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INCORPORATING CITRONELLA OIL INTO EDIBLE COATING TO EXTEND THE STABLE LIFE AND IMPROVE THE QUALITY OF BANANA FRUIT

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Abstract: Carboxymethyl cellulose (CMC) is a linear polysaccharide with long chains soluble in water and anionic. It exhibits high viscosity and possesses non-toxic and non-allergenic properties. The use of plant-based essential oils in edible films to replace vegetable oils, cereals, or seeds. One essential oil with good antibacterial activity is the oil derived from citronella leaves. The main components of this citronella oil compound consist of citronellal, citronellol, and geraniol, which can inhibit bacterial activity. Therefore, it is necessary to develop technology for handling fresh fruits to inhibit excessive ripening and decay. This can be achieved by creating an edible coating with essential oils. This study aims to investigate the impact of using an edible covering (carboxymethyl cellulose) incorporated with lemongrass oil on the storage of bananas. The experimental design employed in this study utilized a Completely Randomized Design (CRD) with one factor, namely citronella oil (0.3%, 0.6%, and 0.9%), as well as the application of edible coating with two elements, namely treatment variations (layer and non-coating) and storage duration (day 0 to day 15). The storage process is conducted until ripening occurs in banana fruits for 16 days, with daily observations. Throughout the storage procedure, various metrics were monitored in bananas, including weight loss, acidity level (pH), and total plate count (TPC). The research findings indicate that the highest weight loss of banana fruit is observed in bananas without edible covering, amounting to 13.10%. Conversely, bananas with edible coating and 0.5% lemongrass oil exhibit a lower weight loss of 7.35%. The application of edible coating significantly impacts the increase in pH and total microbial growth on banana fruits.

Keywords: *Carboxymethyl Cellulose, Edible Coating, Citronella Oil, Essential Oil*

INTRODUCTION

The banana is a widely recognized tropical fruit of significant popularity, particularly in local commercial trade. This substance is rich in essential nutrients and minerals, conferring numerous health benefits. Currently, there is a growing domestic demand for bananas. Nevertheless, it is not unusual for bananas to undergo damage or a decline in quality before reaching the end consumer. Hence, developing novel handling technologies is necessary to prevent rotting [1] effectively. Numerous studies have demonstrated that edible coatings can serve as carriers for a wide range of food additives, including but not limited to anti-browning agents, antimicrobials, coloring agents, flavoring agents, nutrients, and seasonings. One method for preserving the quality of bananas involves the application of edible coatings containing antimicrobial chemicals. These compounds can potentially impede micro-organism growth on the bananas' surface, reducing the likelihood of spoilage [2].

The limited durability of bananas poses an ongoing obstacle for the global banana industry, particularly in regions with inadequate access to refrigerated storage facilities. Various postharvest approaches have been examined to delay fruit ripening, such as low-temperature storage, control room storage, using ethylene antagonists such as 1-MCP, and applying fruit surface coatings [3]. Incorporating natural antibacterial agents into edible films and coatings has been suggested to enhance the quality and extend the shelf life of fruits. There is an increased emphasis among consumers on food safety and nutritional content, leading to a preference for purchasing fruits free from chemical and antibacterial agents. This preference arises from concerns about the potential adverse impacts on human health [3]. Furthermore, it is worth noting that natural antibacterial agents exhibit robust antibacterial properties, minimal toxicity, and reduced environmental impact [4].

Essential oils, sometimes called essential oils (EO), are derived from natural plants and spices. These oils are widely recognized for their notable antibacterial and antioxidant characteristics attributed to their volatile nature. Importantly, their volatile nature ensures that these oils do not leave any lasting effects [5]. Citronella (*Cymbopogon nardus*) is recognized as an essential oil possessing antimicrobial effects. Cymbopogon nardus, also known as fragrant citronella, is a botanical species utilized extensively to extract essential oils. The resulting citronella oil finds widespread application in the food industry and the creation of beverages, fragrances, soaps, personal care items, and medical formulations. Numerous investigations have documented the antibacterial characteristics of citronella (Cymbopogon nardus) essential oil [6]. Citronella oil contains various chemical ingredients, including saponins, polyphenols, and flavonoids. The presence of these active components suggests that citronella essential oil exhibits significant antibacterial properties [7].

