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Citronella Oil as Additives in Active Carrageenan -based Food Packaging Films: A Review

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Authors' contributions

This work was carried out in collaboration among all authors. All authors read and approved the final manuscript.

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Review Article

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ABSTRACT

The natural degradation process of plastics is slow and harmful to the environment, making biodegradable films a more advantageous option. These films not only reduce environmental pollution but also have the potential to preserve food products and extend their shelf life. This study explores the potential of utilizing iota carrageenan-based films in combination with essential oils for food packaging applications. Iota carrageenan, known for its high elasticity and thixothrophic properties, serves as an ideal base material for film formation. Essential oils, with antioxidant and antimicrobial properties, are incorporated into the packaging material to enhance shelf life and prevent food spoilage. Citronella oil demonstrates effective antimicrobial and antioxidant activities for food preservation. The presence of essential oils acts as plasticizers in the film, improving flexibility but requiring careful consideration of concentration to maintain network strength.

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1. INTRODUCTION

"Food packaging is defined as the wrapping of food to protect it from tampering or contamination from physical, chemical, and biological sources, with active packaging being the most commonly used packaging system to preserve food products" [1]. "In general, the natural degradation of plastics starts with photodegradation, which leads to thermo-oxidative degradation. Ultraviolet light from the sun provides the activation energy required to initiate the incorporation of oxygen atoms into the polymer. This causes the plastic to become brittle and break into smaller and smaller pieces, until the polymer chain reaches a low enough molecular weight to be metabolized by microorganisms. These microbes can convert the carbon in the polymer chains into carbon dioxide or combine them into biomolecules. However, this whole process is very slow, and plastics can take 50 years or more to degrade" [2]. "Due to the disadvantages of these plastics that are difficult to degrade naturally, a new solution is currently needed, namely packaging that is easy to degrade naturally, also known as biodegradable films. The utilisation of natural materials to produce biopolymer-based films and coatings has gained increasing attention, as petrochemical-based plastic packaging creates serious environmental problems due to the material's inability to biodegrade. The rise of consumer concerns about the quality and shelf-life of food products and awareness of environmental issues has triggered food scientists to develop biodegradable films" [3]. "Biodegradable films have received attention due to their advantages over plastics. Besides reducing environmental pollution, biodegradable films can be used as carriers of active compounds, preservatives, and antimicrobial agents. In addition, by acting as a barrier to the transfer of water vapor, gas, lipids, and flavor components, biodegradable films can prevent quality deterioration and extend the shelf life of food products" [4]. "The national production of seaweed in Indonesia has increased dramatically in recent decades. According to FAO Statistics (FishStat), cultivated carrageenophyte seaweed production in Indonesia approached 3.4 million tonnes in 2010 [5]. The industrial manufacture of carrageenan is no longer limited to the extraction of refined carrageenan, but also includes semi refinedcarrageenan such as semi-refined iota carrageenan (SRiC). The SRiCproduct is considerably cheaper than refined carrageenan as it does not require involved steps such as centrifugation, filtration and alcohol precipitation that must be recovered" [6]. Therefore, SRiC may have potential for replacing petroleum-based plastic films for food applications at a reasonable cost. However, few studies have explored the use of SRiC for making edible film [3].

2. CHEMICAL STRUCTURE AND PROPERTIES OF CARRAGEENAN

Carrageenan is an advanced preparation of alkali treated cottonii (ATC) or semi-refined carrageenan (SRC). The purity level of SRC is lower than refined carrageenan because the sulfate content is still high, so the gel forming power is low [7]. Carrageenan is an anionic and sulfated polysaccharide consisting of long chains alternating linear chains of (1 → 3)-β-D-galactose and (1 → 4)-3, 6-anhydro-α-D-galactose (3, 6-AG) or (1 → 4)-α-D-galactose with sulfate esters (15-40%) [8]. It is a new renewable natural material that these polymers are derived from the marine red algae family. They can be sorted into six basic forms depending on their source, solubility, and sulfate content: kappa, iota, lambda, mu, nu, and theta; among these, kappa, iota, and lambda. are commonly used as materials for hydrogel manufacturing due to their viscoelastic and gelling properties [5]. Currently, most carrageenans are extracted from *Kappaphycus alvarezii* and *Eucheuma denticulatum* [9] but some of them are still isolated from *Chondrus crispus* [10] native carrageenan, also known as refined carrageenan and filtered carrageenan, is used as a material for hydrogel production.

