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Emulgel as a transport and release system for clove oil (Syzygium aromaticum)

Emulgel como sistema de transporte y liberación del aceite de clavo de olor (*Syzygium aromaticum)*

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Abstract

Clove (Syzygium aromaticum) is a plant that represents a rich source of phenolic compounds such as eugenol, eugenol acetate, and gallic acid. It has great potential for industrial applications due to its antimicrobial, antiviral, anti-inflammatory, hepatoprotective, anti-stress, contraceptive, and anesthetic properties. The essential oils obtained from this plant have direct applications in medicine, aromatherapy, fragrances, and perfumes, among others. To apply this compound, a system is required that not only protects it but also allows its release, thus allowing its diffusion and absorption appropriately, such as creams, lotions, or gels. The compound is obtained by Soxhlet extraction. A prototype emulgel is proposed, using carboxymethylcellulose for the hydrogel formulation and castor oil for the oleogel formulation, placing the extracted oil in one of the phases. The product's physicochemical characteristics were determined, including color, odor, homogeneity and consistency, pH, conductivity, viscosity, swelling index, spreadability, and stability. When evaluating the system, it was found that the bioactive compound within the emulgel could be released under the action of an external stimulus.

Keywords: cloves, emulgel, release system

Resumen

El clavo de olor (Syzygium aromaticum) es una planta que representa una rica fuente de compuestos fenólicos tales como eugenol, acetato de eugenol y ácido gálico, y posee un gran potencial de aplicaciones industriales dada sus propiedades antimicrobianas, antivirales, antiinflamatorias, hepatoprotectivas, anti estrés, anticonceptivas y anestésicas. El uso de los aceites esenciales obtenidos de esta planta tiene aplicaciones directas en medicina, aromaterapia, fragancias, perfumes, entre otras. Para aplicar este compuesto se requiere de un sistema que además de protegerlo permita su liberación, permitiendo así su difusión y absorción de manera apropiada, como cremas, lociones o geles. El compuesto se obtiene mediante extracción Soxhlet. Se propone un prototipo de emulgel, utilizando carboximetilcelulosa para la formulación de hidrogel y aceite de ricino para la formulación de oleogel, colocándose en una de las fases el aceite extraído. Se determinaron las características fisicoquímicas del producto, incluyendo color, olor, homogeneidad y consistencia, pH, conductividad, viscosidad, índice de hinchamiento, extensibilidad y estabilidad. Al evaluar el sistema se encontró que el compuesto bioactivo dentro del emulgel pudo ser liberado bajo la acción de un estímulo externo.

Palabras clave: clavo de olor, emulgel, sistema de liberación

1 Introduction

Clove (*Syzygium aromaticum L.)* is an aromatic flower of the Myrtaceae family, containing approximately 15–20% essential oil. More than 30 compounds have been identified in this oil, where eugenol is the main compound (50–90%), and the complement (10–40%) is composed mainly of β-caryophyllene and α-humulene (Haro-González *et al*., 2023).

Research shows that clove essential oil contains eugenol (83.6%); eugenyl acetate (11.6%) and caryophyllene (4.2%) as major components (Aguilar & López, 2013). This oil represents one of the main vegetable sources of phenolic compounds such as flavonoids, hydroxybenzoic acids, hydroxycinnamic acids and hydroxyphenylpropenes. Also found are caffeic, ferulic, ellagic, and salicylic acids. Flavonoids such as kaempferol, quercetin, and their (glycosylated) derivatives are also found in cloves at lower concentrations (Cortes-Rojas *et al*., 2014).

Eugenol is attributed with antimicrobial activity. This compound can denature proteins and simultaneously alter cell membrane permeability, resulting in the death of microorganisms. It also exhibits antioxidant capacity and free radical scavenging activity, forming complexes with reduced metals. It is also attributed with anti-inflammatory, antimutagenic, antiallergic, analgesic, and antitumor properties (Zari & Hamkeen, 2021).