A limited number of published publications explore the integration of essential oils into composite coatings or films. However, no research has been undertaken on combining carboxymethyl cellulose (CMC) with citronella oil. Carboxymethyl cellulose (CMC), alternatively referred to as sodium carboxymethyl cellulose (Na-CMC), is a derivative of cellulose that has found extensive application across many sectors within the food industry [2]. Nevertheless, it should be noted that CMC lacks antibacterial characteristics and exhibits a significant degree of water vapour permeability, as stated in reference [8]. Published literature exists about the augmentation of moisture barrier and antibacterial characteristics by integrating hydrophobic substances, such as Zataria multiflora Boiss essential oil [9]. Therefore, incorporating essential oils into carboxymethyl cellulose (CMC) is a promising and viable approach to enhance the moisture barrier properties of CMC while concurrently imparting antibacterial attributes. Comprising a composite layer with essential oils does not impede the fruity fragrance or result in adverse consequences [8].

In this research, the investigators selected citronella oil as a hydrophobic and antibacterial substance to augment the characteristics of carboxymethyl cellulose (CMC) for potential use in fruit applications. Hence, this research aimed to assess the efficacy of carboxymethyl cellulose (CMC) in conjunction with citronella oil to enhance the overall quality of bananas. The parameters that were monitored during the storage of bananas were weight reduction, pH levels, and the total plate count (TPC). Banana storage is conducted for 15 days until the ripening process occurs, with daily observations being made.

RESEARCH METHODS

Materials and Equipment Used

The equipment utilized in this investigation included aluminum foil, a measuring flask, an Erlenmeyer flask, a glass beaker, a volume pipette, a separatory funnel, a thermometer, a water bath, an analytical balance, a timer, and a set of distillation apparatus. The primary constituents employed in this study consist of bananas (Musa acuminata *Cavendish*) sourced from the banana market in Langsa Aceh, along with carboxyl methyl cellulose (CMC), glycerol, and Tween 80. In the present context, citronella oil is derived from the leaves of citronella plants cultivated by farmers in the Gayo Lues region of Aceh.

Extraction of Citronella Oil

The hydrodistillation method is employed to extract citronella oil, utilizing a leaf size of 1 cm [10]. During the procedure of removing citronella leaves and water, the components as mentioned above are combined within a round bottom flask, maintaining a feed-to-solvent ratio of 1:2. Subsequently, the flask is positioned onto the heated plate and linked to the Clevenger apparatus, where it is then run for four hours. The fragrant citronella oil is collected within a separatory funnel and subjected to a drying process utilizing anhydrous sodium sulfate. A small quantity of n-hexane is included in the product for the extraction. Next, the citronella oil is carefully measured and placed into vials to determine the overall yield of citronella oil. Lemongrass oil is prepared for utilization as a supplementary component in edible coatings. The oil recovered from the distillation process was further analyzed to identify its active features using Gas Chromatography/Mass Spectrometry (GC-MS) [11].