3. SEMI-REFINED IOTA CARREGEENAN (SRiC) PRODUCTION

The carrageenan production procedure starts with cooking raw red seaweed under alkaline conditions (e.g. sodium hydroxide) to increase the 3, 6-AG content, and then extracting the carrageenan. After extraction, the remaining seaweed is filtered to obtain a concentrated polysaccharide solution. This solution is then precipitated using isopropanol to obtain a fibrous coagulum, which is further separated, pressed, washed, dried, and ground. The alcohol precipitation method can be applied to all types of carrageenan, while the gel method involving potassium chloride is usually used for the extraction of kappa-carrageenan [11--13]. Carrageenan can also be extracted by environmentally friendly technologies, such as microwave-assisted extraction [14] and eutectic solvent method in [12].

4. COMPOSITION AND FUNCTIONAL PROPERTIES OF CITRONELLA OIL (CO)

Essential oils (EOs) have gained attention as natural additives in active food packaging due to their antioxidant and antimicrobial properties [15]. These volatile compounds, extracted from various plant organs, can be incorporated into biodegradable materials like edible films to create active packaging systems [16]. The inclusion of EOs in packaging materials can extend food shelf-life by preventing spoilage and improving preservation properties [17]. EOs can be added as free or encapsulated molecules, with encapsulation showing promising results [11]. However, incorporating EOs may affect packaging properties and consumer acceptability [18]. The use of essential os extracted from agro-industrial by-products in biodegradable packaging can lead to a more sustainable food industry [11]. While ssential oil offer numerous benefits, further research is needed to standardize their use in the food industry and address potential allergic reactions in some individuals [18].

"Citronella oil is a type of essential oil from the citronella plant (*Cymbopogon nardus*) that has been used in various industrial fields, such as the food industry, cosmetics, and also medicine. Just like other types of essential oils, lemongrass oil is volatile at room temperature without causing decomposition, light yellow to dark yellow in color, fragrant (according to the smell of the producing plant), and generally soluble in organic solvents and insoluble in water" [19]. "Regarding therapeutic applications, most citronella essential oils are restricted to their use as a mosquito repellent, antiparasitic, nematocidal, antifungal, and antibacterial agent" [20]. "The main compositions contain citronellol, citronellal and geraniol which had function of antifungal, antimicrobial, Anti-inflammatory, volatile, and well-being" [21]. "On previous research citronella oil has ability against 7 bacterial strains *(Escherichia coli, Salmonella typhi, Pseudomonas aeruginosa, Gram-positive Streptococcus pneumonia, Bacillus cerues, Staphylococcus aureus, Bacillus subtilis*) and 3 fungi *strains (Aspergillus flavus, Candida albicans, Geotrichum candidum)* on food applications. In addition, Sensory evaluation was performed non-significantly on adding essential oils" [22]. "Lemongrass oil is also widely used as a well-known insect repellent and is also registered for use in the United States for human application in eradicating insects and mosquitoes [23]. Lemongrass oil contains natural compounds that are stimulant, anti-inflammatory, antimicrobial, and antioxidant" [24].

"Extraction of citronella essential oil using steam distillation method. The steam distillation process where steam is flowed through a porous steam pipe then the steam will move to the top through the material stored above the filter, the advantage of this steam distillation extraction method is that the process is fast in producing essential oil. The volume of fragrant lemongrass essential oil produced from distillation for 10 kg of fresh fragrant lemongrass has an oil weight of 67.26 grams with a volume of 98 mL. With the known weight of citronella essential oil, the yield can be determined at 0.67%, the calculation of yield is used to determine how much percentage is obtained from the distillation results, so that later it can be known the need for the amount of raw material to be processed" [25].

