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Review Article

Supercritical Carbon Dioxide Extraction of Citronella Oil Review: Process Optimization, Product Quality, and Applications

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ABSTRACT

This review paper explores the utilization of supercritical carbon dioxide (SC-CO_2) extraction to isolate citronella oil, delving into its multifaceted dimensions, including process optimization, product quality enhancement, and diverse potential applications. Citronella oil, renowned for its myriad bioactive compounds with demonstrated health benefits, is a coveted essential oil in the pharmaceutical, cosmetics, and food industries. The transition from traditional extraction techniques to SC-CO_2 extraction presents a paradigm shift due to its manifold advantages, such as heightened yield rates, expedited extraction durations, and elevated product quality. However, the efficacy of SC-CO_2 extraction is intricately interwoven with an array of parameters encompassing pressure, temperature, flow rate, particle size, and co-solvent ratios. Accordingly, meticulous process optimization is indispensable in achieving the desired product quality while maximizing yield. Furthermore, the paper explores the extensive spectrum of potential applications for citronella oil, extending its reach into formulations with antimicrobial,

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insecticidal, and antioxidant properties. These applications underscore the versatility and commercial appeal of citronella oil. The review establishes SC-CO_2 extraction of citronella oil as a promising and sustainable alternative to conventional extraction methodologies, offering myriad applications across the pharmaceutical, cosmetics, and food sectors. This scholarly work provides valuable insights into the intricacies of process optimization and product quality. It outlines future perspectives and avenues for further exploration in SC-CO_2 extraction of citronella oil.

Keywords: Citronella, optimization, product quality, supercritical carbon dioxide

INTRODUCTION

The essential oil is extracted from the leaves and stems of certain species of citronella grass, including *Cymbopogon nardus* and *Cymbopogon winterianus*. The oil is frequently used to produce personal care products, fragrances, and insecticides due to its citrus-like aroma and insect-repelling properties (Wany et al., 2013). Citronella oil has a long history of use in traditional medicine and aromatherapy. Its popularity has increased in recent years as consumer interest in natural and organic products has increased (Carvalho et al., 2016). Traditional cultures have used citronella oil as an insect repellent for centuries, particularly against mosquitoes and other stinging insects (Tyagi, 2016). Geraniol, citronellol, and citronellal, all monoterpenes, are the active compounds in citronella oil responsible for their insect-repelling properties (Pohlit et al., 2011; Sarah et al., 2023). These compounds interfere with the insect's ability to locate its host, making it harder for insects to locate and bite humans and animals. Citronella oil is considered a safe and effective alternative to chemical insecticides, which can negatively impact the environment and human health.

Citronella oil is well-known for its aromatherapy benefits and insect-repelling properties. It is frequently used in fragrances, balms, and candles due to its fresh, invigorating aroma (Bell, 2012). Also believed to have a soothing effect on the mind and body, citronella oil is frequently employed in aromatherapy to promote relaxation and reduce tension. The oil is an alternative therapy for several illnesses, including migraines, muscle pains, and respiratory infections (Ali et al., 2015). Due to its popularity and versatility, citronella oil is now widely produced and used worldwide.

Steam distillation, solvent extraction, and cold pressing are the traditional techniques for extracting citronella oil (Hanif et al., 2019; Khan & Dwivedi, 2018). However, in recent years, supercritical fluid extraction has been a promising alternative method for extracting high-quality citronella oil (Sarah et al., 2023). Extraction of supercritical carbon dioxide requires its supercritical state, which possesses the properties of both a liquid and a gas. This technique is considered more efficient and environmentally favorable than traditional methods because it does not require toxic solvents or high temperatures, which can degrade the extracted oil's quality (Aziz et al., 2022; Arsad et al., 2023; Rizkiyah et al., 2023).

This review paper presents several notable novelties compared to previously published papers. Firstly, it focuses on applying SC-CO_2 extraction to obtain citronella oil, highlighting its distinct advantages over conventional extraction methods. Notably, it delves into the crucial aspect of process optimization, recognizing that parameters such as pressure, temperature, flow rate, particle size, and co-solvent ratio influence the extraction process, offering potential insights into optimizing this technique.

Furthermore, the paper emphasizes product quality enhancement as a significant outcome of SC-CO_2 extraction. It is particularly pertinent for industries like pharmaceuticals, cosmetics, and food, where high-quality citronella oil is in demand. The review also explores the extensive range of potential applications for citronella oil, including its use in antimicrobial, insecticidal, and antioxidant formulations, underscoring its versatility and commercial value.

In addition to its benefits, the paper acknowledges the challenges associated with SC-CO_2 extraction, such as the high equipment and operational costs, as well as the need for standardization in the extraction methods. This comprehensive discussion of advantages, optimization strategies, product quality improvements, challenges, and future prospects ensures that this work offers valuable insights into the SC-CO_2 extraction of citronella oil, making it a significant contribution to the existing body of knowledge in this field.