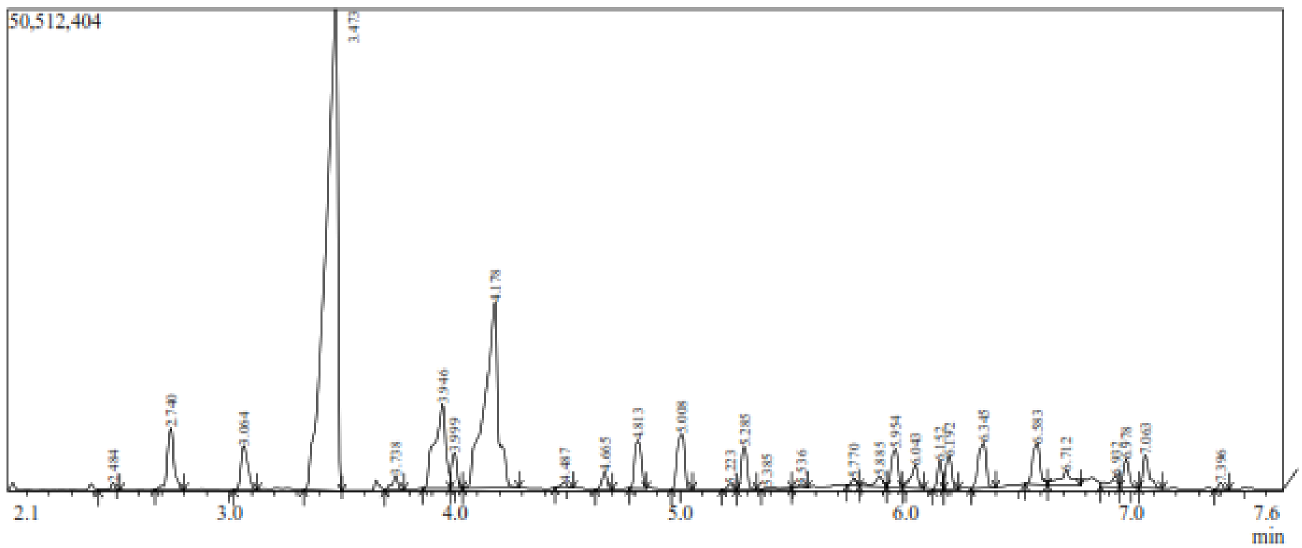

Figure 1. Spectrum analysis test results of citronella oil using GCMS

The achieved yield of citronella oil was 1,174. The components of the essential oil extract were identified using the GC-MS test, and the findings are depicted in Figure 1. The chromatogram data of the essential oil in citronella was analyzed using gas chromatography-mass spectrometry (GC-MS) to determine its three primary components' peak and retention time. The citronellal content was found to be 42.94%, while the geraniol content was determined to be 18.23%. Additionally, the citronellol content was measured to be 6.80%.

Preparation of Composite Coatings

The procedure involves the production of a 500 ml quantity of an edible coating formula using the manual stirring technique, employing a stirrer to facilitate the homogenization process. Initially, the distilled water is heated using a hot plate until it reaches a temperature of 70ºC. Subsequently, a solution of CMC with a concentration of 1% was gradually dissolved in distilled water under continuous stirring for 45 minutes until achieving a state of homogeneity. Subsequently, a 3% concentration of glycerol was introduced to enhance the flexibility of the layer. Simultaneously, a 0.5% potassium sorbate concentration was added while continuously stirring. Finally, a 0.5% concentration of stearic fatty acid was incorporated into the mixture, ensuring homogeneity by continued swirling. Once completely dissolved, the formula for the edible coating was allowed to cool down to the room's ambient temperature, which typically ranges between 25ºC and 30ºC. Subsequently, citronella oil at concentrations of 0.3%, 0.6%, and 0.9% was incorporated into the edible coating formulation as an antibacterial agent. The mixture was continuously stirred for 30 minutes at ambient temperature [12, 13].

Application of Edible Coating with Citronella Oil to Bananas

The bananas undergo a process in which they are immersed in a solution containing 0.5% ascorbic acid for 60 seconds to inhibit browning. Subsequently, they are removed from the solution and subjected to air-drying facilitated by a fan. The control group consisted of banana fruits without any form of coating treatment. Bananas coated with carboxymethyl cellulose (CMC) and infused with varying amounts (0%, 0.5%, 1.0%, and 1.5%) of citronella oil were appropriately marked for identification purposes. During the coating process, the bananas are immersed in the edible coating solution for 60 seconds, after which they are subsequently drained to facilitate drying. The storage of bananas was conducted under ambient conditions, with a subset of bananas serving as control samples that were not treated with an edible covering. The storage process for bananas, until they reach the ripening stage, spans 15 days, during which daily observations are conducted. During the storage of bananas, various parameters were monitored, including weight loss, pH, and total plate count (TPC) [14].

Data Analysis

The present study employed a completely randomized design (CRD) with a single component, specifically the concentration of citronella oil. The acquired data was analyzed using the Statistical Package for the Social Sciences (SPSS) software, employing a one-way analysis of variance (ANOVA) method. If the measurement outcomes indicate the presence of substantial disparities across the treatments, it is advisable to proceed with Duncan's Multiple Range Test (DMRT) at a significance level of 0.05. In order to assess the impact of varying concentrations of basil essential oil on the properties of tilapia fillets, the collected data was subjected to statistical analysis employing a paired t-test at a significance level of 0.05.

RESULTS AND DISCUSSION

Extracting Citronella Oil

The distillation process with a hydrodistillation apparatus is employed to extract fragrant citronella oil. Before the distillation process, preliminary treatment, including drying and size reduction procedures, is conducted. The optimization of conditions and treatment methods for these materials has been found to enhance the percentage yield of essential oils, as supported by existing literature. According to previous studies, the withering process decreases the material glands' water content, facilitating extraction. Additionally, the enumeration process aims to increase the surface area available for evaporation and contact with water, thereby reducing the extraction of essential oils [11].