Based on research conducted by Devi and Singh [26] essential oil from citronella plants (*Cymbopogon nardus*) is quite effective in inhibiting mold growth, so it can be used as a preservative for food commodities. The chemical components contained in essential oils vary, but these components can be classified into large groups that are dominant in determining the properties of essential oils. One of these components is terpenes, which are related to iso-prene or iso-pentane elements, are benzene derivatives, do not contain branch chains, and are straight chain compounds. The chemical components in lemongrass oil are quite complex, but there are main compounds that play an important role, namely citronellal, geraniol, and citronellol. The three main components determine the intensity of the fragrant odor to the selling price of citronella oil [27].

5. SRIC-BASED FILMS CONTAINING CITRONELLA OIL

SRiC-based films containing citronella oil have been researched and developed for various applications.A novel active packaging film containing citronella oil was prepared and characterized [28]. The film exhibited antimicrobial activity and was applied in grape

Table 1. Review of essential oil in food packaging

No	Main Findings	Source
1	Active packaging with essential oils can extend the shelf-life of food by interacting with the external environment. Essential oils can be incorporated into active packaging in the form of films and coatings. Active packaging with essential oils can help maintain temperature, moisture, and control microbial growth and food quality.	[17]
2	Essential oils contain bioactive compounds with antioxidative and antimicrobial properties. Essential oils are used as natural additives to extend the shelf-life of food products.Essential oils can be incorporated into packaging materials to provide "active or smart packaging" with improved properties.	[29]
3	Essential oils can be incorporated into biodegradable packaging materials like edible films to create active packaging systems with improved preservation properties. Encapsulating the essential oils in the packaging material is a particularly promising approach.The addition of essential oils to the packaging can provide bioactive properties that can extend the shelf-life of the packaged food products by preventing spoilage.	[11]
4	Essential oils have beneficial properties but are unstable and have limited applications. Nanoencapsulation of essential oils in biopolymeric nanocarriers can improve their stability and functionality. Essential Oil -loaded biopolymeric nanocarriers have shown promising antimicrobial and antioxidant effects in food products, but further studies are needed to explore their commercial exploitation.	[30]
5	There is an increasing demand for natural antioxidant active packaging due to its advantages over adding antioxidants directly to food. Various natural antioxidants like tocopherol, caffeic acid, catechin, etc. have been incorporated into food packaging in recent years. The paper reviews methods for determining the oxidation protection effect of antioxidant active films and quantifying natural antioxidants in food.	[31]
6	The paper provides an overview of the current knowledge on the antibacterial properties and mode of action of essential oils and their constituents. It identifies the main obstacle for using essential oil constituents as food preservatives, which is that they are not potent enough as single components and cause negative organoleptic effects. It suggests that exploiting synergies between several essential oil compounds could be a solution to this problem, and recommends that future research should focus on exploring the mode of action of individual constituents and investigating the mechanisms of synergy.	[32]
7	The paper provides an overview of the current knowledge on the antibacterial properties and mode of action of essential oils and their constituents. It identifies the main obstacle for using essential oil constituents as food preservatives, which is that they are not potent enough as single components and cause negative organoleptic effects. It suggests that exploiting synergies between several essential oil compounds could be a solution to this problem, and recommends that future research should focus on exploring the mode of action of individual constituents and investigating the mechanisms of synergy.	[33]

preservation [30]. A study characterized cassava starch-based edible films enriched with lemongrass oil. The film showed antimicrobial activity and potential applications in food preservation. Research on PLA films containing montmorillonite nanoclay-citronella essential oil hybrids was conducted. The hybrids showed potential for active film formulations, enhancing the antimicrobial properties of the films [34]. A study investigated the use of citronella oil in edible coatings to extend the shelf life and improve the quality of banana fruit. The results showed that the edible coating with citronella oil significantly reduced weight loss and microbial growth in bananas [35].

6. EFFECT OF CITRONELLA OIL ADDITION ON THE PROPERTIES OF SRIC-BASED FILMS

6.1 General Properties

The addition of citronella oil to SRiC-based films impacts various general properties. Citronella oil's incorporation tends to affect the mechanical properties, water content, and opacity of the films. Research indicates that films with added CO exhibit increased flexibility and reduced tensile strength compared to those without citronella oi. Additionally, the water content in these films is generally higher due to the hydrophobic nature of citronella oi, which influences the water absorption capacity and retention. Furthermore, the opacity of the films tends to increase with higher concentrations of citronella oi, resulting in less transparent films. This change in opacity can be attributed to the distribution and size of citronella oi droplets within the film matrix [36].