CITRONELLA OIL AND ITS BIOACTIVITIES

Essential and regular oils, often referred to as carrier or vegetable oils, exhibit distinct differences in origin, composition, uses, aroma, and volatility. Essential oils are derived from aromatic parts of plants and contain concentrated volatile compounds like terpenes and phenols, giving them unique scents and therapeutic properties (Bakkali et al., 2008). They are commonly used for aromatherapy, perfumery, and flavoring, and they are diluted for topical applications due to their potency (Aziz et al., 2018). In contrast, regular oils, obtained from various sources such as seeds and nuts, consist primarily of fatty acids and lack the volatile aromatic compounds in essential oils. They possess mild, neutral scents, making them ideal base oils to dilute essential oils for skin application and culinary and industrial purposes. Essential oils are highly volatile and evaporate quickly, which enhances their aromatic qualities. In contrast, carrier oils are non-volatile and provide a stable medium for dilution and application of essential oils or other practical uses.

Citronella oil is primarily produced in tropical regions, particularly Indonesia, Sri Lanka, India, and China. The oil is extracted from the leaves and stems of citronella grass, a tall perennial grass with long, thin leaves and a characteristic lemon-like scent. The traditional method of extracting citronella oil is through steam distillation (Hamzah et al., 2014). The plant material is placed in a distillation chamber and heated with steam. The steam causes the essential oil to evaporate and then condense into a liquid collected in a separate container. The resulting oil is filtered to remove impurities (Cassel et al., 2006).

Another method of extracting citronella oil is solvent extraction (Lim et al., 2021). This method involves using a solvent, such as hexane, to extract the oil from the plant material. The solvent is then removed from the oil using a distillation process, leaving behind pure citronella oil. Supercritical carbon dioxide extraction is a newer method of extracting citronella oil (Guedes et al., 2018). The carbon dioxide is used to extract the oil from the plant material, which is then separated from the carbon dioxide. Supercritical carbon dioxide extraction is considered more environmentally friendly than traditional extraction methods, as it does not require toxic solvents or high temperatures (Mohd-Nasir et al., 2021). The active compounds of citronella are shown in Table 1, and an explanation of its compounds is shown below. Citronella oil contains various compounds, including citronellal, geraniol, citronellol, limonene, and camphene. These compounds give citronella oil its characteristic aroma, insect-repelling properties, and other potential health benefits.

- Citronellal: Citronellal is the primary component of citronella oil, accounting for approximately 35%–45% of the oil's total composition (Li et al., 2021). It has a strong lemon-like scent and is often used in fragrances and personal care products. Citronellal has also been found to have antimicrobial properties, making it helpful in treating various infections (Timung et al., 2016).
- Geraniol: Geraniol is another significant component of citronella oil, accounting for approximately 10%–20% of the oil's total composition (El-Kholany, 2016). It has a floral, rose-like scent and is used in producing perfumes, soaps, and lotions. Geraniol has antioxidant and anti-inflammatory properties, potentially helpful in treating various inflammatory conditions (Ammar, 2023).
- Citronellol: Citronellol is present in citronella oil in smaller amounts, accounting for approximately 5%–15% of the oil's total composition (Sreenath & Jagadishchandra, 2012). It has a rose-like scent and is used to produce perfumes and personal care products. Citronellol has also been found to have antimicrobial and anti-inflammatory properties (Chen & Viljoen, 2010).
- Limonene: Limonene is a minor component of citronella oil, accounting for approximately 2%–4% of the oil's total composition

Table 1
Identification of interest compounds of citronella in the chromatogram oil by Silva et al. (2011)

Identification	Components	P(%)
1	Citronellal	**98**
2	Citronellol	**95**
3	Geraniol	**86**
4	β -Element	**95**
5	Germacrene-D	**99**
6	Endemol	**83**
7	Germacredien-5-ol	**91**

(Wijesekera et al., 1973). It has a citrus-like scent and is used in producing fragrances and cleaning products. Limonene has been found to have antioxidant and anti-inflammatory properties, as well as potential anti-cancer effects (Anandakumar et al., 2021).

- Camphene: Camphene is another minor component of citronella oil, accounting for less than 1% of the oil's total composition (Devi et al., 2021). It has a fresh, woody scent and is used in producing fragrances and flavorings. Camphene has been found to have anti-inflammatory properties and potential anti-cancer effects (Alesaeidi & Miraj, 2016).

SUPERCRITICAL CARBON DIOXIDE (SC-CO_2) OF CITRONELLA OIL

SC-CO_2 extraction is a technique for extracting essential oils from plant material. Carbon dioxide is a non-toxic, non-combustible, readily accessible solvent that can extract essential oils without leaving noxious residues or byproducts (Arumugham et al., 2021). When carbon dioxide is heated and pressurized beyond its critical point (73.8 bar and 31.1°C), it transforms into a supercritical fluid with a liquid's density and a gas's diffusion properties (Putra, Rizkiyah, Aziz, et al., 2023). In the case of citronella oil, SC-CO_2 extraction has proven to be an effective and eco-friendly technique for extracting essential oil from citronella grass (Sarah et al., 2023). The plant material is placed in a sealed extraction vessel and then exposed to carbon dioxide at a specific temperature and pressure. The supercritical carbon dioxide can permeate the plant material's cell walls, dissolving the essential oil and transporting it out of the extraction vessel (Sodeifian, Ardestani, et al., 2016; Sodeifian, Sajadian, & Ardestani, 2016).