Numerous physical and pharmacological benefits are attributed to it; in dentistry it is used in patients with toothache, pulpitis and dental hyperalgesia. (Gülçin, 2011). In the cosmetics industry it has been used for the treatment of skin infections, skin lesions and inflammatory disorders. (Nejad *et al*., 2017). However, the volatile nature, hydrophobicity and its impact on the organoleptic properties make it difficult to directly use this oil in food or pharmaceutical products (Franlyne *et al*., 2019).

Gels are a relatively new dosage form created by entrapment of large amounts of aqueous or hydroalcoholic liquid in a network of solid colloidal particles. Gel formulations generally provide a faster release of a compound compared to conventional ointments and creams. (Hiba *et al*., 2018). Emulgels are generally formed by dispersing an oil phase in a gel phase or by inducing gelation of the external phase of an oil-in-water emulsion, it is the approach that uses the help of both emulsion and gels, obtaining the double effect of controlled release (Kumar *et al*., 2016). This research proposes a prototype emulgel as a transport and release system for clove oil.

2 Experimental Procedure

2.1 Extraction and physicochemical characterization of clove oil

The extraction was performed using Soxhlet equipment. A sample was taken and placed in an extraction cartridge, in a 1:20 ratio with the solvent, using petroleum ether. The extraction was carried out at a circulation rate of 6 drops per minute for 6 hours. Once the extraction was complete, the flask was removed and transferred to a rotary evaporator to remove the solvent. The oil obtained was characterized as follows.

The refractive index of the extracted oil was determined using a refractometer (Thermo brand) according to COVENIN (702:2001). As well as its solubility, as proposed by Scotti *et al*., (2020), who used water and linear alcohols of different carbon numbers (C2-C7) by adding oil and solvent in a 1:3 ratios to a test tube, verifying its solubility or not.

The total polyphenol content was determined according to the Folin-Ciocalteu colorimetric method (analytical grade, Merck). For which a volume of 50 µL of the extract was taken to which 125 µL of the Folin-Ciocalteu reagent was added, the mixture was stirred and left to stand for 8 minutes. Subsequently, 400 µL of sodium carbonate (7.1% Na_2CO_3) was added and the solution was topped up with distilled water to 2,000 µL. After 1 hour of standing in the dark, the absorbance at 760 nm was read using a Genesis 20 UV/VIS spectrophotometer (Thermo Scintific, Waltham, Massachusetts, USA). A calibration curve was prepared with a Gallic Acid (Polyhydroxylated Organic Acid) standard. (Sigma-Aldrich, Berlín Alemania) at concentrations of 50, 100, 200, 300, 400, 500 and 1000 ppm.

The 2,2-diphenyl-1-picrylhydrazyl (DPPH) free radical method (Sigma Aldrich Co®) was used for antioxidant capacity determination using a 100 µM DPPH solution in 80% methanol. A glass cuvette contained 100 µL of extract and 2.9 mL of DPPH. Absorbance was monitored every 5 min for 30 min at a wavelength of 515 nm. Reference absorbance (A_0) was obtained by replacing the extract volume with 80% methanol.

2.2. Prototype of emulgel-type transport system. Characterization

For the development of the product prototype, carboxylmethylcellulose (CMC) was selected to form the gel in the aqueous phase, while castor oil was used for the oil phase. The emulsifiers used were Tween 80 and Span 20. The phase ratio was 9:1 (hydrogel:oleogel). The emulsifier was formed by mixing the hydrogel and oleogel using an UltraTurrax at 3500 rpm for 20 s.

The prototype was inspected for physical appearance, including color, homogeneity, and consistency. Its pH and conductivity were determined using a Digimed conductivity meter. A stability study was carried out by subjecting the product to mechanical stress, using a centrifuge at 500 rpm for 5 minutes, then at 1500 rpm for 0 minutes. If no significant changes were observed, the centrifuge was centrifuged at 3000 rpm for 15 minutes.

