceuticals.

Food applications

Food fortified with ω-3 fatty acids could be added for the formulation of dairy products, infant food products, meat and pastas. The stability of the oil in the product remains the major challenge. For example, Rubilar et al., 2012studied the incorporation of encapsulated linseed oil in an optimized formulation of soup. The oxidative stability was enhanced and made it possible to provide a source of ω-3 besides obtaining a value-added product with high consumer acceptability.

Elsewhere, olive oil in lemon juice emulsion microcapsules prepared by freeze-drying were used successfully in instant salad sauce (Silva, Coelho, Calado, & Rocha-LeAão, 2013).

Carneiro, Tonon, Grosso, & Hubinger, 2013, showed thatencapsulation efficiency was higher using a mixture of maltodextrin, whey protein concentrate and modified starch as wall materials which results in good oxidative stability.

Fuel industry

Vegetable oils represent promising alternatives to diesel engine fuel because they can be obtained from different feedstocks and renewable sources; also their properties are close to diesel fuel. Emulsification techniques were used by (Melo-Espinosa, Piloto-Rodríguez, Goyos-Pérez, Sierens, & Verhelst, 2015) in order to obtain emulsified biofuels (emulsions or

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microemulsions). Therefore, it was possible to lower viscosity and improve the atomization through this method to obtain a more engine-friendly fuel.

Cosmetics

The incorporation of oils in an encapsulation system could be a good approach to preserve the bioactive compounds. For example the green coffee oil which has emollient properties and absorption capacity of solar UVB radiation could be applied in cosmetic products. When this oil is encapsulated, it will not be in direct contact with the human skin. This reduces allergenic effect of cinnamic acid, besides allowing a gradual release. Consequently, the microencapsulation by spray drying of this oil is an alternative to transform it in powder, maintaining its active compounds, besides facilitating the handling and application in other products(Carvalho et al., 2014).

Pharmaceutical industry

Evening primrose oil (EPO) (Oenothera biennis) is becoming used in increasing amounts in nutritional and pharmaceutical preparations.The bioactive ingredient of this oil is the gamma-linolenic acid, responsible for many physiological effects, such as hypocholesterolemic effect, therapeutic effects in atopic eczema, diabetic neuropathy, rheumatoid arthritis and premenstrual pains, etc. That ‘s why Liu & Yang, 2011, have optimized the encapsulation of this oil.

Pesticide industry

Encapsulated essential oils could be used to retard postharvest decay which is a significant factor that limits the storage life of fruitsand vegetables causing considerable economic losses. They may be used as alternatives offungicides and pesticides to avoid their harmful impacts on the environment and human health, and the development of fungicide resistance by pathogens. Recently, essential oilsextracted from plants having repellent properties have been studied as replacements for chemical pesticides due to their environmentally friendly and biodegradable status(Bakry et al., 2016). Studies carried by López et al., 2014, showed that the microencapsulation procedure of *Schinus molle* essential oil could be an interesting strategy to obtain a botanical insecticide to control the release of its active ingredient. In this way, essential oils are protected from the external environment during product application and storage time, providing a more persistent insecticidal effect.

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5. Conclusions

Membrane emulsification which is a key step for the microencapsulation has been shown to be a relatively simple and reliable process for making emulsions. The resulting droplets size is dependent on the size of the membrane pores. Microencapsulation involves the incorporation of food ingredients in small capsules. It is useful to protect sensitive food components, to mask or preserve flavours and to transform liquids to easily handled solid ingredients. It is an effective and important tool to prepare oil based high quality and health beneficial products for application in various industries such as food industry, fuel, pesticide, cosmetics and pharmaceutical industry.

Innovation in this field could be directed toward the use of microencapsulation technology to encapsulate a mixture of different oils by varying techniques. In this way, the oils' off-flavour can be masked and safety, quality, and the nutritional value of the product can also be improved. Another novelty includes the purification and use of factory waste as a wall material, thereby reducing the economic cost of the encapsulation process.

Finally, another idea is to form antimicrobial edible films and coatings which contain microencapsulated oil, antimicrobial agent, and which prolong the shelf-life of food products. This could help decreasing the economic losses, especially for the case of cheeses products that are susceptible to fungal attack during ripening and storage.