SC-CO_2 extraction can selectively extract compounds from plant material, one of its benefits (Odunlami et al., 2022; Sodeifian et al., 2017). The solubility of various compounds in SC-CO_2 varies based on their chemical properties (Idham et al., 2021; Sodeifian, Ardestani, et al., 2018). For instance, compounds with high molecular weights and boiling points tend to be more soluble in SC-CO_2 than those with low molecular weights and boiling points (Aziz et al., 2022; Idham et al., 2021). In the case of citronella oil, SC-CO_2 extraction has proven effective at removing the oil's primary components, including citronellal, geraniol, and citronellol. It has also been discovered that the process produces a higher yield of essential oil than traditional steam distillation methods (Chai et al., 2020). In addition to its efficiency and selectivity, SC-CO_2 extraction of citronella oil has yielded a high-quality product with minimal chemical degradation (Odunlami et al., 2022). It is because neither high temperatures nor exposure to oxygen, which can cause chemical changes in the essential oil, are involved in the extraction process (de Melo et al., 2020). Overall, SC-CO_2 extraction is a prospective method for extracting citronella oil, as it provides a safe and environmentally friendly alternative to conventional extraction techniques. This method is advantageous for producing citronella oil for various applications, including fragrances, personal care products, and insect repellents, due to the possibility of selective extraction and the production of high-quality essential oils. The parameters' influence on SC-CO_2 for citronella oil is shown in Table 2.

Effect of Pressure

The pressure in SC-CO_2 extraction can positively and negatively affect oil recovery, including citronella oil. Increasing the pressure in SC-CO_2 extraction can increase the oil yield by increasing the solubility of the oil components in the supercritical fluid. SC-CO_2 behaves like a gas at low pressure and a liquid at high pressure, which makes it an excellent solvent for extracting various organic compounds, including essential oils (Silva et al., 2021). When the pressure increases, the supercritical fluid's density and solubility increase. This increase in density and solubility allows the SC-CO_2 to penetrate deeper into the plant material and dissolve more oil components (de Oliveira et al., 2019; Sodeifian et al., 2022). The pressure also influences the polarity of the SC-CO_2, which can affect its ability to dissolve different compounds (Ahmad et al., 2019).

In the case of citronella oil extraction, citronellal and geraniol are the major components of the oil, which are responsible for its aroma and insect-repellent properties. These components have relatively high solubility in SC-CO_2, and increasing the pressure can enhance their solubility and, hence, the oil yield (Manaf et al., 2013; Sodeifian, Sajadian, et al., 2018). The pressure can also affect the phase behavior of the supercritical fluid and the oil, leading to changes in the partitioning of the oil components between the plant material and the supercritical fluid (Machado et al., 2022). It can result in more efficient extraction of the oil components, leading to higher oil yield.

Reducing the pressure during SC-CO_2 extraction can reduce oil recovery for various reasons. Initially, as the pressure decreases, the density and solubility of the supercritical fluid decrease (Sathasivam et al., 2022). Consequently, SC-CO_2's ability to dissolve oil components decreases, resulting in a lower oil yield (Ongkasin et al., 2019; Sodeifian, Ardestani, et al., 2016). It occurs because the solubility of a compound in a supercritical fluid depends on the fluid's density, and a decrease in pressure causes a decrease in density, thereby decreasing solubility (Putra et al., 2022). Second, lowering the pressure can cause the supercritical fluid to expand, decreasing its concentration and density (Aziz et al., 2021, 2023; Putra, Rizkiyah, Aziz, Mamat et al., 2023; Putra, Rizkiyah, Aziz, Yunus et al., 2023; Wang et al., 2023). It can result in the formation of gas bubbles, which can inhibit the penetration of the supercritical fluid into the plant material and shorten the contact period between the supercritical fluid and the oil components. It reduces oil recovery. A decrease in pressure can also reduce the mass transfer coefficient, which is the rate at which the solute (oil components) diffuses from the solid matrix (plant material) into the supercritical fluid (Soh et al., 2019). The mass transfer coefficient is affected by the density and diffusivity of the supercritical fluid, and a decrease in pressure causes both of these variables to decrease. It may result in a slower extraction rate and a reduced oil yield (Kim & Lim, 2020; Putra, Aziz, et al., 2018).

Effect of Temperature

In SC-CO_2 extraction, increasing the temperature can increase the oil yield by reducing the oil's viscosity and increasing the diffusion rate of the supercritical fluid in the plant material (Leila et al., 2022). However, increasing the temperature beyond a certain point can reduce the oil yield due to various factors. In SC-CO_2 extraction, when the temperature increases, the oil's viscosity decreases, making it more straightforward for the supercritical fluid to permeate the plant material and dissolve the oil components. Therefore, it will increase the oil yield. Increasing the temperature can also increase the diffusion rate of the supercritical fluid in the plant material, facilitating the mass transfer of the oil components from the plant material to the supercritical fluid, resulting in a greater oil yield (Moges et al., 2022; Putra, Aziz, et al., 2018).

However, if the temperature increases beyond the optimal range, the oil yield may decrease due to several factors. First, high temperatures can lead to the thermal degradation of oil components, resulting in the formation of undesirable byproducts and the loss of desirable oil components (Aprodu et al., 2020). High temperatures can also increase the volatility of oil components, resulting in their loss during extraction (Muhammad et al., 2022). In addition, elevated temperatures can reduce the solubility of oil components in SC-CO_2, decreasing the extraction efficiency and, consequently, the oil yield (Sodeifian et al., 2022).