This investigation yielded a percentage of 1.174% utilizing the hydrodistillation process, which required a total extraction time of 7 hours. The analysis of active compounds was conducted using the GCMS-QP2010 Ultra instrument, as depicted in Figure 1. The citronellal concentration was 42.94%, the geraniol content was measured at 18.23%, and the citronellol content was determined to be 6.80%. Additionally, the investigation detected 29 chemicals identified in the citronella oil sample.

Citronella oil comprises many chemicals, including citronellal, citronellol, geraniol, geranyl acetate, citronellal acetate, limonene, and linalool. These chemicals possess several advantages, including antibacterial, antifungal, and anti-inflammatory properties [4]. Edible coatings using essential oils can create a film layer that exhibits impermeability to both water and air. The application of a film layer has the potential to impede the ingress of micro-organism into the fruit or vegetable. Furthermore, this film layer can restrict the proliferation of micro-organism, prolong the duration of storage, enhance the overall characteristics, and

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augment the nutritional composition of fruits or vegetables [15].

Weight Loss

The application of an edible coating on fruit has the potential to mitigate weight loss by effectively reducing the rate of water loss in the fruit. Applying an edible layer is an effective barrier against the permeation of water and oxygen while also exhibiting the capability to regulate the respiration rate. The phenomena of weight loss can be attributed to the depletion of water and other volatile constituents through the processes of respiration, involving the evaporation of water, gas, and energy, as well as transpiration, which entails the release of water in the form of water vapour, during the storage period [1]. The weight loss value that exhibited the least magnitude was observed in the case of bananas subjected to an edible coating containing an additional 0.5% citronella oil (F2 = 7.35%).

This observation demonstrates that the incorporation of citronella oil into the edible coating enhances the hydrophobic properties of the carboxymethyl cellulose (CMC) coating [16]. The addition of citronella oil has been observed to impact the hydrophilic characteristics of carboxymethyl cellulose (CMC). The presence of low concentrations of citronella oil can result in a cross-linking effect when interacting with CMC. This cross-linking effect reduces the accessibility of hydroxyl groups for interaction with water molecules, ultimately forming a composite coating that is more resistant to water [15].

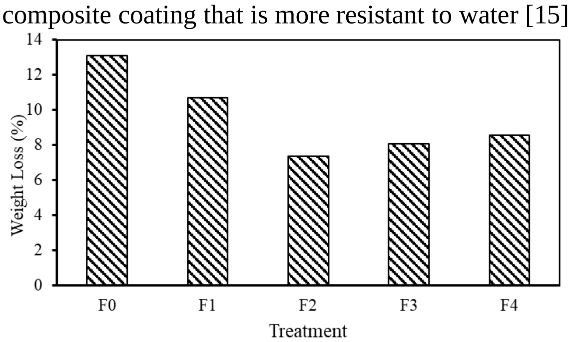

Figure 2. The present study investigates the potential association between weight loss and different amounts of citronella oil incorporated into bananas.

In contrast, it was observed that an increase in starch concentration resulted in a decline in the quality of both citronella oil-infused and non-infused edible coatings. Figure 2 illustrates that the weight loss percentage of bananas lacking an edible coating (F0) was 13.10%. In contrast, the weight loss percentage of bananas with an edible covering but without the inclusion of citronella oil (F1) was 10.69%. This finding demonstrates that applying an edible coating can effectively impede the process of water loss in bananas. Figure 2 illustrates the edible layer incorporating citronella oil at varying concentrations (0.5% F3, 1.0% F4, and 1.5% F5), resulting in weight percentages of 7.35%, 8.10%, and 8.56%.

The decline in the efficacy of weight reduction associated with higher concentrations of citronella oil can be attributed to the thermogenic qualities inherent in citronella oil. The findings of this study indicate that the optimal concentration of citronella oil for bananas was determined to be 0.5%. According to Tavassoli Kafrani et al. (2016), incorporating citronella oil into an edible coating enhances the hydrophobicity of the coating layer. It improves permeability, effectively resisting water vapor transmission [17].

Additionally, this combination exhibits favorable barrier capabilities against oxygen and carbon dioxide. The addition of citronella oil to edible coatings can lead to a reduction in their hydrophilic characteristics. This is due to the cross-linking effect when the edible layer interacts with low concentrations of citronella oil. As a result, the availability of hydroxyl groups for interaction with water molecules is diminished, resulting in a more durable composite coating [15].