2.3. Evaluation of the oil release system incorporated in the emulgel

1 g of the emulgel was dissolved in an appropriate solvent and filtered to obtain a clear solution. A standard solution was prepared with a sample of commercial eugenol in the same solvent and a solution with the extracted oil. The concentration of oil in the product is determined by UV-visible spectrophotometry. The calibration curve was prepared with a commercial eugenol standard solution at concentrations of 5, 10, 15, 20 and 25 ppm at a wavelength of 280.5 nm.

3 Discussion and Results

3.1 *Physicochemical characterization of the oil*

The clove oil obtained was characterized by its appearance, as well as its refractive index as shown in table 1., which displays a characteristic odor, yellowish to brown color, pungent flavor, and oily texture, Nejad *et al*., (2017) points out that this oil acquires a brown color with aging or in contact with air, it is photosensitive and thermolabile, so its storage life is short if not properly protected.

Clove contains 15–20% volatile essential oil; 10–13% tannin (gallotanic acid), resin, chromone, and eugenin. The essential oil of clove buds contains eugenol (70–90%), eugenol acetate (2–17%), and caryophyllene as its main component (Misar *et al*., 2020).

The density of most oils is less than 1 g/cm³, which means they are less dense than water and will float on it, the presence of phenols or derivatives affect the density value. The refractive index obtained is within the range reported by Nejad *et al*., (2017) and Valladolid (2016) who indicate that the refractive index of this oil is between 1.515 and 1.535. Misar *et al.,* (2020) reports a refractive index1.529.

The oil is insoluble in water, partially soluble in ethanol, and soluble in linear alcohols (C2 to C7), given the structure of the predominant compounds in the extract, such as the presence of eugenol (Nejad, Özgünes, & Basaran, 2017). As the molecular weight of the alcohols increases, a decrease in polarity is observed. This can be attributed to the predominance of the nonpolar part of the molecule, i.e., the hydrocarbon chain, and the influence of the OH decreases, as is the case with the eugenol molecule.

Table 1. Physical and organoleptic characteristics of the essential oil

Parameters	Value obtained
Color	Dark yellow
Odor	Characteristic
Density (g /mL)	0,99 ± 0,05
refractive index	1,519 ± 0,001

Plant phenols are one of the main compounds that act as primary antioxidants or free radical scavengers. Eugenol, or 4-allyl-2-methoxyphenol, is a phenolic derivative composed of an aromatic hydroxyl nucleus. Table 2 shows the total phenol content found in the oil, as well as its antioxidant capacity. The principle of antioxidant activity is based on the availability of electrons to neutralize free radicals. Furthermore, antioxidant activity is related to the number and nature of the hydroxylation pattern on the aromatic ring. In general, the ability to act as a hydrogen donor and the inhibition of oxidation increase with the increasing number of hydroxyl groups on the phenolic ring. (Gülcin, 2011). Radünz *et al*. (2019) reported a percentage of 94.86% for the antioxidant capacity of this oil. In this study, the antioxidant and radical scavenging activities of eugenol were compared with those of a 6-hydroxy-2,5,7,8-tetramethylchromium-2-carboxylic acid solution.

Table 2. Physical and organoleptic characteristics of the essential oil

Parameters	Value obtained
Total polyphenols	(9321,93 ± 1,32)mg/L
Antioxidant capacity	(83,76 ± 0,32) %
Trolox	(96,43 ± 1,2) %

3.2. Characterization of the emulgel prototype

Emulgel consists of emulsions, either oil-in-water or water-in-oil, that gel when combined with a gelling agent. Emulgel is more effective in healing than conventional gel. Furthermore, this system features dual controlled release, attributed to both the emulsion and the gel (Malavi *et al.,*2022).