Depending on various factors such as the pressure, solvent-to-feed ratio, and the properties of the plant material being extracted, a decrease in temperature during SC-CO_2 extraction can either decrease or increase the oil yield. The oil's viscosity increases at low temperatures, making it more difficult for the supercritical fluid to penetrate plant material and dissolve oil components (Dhara et al., 2022). Therefore, it can reduce oil recovery by SC-CO_2. In addition, decreasing the temperature can reduce the diffusion rate of the supercritical fluid in the plant material, which delays the mass transfer of oil components from the plant material to the supercritical fluid, resulting in a decreased oil yield (Dhara et al., 2022). On the other hand, in certain conditions, a decrease in temperature can also increase oil yield. For instance, at low temperatures, the solubility of specific oil components in SC-CO_2 may increase, resulting in greater extraction efficiency and, consequently, a greater oil yield (Aziz et al., 2022). In addition, low temperatures can diminish the vapor pressure of oil components, thereby minimizing their loss during extraction (Aziz et al., 2020).

It should be noted that the effect of temperature on oil yield is highly dependent on the pressure of the extraction process. Due to the causes above, decreasing the temperature at moderate forces can reduce the oil yield. However, a temperature decrease at high pressures can increase oil yield as the solubility of oil components in SC-CO_2 rises with decreasing temperature (Putra et al., 2021). Consequently, the effect of temperature on oil yield during SC-CO_2 extraction is complex and highly variable. The temperature must be optimized alongside other extraction parameters to obtain the highest oil yield with desirable qualities. Depending on the pressure and other extraction parameters, the optimal temperature range for citronella oil extraction using SC-CO_2 is reported to be between 40 and 60°C, as shown in Table 2.

Table 2
Parameter conditions of SC-CO_2 extraction for citronella oil recovery

Conditions	Yield	Summary	References
T= 313.15 to 353.15 K P= 6.2, to 180.0 MPa	± 3%	essential oil more concentrated with rich active compounds. A high selectivity was obtained at 353.15K and 18.0MPa, with a pure	Silva et al. (2011)
Ext. time = 120 min P= 25 MPa T=35°C CO_2 flow=18 L/h	4.40%	The antioxidant and antimicrobial activities of SFE oil are better than hydro distillation. Alcohols and aldehydes were the main compositions in the essential oils.	Wu et al. (2019)
P= 20 MPa T=40 °C	3.5%	Higher pressures significantly increased yield.	Guedes et al. (2018)
P= 15 MPa T= 50 °C Ext. time = 180 min.	1.4%	temperature is the most significant factor in maximizing citronella oil yield	Salea et al. (2018)
P =11 MPa T= 50 °C	1.92%	Higher pressures significantly increased the yield	Rosli et al. (2007)
P= 20 MPa T=40 °C	1.674%	oil yield was increased with increasing the pressure and decreasing the temperature	Abd Manaf et al. (2013)

Effect of Flowrate

Depending on various factors, increasing the flow rate of the supercritical fluid can have positive and negative effects on the oil yield. Increasing the flow rate can increase the mass transfer of the oil components from the plant material to the supercritical fluid, resulting in higher extraction efficiency and yield (Pavlić et al., 2020). Furthermore, it can increase the penetration of the supercritical fluid into the plant material, resulting in a quicker diffusion of the oil components into the supercritical fluid (Dimić et al., 2021). Increasing the flow rate can also diminish oil recovery. Increasing the flow rate decreases the contact duration between the supercritical fluid and the plant material, which may result in insufficient oil extraction. In addition, increasing the flow rate may also increase the pressure loss across the extraction vessel, reducing extraction efficacy (Peng et al., 2020).

On the other hand, decreasing the flow rate can also lead to a decrease in the oil yield. Decreasing the flow rate can increase the residence time of the supercritical fluid in the extraction vessel, leading to a higher risk of thermal degradation of the oil components due to prolonged exposure to high temperatures (Dhakane-Lad & Kar, 2021). Notably, the effect of flow rate on oil yield is highly dependent on other extraction parameters such as pressure, temperature, and the ratio of solvent to feed. At high pressures, for instance, increasing the flow rate can result in a higher oil yield because the mass transfer rate of the oil components from the plant material to the supercritical fluid is already high (Argun et al., 2022). Increasing the flow rate can further improve it. However, increasing the flow rate may not inherently increase the oil yield at low pressures, as the pressure may limit the mass transfer rate (Rizkiyah et al., 2022; Soh et al., 2019).

Consequently, the influence of flow rate on oil yield in SC-CO_2 extraction is complex and highly variable. Optimizing the flow rate and other extraction parameters is essential to obtain the maximum oil yield with desirable qualities. According to reports (Table 2), the optimal flow rate range for citronella oil extraction using SC-CO_2 is 0.3 L/min, depending on the extraction pressure and other parameters.

Effect of Extraction Time

Depending on various factors, increasing the extraction time in SC-CO_2 extraction can also positively and negatively affect the oil yield. On the one hand, increasing the extraction time can increase the contact time between the supercritical fluid and the plant material, allowing the oil components to dissolve in the solvent for an extended period, resulting in a higher oil yield (Capuzzo et al., 2013; del Valle, 2015). Increasing the extraction time gives the supercritical fluid more time to penetrate the plant material and dissolve its oily components. On the other hand, increasing the extraction time can also reduce oil production because increasing the extraction time can increase the exposure of oil components to high-pressure and high-temperature conditions, potentially resulting in thermal degradation or oxidation of oil components, which can reduce the overall oil yield (Golmohammadi et al., 2018).