Acidity Degree (pH)

The pH value, which indicates the degree of acidity of bananas during the storage period, is depicted in Figure 3. This study conducted a pH test during the storage period to assess the storability of bananas within the range of consumer acceptance limitations. The bananas were subjected to two treatments: coating with citronella oil and non-coating. This phenomenon can be attributed to the ongoing respiration process and enzyme activity that bananas experience during storage, gradually diminishing until reaching a near-zero rate. The observed decrease serves as an exemplification of the denaturation process of the enzyme [2].

Figure 3 illustrates the pH changes of banana fruit throughout the storage period. Figure 3 demonstrates that bananas underwent a pH increase in the citronella oil coating and non-coating treatments. However, it is noteworthy that the non-coating treatment resulted in a more rapid pH increase in bananas. A coating layer on the fruit effectively impedes respiration, leading to an elevation in CO_2 production. Consequently, the organic acids within the fruit can resist rapid decomposition when exposed to aerobic circumstances.

Furthermore, it has been shown that essential oils can impede the functioning of respiratory enzymes, thus decelerating the fruit's respiration rate. Respiration is a fundamental metabolic process that yields energy, albeit with the potential to induce detrimental effects on fruit quality. Consequently, suppressing respiratory enzyme activity can aid in preserving fruit's freshness [1].

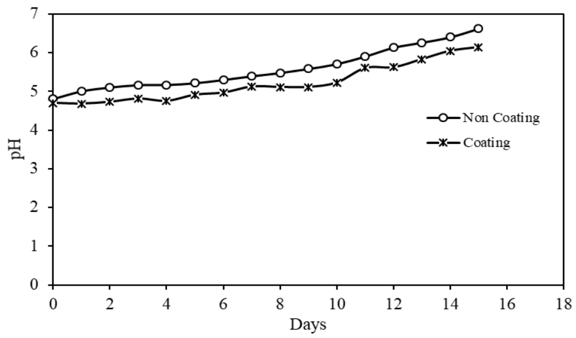

Figure 3. Comparative analysis of the acidity degree (pH) on bananas coated with citronella oil versus non-coated bananas.

Edible coatings consist of polysaccharides that create a thin film exhibiting semi-permeable characteristics. This film helps maintain the balance of aerobic and anaerobic respiration gases, thus preventing senescence [18]. The results of the non-coating treatment demonstrated a significant and swift rise in the pH levels of bananas. This phenomenon can be attributed to the thin surface skin of bananas, which allows for a high permeability of the fruit's stomata (pores).

Consequently, there is an increased rate of transpiration due to the easy passage of CO_2 from the external atmosphere to the inner tissues. As a result, the acids present in the fruit are affected. Under aerobic circumstances, organic matter experiences a fast breakdown. Alterations in pH levels directly result from fluctuations in the concentration of organic acids present within bananas. The amounts of organic acids increase during the fruit's ripening process, thus leading to a rise in the fruit's pH. During the growth period, fruit ripening is accompanied by a subsequent elevation in the amounts of simple sugars, resulting in a heightened sweetness in the fruit's taste. The observed phenomenon can be attributed to a reduction in the concentration of organic acids [19].

The presence of organic acids influences the pH level of the fruit. A corresponding elevation in the pH level characterizes the reduction in acidity. A low pH value indicates that the fruit's organic acids are preserved in a satisfactory state. The observed rise in pH can be attributed to reduced acid production and a drop in organic acid concentration over the storage period. Variations in the acidity of bananas are contingent upon the degree of maturity and the temperature at which they are stored [20]. The process entails the conversion of malic acid present in mature fruit into citric acid. The initial depletion of malic acid followed by citric acid indicates a potential pathway for citric acid catabolism involving malic acid. Throughout the fruit ripening process, the complete starch composition undergoes hydrolysis, forming sucrose, glucose, and fructose. During ripening, there is typically an increase in the concentration of simple sugars, a decrease in organic acids and phenolic compounds that contribute to the tart and sour taste, and an increase in volatile chemicals that provide a characteristic flavor to the fruit [15].

Total Plate Count (TPC)

One potential benefit of incorporating antimicrobial agents into edible coatings is the possible shelf life extension. Furthermore, incorporating antimicrobial active components in the film layer enhances its barrier qualities, inhibiting putrefactive bacteria and a subsequent reduction in potential health concerns. The utilization of antimicrobial agents derived from natural sources is considered to be a safer alternative compared to manufactured antimicrobial agents. According to a study conducted by Rojas Grau et al. (2009), the effectiveness of antimicrobial agents applied directly to the surface of fruits may be compromised due to certain components within the fruit [21]. Figure 4 illustrates bananas' Total Plate Count (TPC) value over the storage period.

