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Original Research

Liquid Hand Soap Formulation of Java Citronella (*Cymbopogon winterianus*) Oil Using Surfactant Combination as Foaming Agent

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ABSTRACT

Background: *Unhygienic hands can cause microorganism infection. Citronella oil contains citronellal, citronellol, and geraniol, which have antibacterial activity. Liquid hand soap removes the dirt and microorganisms; however, it commonly uses sodium lauryl sulphate (SLS) as a foaming agent that may irritate the skin. Lauryl glucoside is a non-ionic, non-irritating surfactant. This study aimed to formulate and characterise liquid hand soaps from citronella oil using a combination of SLS and lauryl glucoside as surfactants.*

Methods: *The research was conducted using an experimental laboratory method. Three liquid hand soap formulas were developed from 15% citronella oil with different weight ratios of SLS and lauryl glucoside: F1 (1:2), F2 (1:1), and F3 (2:1). Then, all liquid hand soaps were evaluated for organoleptic properties, homogeneity, viscosity, pH, and foam height. Then the foam stability test was conducted after the formed foam was left for 5 minutes.*

Results: *The results showed that all liquid hand soaps were homogeneously distributed, clear viscous liquids having a distinctive odour of citronella oil. The liquid hand soap preparations had a viscosity range of 549.7–565.8 cps, a pH in the range of 7.39–7.89, a foam height of 74–85 mm, and a stability foam of 63.52–68.91%. All formulas complied with SNI 2588:2017 criteria for liquid hand soap.*

Conclusion: *This study presented that citronella oil could be formulated into liquid hand soap using a combination of SLS and lauryl glucoside. The optimal formula was F1 with a 1:2 ratio of SLS and lauryl glucoside. F1 had the most stable foam due to the higher concentration of lauryl glucoside, which enhances foam stability.*

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INTRODUCTION

Hands commonly become carriers for pathogenic bacteria to enter the human body. The COVID-19 pandemic increased our awareness of maintaining hand hygiene in order to lower the virus transmission rate. Moreover, WHO recommends people wash their hands thoroughly for a minimum of 20 seconds; this is especially important after being in public spaces, before eating, after coughing or sneezing, after using the restroom, and whenever hands are visibly dirty (Rundle et al., 2020).

Therefore, hand hygiene should be routinely conducted by washing hands with antibacterial soap. Hand soap is effective in removing dirt and dust from the surface of the skin and reducing the number of pathogenic microorganisms such as viruses, bacteria, and other parasites (Y.P.M. & Azizah, 2021). Liquid hand soap as a household health supply is more widely used because it is more practical and hygienic than the other preparation forms (Rinaldi et al., 2021).

Currently, there is an increase in the usage of herbal ingredients for soap formulation because it is considered safer than synthetic chemical compounds. Citronella's antibacterial activity makes it a potential active ingredient in liquid soap formulations. Citronella produces essential oil with main compounds consisting of citronellal, citronellol, and geraniol. Citronella oil is categorized into two types, the Ceylon and Java types.

The Ceylon type is obtained from *Cymbopogon nardus* Rendle, and the Java type is generated from *Cymbopogon winterianus* Jowitt. The Ceylon type produces essential oil with a lower level of citronellal and geraniol than the Java type (Sulaswatty & Adilina, 2019). Citronella oil derived from *Cymbopogon winterianus* contains major components such as citronellal (2.2–55.4%), geraniol (14.2–53.0%), citronellol (8.2–16.4%), isopulegol (0.3–12.6%), elemol (0.8–8.2%), and limonene (0.2–5.0%) (Verma et al., 2020).

A previous study found that Java citronella oil performed good antibacterial activity against E. coli, S. typhimurium, S. aureus, and Listeria monocytogenes (Verma et al., 2020). Similar results were reported by Munda (2019) that Java citronella oil showed good inhibition diameter for *E. coli* (28±0.176 mm), *B. cereus* (25±0.305 mm), and *B. subtilis* (19±0.289 mm) (Munda et al., 2019). SLS is widely used in liquid hand soap formulation as the foaming agent. It has high cleansing performance at a low cost (Takagi et al., 2014). As an anionic surfactant, SLS is prone to cause skin irritation, so its concentration in topical cleanser preparations is limited to 1-2% (Bondi et al., 2015).

People regularly use liquid hand soap to remove dirt and microorganisms on their hands. Moreover, it should have mild characteristics and effective cleansing ability. The formula combining anionic surfactant and nonionic surfactant is developed to produce nonirritating liquid hand soap. Lauryl glucoside, a nonionic surfactant, is known for its excellent foaming ability and low irritation profile (Takagi et al., 2014).

It has an emollient effect on the skin, so it will reduce irritation of the skin (Rohmani et al., 2022). Therefore, this study aimed to formulate Java citronella oil into liquid hand soap using a combination of SLS and lauryl glucoside as foaming agents to produce a mild formula as well as a good foaming ability to rinse off the dirt and microorganisms.