However, reducing the extraction time may also reduce the contact time between the supercritical fluid and the plant material, decreasing extraction efficiency and oil yield (Park et al., 2007; Wu et al., 2019). Therefore, the optimal extraction duration for SC-CO_2 extraction of citronella oil highly depends on the specific extraction conditions, such as pressure, temperature, and solvent-to-feed ratio. Depending on the pressure and other extraction parameters, the optimum extraction time for citronella oil extraction using SC-CO_2 is typically reported to be between 30 minutes and 3 hours (Table 2).

Effect of Particle Size

In the SC-CO_2 extraction of citronella oil, the particle size of the plant material has a complex effect on the oil yield. However, these parameters have yet to be investigated by previous research. Therefore, this review will suggest that the effect of particle size in the extraction process can be applied to extract the citronella oil by SC-CO_2. When the particle size of the plant material is increased, the surface area of the material exposed to the supercritical fluid is decreased, which may decrease oil yield due to inefficient contact between the plant material and the solvent (Snyder et al., 1984; Sodeifian, Saadati Ardestani, et al., 2016). A larger particulate size will have a lower ratio of surface area to volume, resulting in lower extraction efficiency and, consequently, a lower oil yield. On the other hand, increasing the particle size can also increase the porosity of the plant material, allowing for greater solvent penetration and mass transfer of oil components from the plant material to the solvent (Uwineza & Waśkiewicz, 2020). It can result in a greater oil yield due to increased extraction efficacy.

Similarly, reducing the particle size can increase the surface area of the plant material, which can result in improved contact between the plant material and the solvent and, consequently, a greater oil yield (Jha & Sit, 2022). However, reducing the particle size can also increase the flow resistance of the supercritical fluid, which can reduce the mass transfer rate of oil components from the plant material to the solvent, resulting in a lower oil yield (Yousefi et al., 2019). Consequently, the optimal particle size for SC-CO_2 extraction of citronella oil depends on several variables, such as the extraction conditions, pressure, temperature, and solvent-to-feed ratio. SC-CO_2 extraction of plant material typically employs a particle size range of 0.5 to 3 mm (Putra, Rizkiyah, et al., 2018).

Effect of Co-solvent

In the SC-CO_2 extraction of citronella oil, using co-solvents can have complex effects on oil yield. However, these parameters have also yet to be investigated by previous research. Increasing the co-solvent ratio increases the solubility of the oil components in the supercritical fluid, resulting in a more significant mass transfer of the oil components from the plant material to the solvent and a higher oil yield (Sodeifian, Sajadian, & Saadati Ardestani, 2016). Co-solvents can also reduce the viscosity of SC-CO_2, thereby increasing the mobility of the solvent within the plant material and further improving extraction efficiency and oil yield (Benelli et al., 2010). The type of solvents that can be used in SC-CO_2 extraction is shown in Table 3. One of the safe co-solvents used in SFE for essential oils is ethanol. Ethanol is generally recognized as safe (GRAS) by the Food and Drug Administration (FDA) and is a common solvent in the food industry (Aziz et al., 2021). Ethanol has been used as a co-solvent in SFE for essential oils, and it has been found to increase the yield and quality of the extracted oil. However, using ethanol as a co-solvent in SFE requires careful optimization of the extraction parameters, such as temperature, pressure, and flow rate, to avoid denaturation of the essential oil components.

Table 3
Common co-solvent can be applied to assist SC-CO_2 extraction

Co-solvent	Chemical Formula	Source
Ethanol	C_2H_5OH	Daud et al., 2022
Methanol	CH_3OH	Marcus, 2018
Propanol	C_3H_7OH	Akkarawatkhoosith et al., 2019
Ethyl acetate	$C_4H_8O_2$	Klein et al., 2019
Acetone	C_3H_6O	Byun, 2020
Dimethyl sulfoxide (DMSO)	C_2H_6OS	Zhou et al., 2022
Carbon disulfide	CS2	Daneshyan and Sodeifian, 2022

However, an excessive quantity of co-solvent can cause the density and solvation power of the SC-CO_2, thereby decreasing the solubility of the oil components in the solvent and resulting in a reduced oil yield (Ahmad et al., 2019; Moges et al., 2022). Increasing the co-solvent ratio may also increase the cost of the extraction process, thereby reducing the process's economic viability (Hayyan et al., 2022). Similarly, diminishing the co-solvent ratio can reduce the oil components' solubility in SC-CO_2, reducing oil yield. However, diminishing the co-solvent ratio can reduce the cost of the extraction process, thereby increasing the process's economic viability. In the SC-CO_2 extraction of plant materials, including citronella, a co-solvent ratio of 5% to 20% is typically used (Machmudah et al., 2006).