6.2 Morphology

The morphology of SRiC-based films is notably altered by the addition of citronella oil. Microstructural analysis reveals that citronella oil droplets undergo varying degrees of aggregation depending on the concentration of citronella oil used. At lower concentrations, citronella oil droplets are more uniformly dispersed within the polymer matrix, leading to a homogeneous film structure. However, as the concentration increases, citronella oil droplets tend to coalesce, forming larger aggregates. This aggregation can create microvoids and heterogeneities within the film, potentially impacting the film's barrier properties and mechanical integrity [37].

Additionally, scanning electron microscopy (SEM) images typically show that the surface of SRiC-based films with citronella oi is rougher and more irregular compared to control films. The roughness can be linked to the uneven distribution and partial migration of citronella oi to the film's surface during the drying process. This phenomenon affects not only the film’s mechanical properties but also its optical properties, such as gloss and clarity [38]. In conclusion, the addition of citronella oil to SRiC-based films significantly influences both their general properties and morphology. These changes are largely dependent on the concentration of citronella oil used, affecting the films' mechanical strength, water content, opacity, and structural uniformity.

6.3 Opacity of SRIC Film Incorporating Citronella Oil

Semi-refined iota carrageenan (SRiC) films show promise as biodegradable food packaging materials. These films can be enhanced with various additives to improve their properties. Incorporating plasticizers like sorbitol or glycerol increases film flexibility and thermal stability [30]. Adding essential oils, such as cinnamon oil, can impart antimicrobial and antioxidant properties [30]. ZnO nanoparticles improve UV-screening, transparency, and water barrier properties [30]. α-tocopherol incorporation enhances antioxidant activity and can prolong food shelf life [24]. However, additives may affect mechanical properties, with some decreasing tensile strength while increasing elongation at break [24]. Overall, SRiC films demonstrate comparable properties to refined carrageenan films, suggesting their potential as cost-effective, sustainable packaging materials [24]. These studies highlight the versatility of SRiC films and their potential for various food packaging applications. citronella oil affects light transmission, and light can scatter at the interface of citronella oil droplets [30] In general, antibacterial protein coatings effectively block UV light and visible light. These properties can delay the oxidative spoilage of food products [39].

6.4 Water Solubility

Previous research reported that Citronela Oil increased the WVP of cuticle-chitosan composite films [40]. It is known that water vapor usually occurs in the hydrophilic part of the film network. Thus, the WVP also depends on the hydrophilic/hydrophobic ratio of the film composition [41]. In contrast, interfacial interactions between citronella oil and the film substrate lead to reduced protein-water interactions, potentially allowing free passage of water molecules and thus increasing the WVP [42]. It has also been reported that the increase in WVP may be due to the plasticizing effect of citronella oil on the film due to the weakening of protein-protein interactions in the film [39]. Other factors, such as film thickness and relative humidity, may also affect WVP. For hydrophilic films, studies have shown a positive positive correlation between WVP and film thickness [43]. According to research conducted by Go and Bin Song [44] increasing the concentration of essential oil above 1% (b/b) in the film composite also results in higher solubility values. Citronella oil can loosen the film network, which causes water molecules to diffuse into the film network and causes the film to dissolve in water [41].

6.5 Moisture and Solubility of Films

In [25] the moisture content of the films varied between 20% - 27.5%. this was due to the different drying treatments. Drying was carried out under the same conditions, and the moisture content of the film decreased with increasing citronella oi concentration. Similar results were observed in oregano oil biocomposite films [41]. Due to the hydrophobic nature of citronella oil, citronella oil-containing films are more soluble than SPI films. It is possible that protein-water partially replaces protein-water interactions [45]. Water solubility of films is an important characteristic of food packaging films, and potential commercial films should usually be insoluble or water-resistant. The low film solubility in this study may be because protein-citronella oi interactions mainly occur through hydrogen hydrogen, which facilitates film development [46]. In some situations, edible films are made to dissolve in water before consumption. However, low solubility films have the advantage of improving the integrity and water resistance of packaged foods. Therefore, the solubility of the film should be adjusted according to the needs of the industry, as too high solubility cannot protect the product from water loss and moisture [4].