According to the data presented in Figure 4, microbial populations have a noticeable rise during the storage period. The total plate count (TPC) values of the banana samples subjected to the coating treatment varied between 2.83 and 5.31 log cfu/g. In the present study, the total plate count (TPC) values of banana samples subjected to treatment with non-coated edible film composites containing pectin and essential oils exhibited a range of 2.85 to 6.18 log cfu/g. The statistical analysis, employing Duncan's Multiple Range Test with a significance level of $\alpha = 0.05$, revealed significant differences in the total phenolic content (TPC) values among treatments and storage times ($\alpha > 0.05$). The experimental results indicated that a coating treatment supplemented with citronella oil exhibited superior efficacy in preventing microbiological deterioration compared to the non-coating treatment when applied to bananas. This study selected the incorporation of antibacterial agents in citronella oil. The inhibitory effects of secondary metabolites found in plants, specifically those belonging to the flavonoid, phenol, terpenoid, and essential oil groups, have been observed in various harmful pathogens such as Escherichia coli bacteria, Bacillus subtilis, Staphylococcus aureus, Neurospora sp. fungus, Rhizopus sp., and Penicillium sp [22][6]. The primary constituents of citronella oil consist of citronellal, citronellol, and geraniol. The abovementioned chemicals are classified as aldehydes, alcohols, and esters, each possessing a discernible and potent fragrance. In addition to the three primary constituents, citronella oil comprises several additional chemicals, including geranyl acetate, citronellal acetate, limonene, and linalool. Citronella has a range of chemical components that offer diverse advantages, including antibacterial, antifungal, and anti-inflammatory properties [4]. This study additionally demonstrated that applying various treatments on bananas, which were coated with edible coatings containing essential oil, reduced the overall

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microbial count. Specifically, the bananas with edible coating exhibited a microbial count, but those without the layer had a count after a storage period of 15 days.

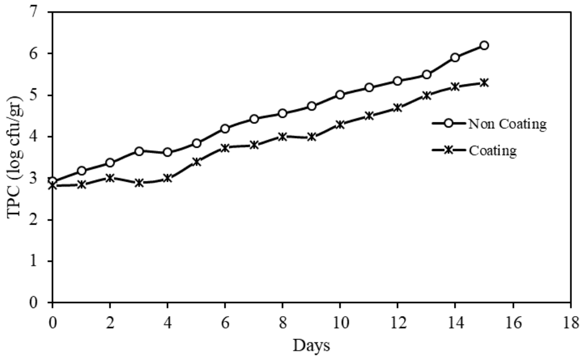

Figure 4. A comparative analysis of the total plate count (TPC) on bananas coated with citronella oil versus non-coated bananas.

Citronella oil exhibits antimicrobial properties that effectively inhibit the proliferation of micro-organism on edible coatings. The reason behind this is that citronella oil possesses antibacterial and antifungal properties. The antibacterial characteristics of citronella oil can be attributed to its composition of citronellal, citronellol, and geraniol molecules [6]. These chemicals possess the ability to impede bacterial growth by the infliction of damage upon the cell membrane of the bacteria. The antifungal characteristics of citronella oil can be attributed to specific chemical components, namely limonene, linalool, and geranyl acetate. These chemicals can impede the proliferation of fungus through the mechanism of fungal cell wall degradation. Using citronella oil within an edible coating can create a film layer that exhibits impermeability to both water and air. The application of a film layer has the potential to impede the ingress of micro-organism into the fruit or vegetable. Furthermore, this film layer can hinder the proliferation of pre-existing micro-organisms present on the surface of the fruit or vegetable [23].

CONCLUSION

Based on the research, it can be inferred that the hydrodistillation technique yielded a citronella oil extraction with a percentage yield of 1.17%. Furthermore, the analysis detected 29 active components in the extracted citronella oil. The bananas that exhibited the least weight loss had been treated with an edible coating containing 0.5% essential oil at a concentration of 7.35%. The pH of bananas showed an elevation in both the treatment involving a citronella oil coating and the treatment without any coating. The TPC analysis conducted on bananas also revealed a reduced bacterial count. The observed total plate count (TPC) values for banana samples subjected to essential oil treatment range of 2.83 and 5.31 log cfu/g.

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