BIOACTIVITY OF CITRONELLA OIL

Citronella oil is known to have a range of bioactive properties that have been the subject of research for many years. Some of the most commonly studied bioactivities of citronella oil include:

- Insect repellent: Citronella oil is well-known for its insect-repelling properties, particularly against mosquitoes. It is effective in repelling several species of mosquitoes and has been used as a natural alternative to synthetic insect repellents (Nollet & Rathore, 2017).
- Antimicrobial: Citronella oil has exhibited antimicrobial activity against various bacteria and fungi (Nakahara et al., 2013). It is particularly effective against Gram-positive bacteria such as *Staphylococcus aureus*, *Bacillus subtilis*, and *Streptococcus pneumoniae*.
- Antioxidant: Citronella oil has been found to possess significant antioxidant activity, which can help to protect the body from oxidative damage caused by free radicals (Sinha et al., 2011). This activity may be due to phenolic compounds in the oil.
- Anti-inflammatory: Citronella oil has been found to exhibit anti-inflammatory

activity in both in vitro and in vivo studies. This activity may be due to geraniol and citronellal (Salaria et al., 2020).

- Analgesic: Citronella oil has been shown to possess analgesic properties, which can help to reduce pain and inflammation (Ganjewala, 2009). This activity may be due to compounds such as citronellal, which have been found to exhibit analgesic effects in animal studies.
- Anti-cancer: Some studies have suggested that citronella oil may possess anti-cancer activity, particularly against breast cancer cells (Manosroi et al., 2006). This activity may be due to geraniol and citronellol, which have been found to exhibit anti-cancer anti-cancer effects in vitro.

OPTIMIZATION METHODS FOR SC-CO_2 EXTRACTION PARAMETERS

SC-CO_2 extraction is a versatile and environmentally friendly method to obtain valuable compounds from various natural sources, including essential oils like citronella. The success of SC-CO_2 extraction depends on the precise control of multiple operating parameters, such as temperature, pressure, particle size, and co-solvent ratios. Achieving the desired product quality, maximum yield, and process efficiency necessitates optimizing these parameters systematically. In this context, optimization methods are pivotal in fine-tuning SC-CO_2 extraction processes. These methods offer structured frameworks to explore the complex interplay of variables and identify optimal conditions. Three commonly employed optimization approaches in SC-CO_2 extraction are Response Surface Methodology (RSM) with Central Composite Design (CCD), Box-Behnken Design (BBD), and the Taguchi method.

Response Surface Methodology with Central Composite Design

Response Surface Methodology (RSM) with Central Composite Design (CCD) is a robust statistical technique for optimizing complex processes with multiple variables. When applied to the supercritical carbon dioxide (SC-CO_2) extraction of citronella oil, RSM-CCD offers a structured and systematic approach to investigating and optimizing critical parameters like temperature, pressure, and flow rate.

RSM allows researchers to systematically vary the selected parameters within predetermined ranges. In the case of SC-CO_2 extraction, these parameters can include temperature (T), pressure (P), and flow rate, among others. By systematically altering these factors, researchers can evaluate their individual and interactive effects on the extraction process (Arumugham et al., 2022). The Central Composite Design (CCD) is employed within RSM to construct a response surface. This surface accounts for quadratic effects, enabling a comprehensive analysis of how parameter interactions affect the response variable (Moges et al., 2022). In this case, the yield and quality of citronella oil. The CCD involves a series of experiments strategically placed within the parameter space, allowing for generating a mathematical model that approximates the relationship between the parameters and the response.

Once the response surface is constructed, researchers can analyze it to identify the optimal extraction conditions. These conditions represent the combination of parameters that maximize the desired outcome, such as citronella oil yield and product quality. By studying the response surface, researchers gain insights into the critical factors that significantly impact the extraction process. RSM-CCD has found practical application in SC-CO_2 extraction studies, as exemplified in Timung et al. (2016). This study employed the response surface to visualize the interplay between temperature and pressure, shedding light on their combined effects on citronella oil yield and quality. Researchers pinpointed the optimal conditions within the parameter space, enhancing process efficiency and product quality.

In summary, RSM-CCD is a valuable optimization methodology that offers a structured approach to fine-tune critical parameters in SC-CO_2 extraction processes. It leverages the construction of response surfaces to identify optimal conditions for citronella oil extraction. This statistical technique provides researchers with valuable insights into the complex interactions between parameters and their effects on yield and product quality, ultimately contributing to the advancement of SC-CO_2 extraction methods in various industries.

Box-Behnken Design

Box-Behnken Design (BBD) is a sophisticated experimental design method that efficiently optimizes SC-CO_2 extraction parameters (Rizkiyah et al., 2023). This methodology is particularly well-suited for situations where factors have three-level settings, making it a valuable tool for optimizing complex processes like SC-CO_2 extraction. BBD is designed to efficiently explore the design space by systematically varying critical parameters. In the case of SC-CO_2 extraction, researchers can select factors like temperature (T) and pressure (P) and explore multiple levels for each factor. BBD strategically places experiments at the low, middle, and high levels of these parameters, facilitating a thorough investigation of their effects on the extraction process (Putra, Rizkiyah, Idham, et al., 2023). Like RSM-CCD, BBD also involves the construction of a response surface. This mathematical model approximates the relationship between the selected parameters and the response variable, which includes factors like citronella oil yield and product quality. The response surface aids in visualizing how changes in parameter levels influence the response.