6.6 Antioxidant Properties

“Carrageenan iota film (SRiC) is known for its antioxidant properties, which can be enhanced by incorporating natural antioxidants such as citronella oil. Citronella oil, which is derived from the leaves of the *Cymbopogon nardus* plant, is rich in antioxidants and has been shown to exhibit significant antioxidant activity. SRiC film with citronella oil has been shown to exhibit high antioxidant activity in the DPPH radical scavenging assay, indicating its ability to effectively neutralize free radicals and protect against oxidative damage. The antioxidant activity of SRiC films with citronella oil has also been evaluated using the β-carotene bleaching assay, which measures the ability of the film to inhibit β-carotene oxidation. The results showed that the film with citronella oil exhibited significant antioxidant activity, indicating its ability to protect against lipid peroxidation” [47]. “Antioxidant compounds in citronella oil come from terpenoid compounds and secondary metabolites such as phenolic groups, where phenol groups have an important role in antioxidant activity” [48]. “The mechanism of action of phenolic compounds as antioxidants is through the ability of the phenol group to bind a free radical by donating a hydrogen atom through an electron transfer process, so that phenol turns into a phenoxyl radical” [4].

6.7 Antimicrobial Properties

The antibacterial activity of essential oils is determined by differences in chemical composition, one of which is terpenoid compounds. According to Evans and Cowan [49] citronella oil content consists of terpenoid metabolites, namely monoterpenes and sesquiterpenes. Terpenoids have antibacterial effects, and the mechanism of action of these compounds reacts with porins (transmembrane proteins) found in the outer membrane of the bacterial cell wall. This bond creates a strong polymeric bond that damages the porin. Damaged porins affect the entry and exit of cellular components contained in bacteria, reduce the permeability of the bacterial cell wall and bacterial cells experience nutrient deficiencies inhibit bacterial growth or die [50]. The chemical content contained in citronella plants *(Cymbopogon nardus L.)* that can inhibit bacterial growth are saponins and flavonoids. Saponins can cause microbial cells to lyse by disrupting the stability of their cell membranes Raningsih et al. [51]. Saponins are polar surfactants that will reduce the surface tension of the sterol membrane of the bacterial cell wall, causing disruption of membrane permeability which results in the entry of materials or substances needed to be disrupted eventually the cell swells and ruptures. Flavonoids work by denaturing proteins, disrupting the lipid layer and causing cell wall damage. This can occur because flavonoids are lipophilic so that they will bind to phospholipids phospholipids in fungal cell membranes and disrupt cell membrane permeability [25].

7. CONCLUSION

The incorporation of citronella oil into carrageenan-based food packaging films offers a promising solution for sustainable and effective food preservation. Citronella oil, known for its antimicrobial and antioxidant properties, enhances the functional properties of these biodegradable films, making them suitable for extending the shelf life of food products. The addition of citronella oil increases the film's flexibility and water content, while reducing its tensile strength and transparency. The essential oil's chemical components, such as citronellal, geraniol, and citronellol, contribute to its effectiveness as a plasticizer and antimicrobial agent. These compounds disrupt bacterial cell membranes, preventing microbial growth and spoilage. Despite the potential sensory impacts and the need for further research to standardize its use, the application of citronella oil in food packaging represents a significant advancement in creating environmentally friendly and efficient preservation methods. Although preliminary results show the effectiveness of the formulation, several challenges need to be addressed for industrial scale production. One of them is ensuring the stability and consistency of the product in large quantities, given that composite film production requires strict control of mixing and drying conditions. In addition, the sensory properties of citronella oil-such as its strong aroma-can affect product acceptance in food packaging applications. Therefore, further development could involve consumer acceptance testing or selection of alternative essential oils with sensory profiles more suitable for food, while maintaining the desired functional properties.

DISCLAIMER (ARTIFICIAL INTELLIGENCE)

Author(s) hereby declare that NO generative AI technologies such as Large Language Models (ChatGPT, COPILOT, etc.) and text-to-image generators have been used during the writing or editing of this manuscript.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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