The product was formulated with organoleptic and rheological properties suitable for topical administration. An oil-in-water emulsion was proposed; the polymers and surfactants used to obtain the emulsion are used in the food and cosmetics industries. Upon mixing the aqueous and oily phases, a whitish product, a typical emulsion color, was obtained (Figure 1). The emulsion is homogeneous, with good spreadability and a characteristic clove odor due to its aromatic compounds.

Figure 1. Emulgel prototype

Spreadability is closely related to the ability to spread the product over a surface, Whader *et al*., (2018), who reported a capacity of 1,875 g.cm/s, for a carbopol-based aqueous external phase emulsifier, this value is higher than that reported in this research (1.68 ± 0.05) g.cm/s, due to the polymer used (carboxylmethylcellulose), however, the result shows that it spreads easily when applied to a surface or to the skin, providing a suitable delivery system for the extracted oil.

The product has a pH of 6.0, which falls within the skin pH range of 5.5–6.5. The advantage of being slightly acidic is that it cannot attack skin tissue and does not cause skin disorders (Arshad *et al*., 2020) when used in cosmetics. The conductivity (1722 µS/cm) allows us to verify that the external phase is aqueous, based on the measurement of free ions and water present in the sample. The value obtained is higher than water, due to the presence of other components in the product, which causes a grouping of the globules in emulsion and the movement or exchange of ions within the aggregates (Mehta, Gurpeet, Bhasin, 2010). Regarding its viscosity, Wadher *et al*., (2018) point out that the viscosity of emulgel formulations that have more than 1% of gelling agent exceeds 10,000 cP, with that of this manufactured product being 19,500 cP, this value is in accordance with that described by the researcher.

Another characteristic that different gels have is their swelling capacity, due to their ability to form a three-dimensional polymeric network that allows them to absorb a large amount of liquid, swelling and considerably increasing their volume without losing their shape, until they reach their maximum degree of hydration or swelling index (Ramirez_*et al*., 2016). The swelling index in an emulgel is associated with the structure and composition, as well as the medium where it is found. The value obtained was (9.82 ± 0.01) %, with indexes of 11.18% reported in the literature (Kapadiya *et al*., 2016).

On the other hand, the stability of the product was measured, finding that at 500 rpm for 5 min, and then 1500 rpm for 15 min there is no phase separation, however, at 3000 rpm for 15 min small oil droplets were observed on the surface. The emulgel behaves as a viscoelastic fluid, where the droplets that form part of the elaborated product are perfectly organized forming a hexagonal network with a negligible film thickness of the continuous phase, and is capable of storing the deformation energy and returning to its initial shape, just like the behavior of an elastic solid, dissipating part of that energy as if it were a viscous fluid. By increasing the shear stress for a considerable time, a greater kinetics of the dispersed agent (oil) is produced in the solution and the molecules in the dispersion proceed to join or agglomerate and the process known as flocculation is formed, where there is an increase in the drops due to their grouping, which leads to a breaking of the emulsion forcing it to a phase separation and due to the difference in densities, the oil being of lower density than the aqueous solution, a migration of the drops on the surface takes place (Nava, 2017).

3.3. Evaluation of the release system of the bioactive compound incorporated in the emulgel

The release of the compound was quantified by spectrophotometry showing its maximum absorbance at 280 nm (Olvera *et al*., 2019). A standard curve was constructed using commercial Eugenol as a standard. The emulgel was diluted, estimating an initial concentration of 200 ppm of eugenol in the clove extract. The emulgel dissolved and a concentration of 10.12 ppm was determined, which represented an absorbance of 0.408 a.u. This result shows that the compound trapped in the emulgel network can be released, suggesting that the system is capable of transporting and releasing the compound. The characteristics of the transport system can be modulated so that the compound is released under a certain external stimulus.

4 Conclusion

The resulting extract has a significant antioxidant capacity, providing excellent properties for use in cosmetic or pharmaceutical products. The designed prototype exhibits good properties as a topical product due to its good spreadability and pH. The proposed system is capable of transporting and releasing the compound present in cloves under external stimulation.

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