Researchers analyze the response surface generated through BBD to identify the optimal extraction conditions. These conditions represent the combination of parameter settings that maximize the desired outcome, such as achieving the highest yield of citronella oil with optimal product quality. BBD helps researchers navigate the parameter space efficiently to reach these optimal conditions. Furthermore, BBD systematically investigates both the individual and interactive effects of parameters on the extraction process. It provides a comprehensive understanding of how each parameter contributes to the response and how they interact. Understanding these effects is crucial for fine-tuning the extraction process.

BBD is a powerful experimental design method that efficiently explores parameter settings in SC-CO_2 extraction. It offers a structured approach to identifying optimal conditions for citronella oil extraction, taking into account the individual and interactive effects of parameters. BBD aids researchers in navigating the complexities of the extraction process, ultimately contributing to enhanced process efficiency and product quality in various industrial applications.

Taguchi Method

The Taguchi Method is a robust optimization technique that prioritizes the robustness and performance of a process. When applied to SC-CO_2 extraction, it becomes a valuable tool for optimizing a range of parameters, including those critical for citronella oil extraction, such as particle size and co-solvent ratios. The Taguchi Method is rooted in the concept of robust optimization, which aims to achieve consistent and high-quality results while accounting for variability in process parameters. It is particularly well-suited for processes like SC-CO_2 extraction, where maintaining product quality and reliability are paramount.

The Taguchi Method allows for the systematic optimization of multiple parameters simultaneously. SC-CO_2 extraction might include factors such as particle size, co-solvent ratios, pressure, and temperature. By optimizing these parameters together, researchers can ensure that the extraction process is not overly sensitive to variations, thus enhancing its robustness. The Taguchi Method employs orthogonal arrays to organize experiments efficiently (Ramezani et al., 2020). These arrays help in planning a relatively small number of experiments while providing a comprehensive understanding of the parameter interactions and their effects on the response variable, which in this case would be citronella oil yield and quality.

A key aspect of the Taguchi Method is the use of Signal-to-Noise (S/N) ratios to assess the performance of different parameter combinations (Ribeiro et al., 2017). These ratios help identify which parameter settings result in the most desirable outcomes. Researchers aim to maximize the S/N ratios associated with the desired response, indicating optimal process conditions. In summary, the Taguchi Method is a powerful and structured optimization technique that is particularly useful for ensuring the robustness and quality of SC-CO_2 extraction processes, including citronella oil extraction. It offers a multi-factorial approach to parameter optimization, utilizing orthogonal arrays and S/N ratios to systematically fine-tune process conditions. This methodology is instrumental in achieving consistent and desirable product characteristics while minimizing the impact of parameter variations.

COMPARISON OF SUPERCRITICAL CARBON DIOXIDE WITH TRADITIONAL EXTRACTION METHODS

Citronella oil has been traditionally extracted using steam distillation and hydrodistillation methods. However, the emergence of SC-CO_2 extraction has shown to be a promising alternative method for extracting essential oils, including citronella oil. The SC-CO_2 extraction method utilizes carbon dioxide in its supercritical state, where it behaves as both a gas and a liquid, to extract essential oils from plant materials.

Due to its many advantages, there has been a growing interest in using SC-CO_2 extraction as an alternative to traditional methods. Compared to traditional methods, SC-CO_2 extraction offers a more efficient and environmentally friendly process with higher yields and does not require toxic solvents. The SC-CO_2 extraction method is also highly tunable, optimizing extraction conditions to target specific compounds of interest in the extracted oil. Table 4 summarizes the advantages and disadvantages of conventional extraction and SC-CO_2 extraction.

Table 4
Advantages and disadvantages of conventional extraction and SC-CO_2 extraction

Extraction Method	Advantages	Disadvantages	Ref
Steam Distillation	• Low-cost and widely used • Preserves aroma and flavor	• Low yield • Time-consuming • Sensitive to heat • Degradation of heat-sensitive compounds	Hamzah et al., 2014; Weng et al., 2015
Solvent Extraction	• High yield • Can extract a wide range of compounds • Short extraction time	• Use of toxic solvents • Residual solvent in the final product • Expensive and time-consuming solvent recovery	Okpo and Otaraku, 2020
Hydrodistillation	• Efficient for extraction of high boiling point compounds • Preserves the aroma and flavor	• High energy consumption • Low yield • Long extraction time	Gavahian et al., 2018; Timung et al., 2016
SC-CO_2 Extraction	• Non-toxic and environmentally friendly • High selectivity and purity • Short extraction time • High yield • No residual solvents in the final product	• High equipment cost • Requires expertise to operate and optimize	Wu et al., 2019

Several methods are employed to extract citronella oil, each with its advantages and disadvantages. Steam distillation, a widely used method, is characterized by its cost-effectiveness and ability to preserve the aroma and flavor of the oil. However, it suffers from drawbacks such as relatively low yields, time-consuming processes, and sensitivity to heat, which can degrade heat-sensitive compounds within the oil. Solvent extraction offers high yields and can quickly extract a wide range of compounds. Nevertheless, it involves toxic solvents, potentially resulting in residual solvents in the final product, and can be expensive due to the need for solvent recovery. Hydrodistillation is efficient for compounds with high boiling points and maintains the oil's aromatic and flavorful qualities. However, it comes with the trade-offs of high energy consumption, lower yields, and lengthy extraction times. In contrast, SC-CO_2 extraction offers a non-toxic and environmentally friendly alternative. It excels in purity, yield, and speed, with no residual solvents in the final product. Nonetheless, the method requires a substantial equipment investment and specialized expertise for optimal operation. The choice of extraction method hinges on specific project requirements and considerations, ranging from yield and product quality to environmental concerns and available resources.