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References

Bakry, A. M., Abbas, S., Ali, B., Majeed, H., Abouelwafa, M. Y., Mousa, A., & Liang, L. (2016). Microencapsulation of Oils: A Comprehensive Review of Benefits, Techniques, and Applications. *Comprehensive Reviews in Food Science and Food Safety*, *15*(1), 143–182.

Calvo, P., Hernández, T., Lozano, M., & González-Gómez, D. (2010). Microencapsulation of extra-virgin olive oil by spray-drying: Influence of wall material and olive quality. *European Journal of Lipid Science and Technology*, *112*(8), 852–858.

Carneiro, H. C. F., Tonon, R. V, Grosso, C. R. F., & Hubinger, M. D. (2013). Encapsulation efficiency and oxidative stability of flaxseed oil microencapsulated by spray drying using different combinations of wall materials. *Journal of Food Engineering*, *115*(4), 443–451.

Carvalho, A. G. S., Silva, V. M., & Hubinger, M. D. (2014). Microencapsulation by spray drying of emulsified green coffee oil with two-layered membranes. *Food Research International*, *61*, 236–245.

Charcosset, C. (2009). Preparation of emulsions and particles by membrane emulsification for the food processing industry. *Journal of Food Engineering*.

Desai, K. G. H., & Jin Park, H. (2005). Recent Developments in Microencapsulation of Food Ingredients. *Drying Technology*.

Dickinson, E. (2009). Hydrocolloids as emulsifiers and emulsion stabilizers. *Food Hydrocolloids*, *23*(6), 1473–1482.

DJ McClements - 2015 - books.google.com.

Đorđević, V., Balanč, B., Belščak-Cvitanović, A., Lević, S., Trifković, K., Kalušević, A., … Nedović, V. (2014). Trends in Encapsulation Technologies for Delivery of Food Bioactive Compounds. *Food Engineering Reviews*.

Fernandes, R. V. D. B., Borges, S. V., & Botrel, D. A. (2014). Gum arabic/starch/maltodextrin/inulin as wall materials on the microencapsulation of rosemary essential oil. *Carbohydrate Polymers*, *101*(1).

Gharsallaoui, A., Roudaut, G., Chambin, O., Voilley, A., & Saurel, R. (2007). Applications of spray-drying in microencapsulation of food ingredients: An overview. *Food Research International*.

Joscelyne, S. M., & Trägårdh, G. (2000). Membrane emulsification — a literature review. *Journal of Membrane Science*, *169*, 107–117.

Liu, T. T., & Yang, T. S. (2011). Optimization of Emulsification and Microencapsulation of Evening Primrose Oil and Its Oxidative Stability During Storage By Response Surface

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Methodology. *Journal of Food Quality*, *34*(1), 64–73.

López, A., Castro, S., Andina, M. J., Ures, X., Munguía, B., Llabot, J. M., … Domínguez, L. (2014). Insecticidal activity of microencapsulated Schinus molle essential oil. *Industrial Crops and Products*, *53*, 209–216.

Melo-Espinosa, E. A., Piloto-Rodríguez, R., Goyos-Pérez, L., Sierens, R., & Verhelst, S. (2015). Emulsification of animal fats and vegetable oils for their use as a diesel engine fuel: An overview. *Renewable and Sustainable Energy Reviews*.

Quang, N., & Le, *. (2017). *Combination of whey protein and carbohydrate for microencapsulation of pumpkin (Cucurbita spp.) seed oil by spray-drying*. *International Food Research Journal* (Vol. 24). Retrieved from

Rubilar, M., Morales, E., Contreras, K., Ceballos, C., Acevedo, F., Villarroel, M., & Shene, C. (2012). Development of a soup powder enriched with microencapsulated linseed oil as a source of omega-3 fatty acids. *European Journal of Lipid Science and Technology*, *114*(4), 423–433.

Silva, K. A., Coelho, M. A. Z., Calado, V. M. A., & Rocha-LeAão, M. H. M. (2013). Olive oil and lemon salad dressing microencapsulated by freeze-drying. *LWT - Food Science and Technology*, *50*(2), 569–574.

Zanatta, V., Rezzadori, K., Penha, F. M., Zin, G., Lemos-Senna, E., Petrus, J. C. C., & Di Luccio, M. (2017). Stability of oil-in-water emulsions produced by membrane emulsification with microporous ceramic membranes. *Journal of Food Engineering*.

Zhu, F. (2017, August 15). Encapsulation and delivery of food ingredients using starch based systems. *Food Chemistry*. Elsevier Ltd.

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