APPLICATION CITRONELLA OIL

Due to its unique chemical composition and properties, citronella oil has numerous applications in numerous industries. The following are some of the most common applications of citronella oil:

Due to its delightful, fresh, and citrus aroma, Citronella oil is extensively employed in the fragrance industry. Popular in fragrances, balms, candles, and air fresheners (Ganjewala, 2009).

- Citronella oil is effective against mosquitoes, flies, and other insects as a natural insect repellent. It is commonly used in candles, perfumes, mists, and mosquito coils (Hsu et al., 2013).
- Aromatherapy utilizes citronella oil to promote relaxation, reduce tension, and enhance mood. Typical applications include diffusers, massage oils, and bath additives (Barbas et al., 2017).
- Citronella oil is a flavoring agent in the food and beverage industry (Neequaye et al., 2017). It is utilized frequently in soft beverages, ice creams, chocolates, and baked products.
- The antifungal, antibacterial, and antiseptic properties of citronella oil make it useful in the pharmaceutical industry (Kaur et al., 2021). It is incorporated into topical ointments, balms, and cosmetics.
- In veterinary medicine, citronella oil is used as an insect repellent and a natural treatment for various animal diseases.

- Citronella oil's antiseptic and astringent properties are utilized in cosmetics (Happy et al., 2021). It is commonly found in skin care products such as facial cleansers and toners.
- Citronella oil produces various industrial products, including insecticides, cleansing agents, and lubricants (Isman et al., 2011).

CHALLENGES AND FUTURE PERSPECTIVE IN SC-CO_2 EXTRACTION FOR CITRONELLA OIL

SC-CO_2 Extraction as an alternative to conventional methods for extracting essential oils, including citronella oil, has shown tremendous promise. However, some obstacles must be overcome for the widespread utilization of this technology in the citronella oil industry. Optimization of the extraction parameters is one of the most significant obstacles. Although the effects of pressure, temperature, flow rate, particle size, and co-solvent ratio on the extraction yield of citronella oil have been investigated, further optimization of these parameters is required to maximize the extraction yield and minimize operating costs. Additionally, the use of various co-solvents and their effects on the extraction yield and quality of the extracted oil should be investigated further.

Scaling up the SC-CO_2 extraction method presents further difficulty. While the laboratory-scale extraction yielded promising results, the scale-up process necessitates substantial investment and engineering expertise to ensure the process's efficacy and safety. In addition, the cost of the apparatus required for SC-CO_2 extraction is still comparatively high compared to conventional extraction techniques, which may prevent small-scale producers from adopting this technology.

The stability of the extracted citronella oil is an additional crucial factor to consider. The SC-CO_2 extraction method can yield high-quality essential oils with a minimal impurity content. To ensure the purity and efficacy of these oils, however, it is necessary to investigate their stability during storage and transportation. In addition, the effects of the extraction procedure on the bioactive compounds in citronella oil should be evaluated for their possible health benefits.

The future of SC-CO_2 extraction in the citronella oil industry appears promising despite these obstacles. The technology provides several advantages over conventional extraction techniques, including a higher yield, a lower impurity level, and a shorter extraction time. In addition, SC-CO_2 extraction has a lower environmental impact than conventional extraction methods because it does not require organic solvents, which can be detrimental to the environment. In addition, developing new technologies and optimizing existing ones can significantly enhance the SC-CO_2 extraction process's efficacy and cost-effectiveness (Chemat et al., 2020; De Oliveira et al., 2019). Using co-solvents and optimizing their ratio, for instance, can improve the extraction yield and the quality of the extracted oil.

Integrating SC-CO_2 extraction with other technologies, such as microwave and ultrasound, can improve the process's efficacy and selectivity.

The future of SC-CO_2 extraction for citronella oil appears promising, even though several obstacles still need to be addressed. Its future applications in the culinary, cosmetic, and pharmaceutical industries are extensive. Continued research and development in this field can significantly enhance the process's efficacy and cost-effectiveness, making it a viable alternative to conventional extraction techniques.

CONCLUSION

This review paper focused on the SC-CO_2 extraction of citronella oil, discussing topics such as the production and interesting compounds of citronella, the theory of SC-CO_2 extraction, and the various parameters that affect the yield and quality of the extracted oil. According to the review's findings, SC-CO_2 extraction is a promising alternative to conventional citronella oil extraction methods. It offers several advantages, including reduced solvent usage, shorter extraction times, and higher oil yields with a higher concentration of interesting compounds. However, there are still obstacles to maximizing the yield and quality of the extracted oil by optimizing the process parameters. Prospects for SC-CO_2 extraction of citronella oil include the development of new technologies and equipment, such as the use of co-solvents and the integration of ultrasound or microwave-assisted techniques, as well as the investigation of new applications for the extracted oil in various industries, such as the pharmaceutical, cosmetic, and food industries. This review provides valuable insights into the SC-CO_2 extraction of citronella oil, highlighting its potential as a sustainable and efficient alternative to traditional extraction methods and identifying research gaps.

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