MATERIALS AND METHOD

Citronella oil was obtained from Triefta Aroma Nusantara, Indonesia. Glycerin (Ecogreen, Indonesia), hydroxypropyl methylcellulose (HPMC) (Dow Chemical Pacific, Singapore), Tween 80 (Fagron Hellas, Greece), sodium lauryl sulphate, lauryl glucoside, methyl paraben, and propyl paraben were purchased from Dipa Prasada Husada, Indonesia.

Formulation of Liquid Hand Soap of Citronella Oil

Liquid hand soap was prepared using three different weight ratios of SLS and lauryl glucoside: F1 (1:2), F2 (1:1), and F3 (2:1). Citronella oil as an active antibacterial agent was used in a concentration of 15%. The composition of liquid hand soap is presented in Table 1.

Table 1. Formulation of Liquid Hand Soap

Material	Function	Formula (% w/w)		
		F1	F2	F3
Citronella oil	Active ingredient	15	15	15
Glycerin	Humectant	5	5	5
Sodium lauryl sulfate	Surfactant	1	1.5	2
Lauryl glucoside	Surfactant	2	1.5	1
HPMC	Thickening agent	2	2	2
Methyl paraben	Preservative	0.18	0.18	0.18
Propyl paraben	Preservative	0.02	0.02	0.02
Tween 80	Surfactant	15	15	15
Aquadest	Solvent		ad 100	

Firstly, HPMC was dispersed in hot distilled water (80-90°C). It was stirred until forming a homogeneous gel (mixture A). Lauryl glucoside was dissolved in hot distilled water at 70°C in a different beaker glass (mixture B). Citronella oil was dissolved with Tween 80 (mixture C). Meanwhile, methyl paraben and propyl paraben were dissolved in glycerin (mixture D).

Then, the dissolved SLS in distilled water was poured and mixed into mixture A until a homogeneous dispersion was formed. Subsequently, mixtures B, C, and D were added to mixture A. It was stirred to form a homogeneous dispersion. Then, the remaining distilled water was added to reach the volume of 100 ml.

Evaluation of Liquid Hand Soap of Citronella Oil

The obtained liquid hand soaps were analyzed for organoleptic properties, homogeneity, pH value, viscosity, foam height, and foam stability.

Organoleptic test: The organoleptic test was carried out by directly observing the odor, colour, and form of the liquid hand soap preparation using sensory organs. This test was conducted to determine the physical appearance visually. Based on SNI criteria for liquid hand soap, the preparations should have a liquid form with a distinctive odor and colour (Y.P.M. & Azizah, 2021).

Homogeneity test: The liquid hand soap preparations were dropped and spread evenly onto a glass object. The preparation is homogeneous when there are no coarse particles observed visually (Y.P.M. & Azizah, 2021).

The pH test: One gram sample of citronella oil liquid hand soap was dispersed into 10 ml of distilled water. Then the pH value was measured using a pH meter. The pH test aims to determine the safety of a preparation. An acid pH can irritate the skin, while a pH that is too high results in scaly skin. According to SNI 2588:2017, the pH value of liquid hand soap should be in the range of 4-10 meters (Y.P.M. & Azizah, 2021).

Viscosity test: The viscosity of the preparations was determined using a Brookfield viscometer. A total of 15 ml of liquid hand soap was put into a cylindrical container, and then the viscosity was measured using spindle number 2 at a speed of 60 rpm (Ardana et al., 2015). The viscosity requirement for liquid hand soap is 400–4,000 cPs.
The foam height and stability test: The liquid hand soap preparation of citronella oil was put into a tube filled with distilled water. The tube was closed and shaken for 20 seconds, and the height of the formed foam was measured. The foam height was measured again after 5 minutes (Y.P.M. & Azizah, 2021). Then the stability of the foam was determined using the following equation:

$$\text{The foam stability} = \frac{H}{H_0}\ x\ 100\%$$

Ho is the initial foam height measurement, and H is the foam height measurement after 5 minutes. The preparations should have a foam height in the range of 13–220 mm, and the foam stability should be 60–70%.

RESULTS

Three formulas of liquid hand soap have been evaluated in terms of organoleptic properties, homogeneity, viscosity, pH, foam height, and foam stability. The organoleptic test showed that all formulas were clear, viscous liquids having the distinctive odor of citronella oil. Table 2 illustrates the results of the organoleptic test.

Table 2. Organoleptics of liquid hand soap of citronella oil

Formula	Form	Color	Odor
F1	Viscous liquid	Clear	Distinctive odor of citronella oil
F2	Viscous liquid	Clear	Distinctive odor of citronella oil
F3	Viscous liquid	Clear	Distinctive odor of citronella oil

The preparations then were observed for homogeneity, viscosity and pH. The results is presented on Table 3.

Table 3. Homogeneity, viscosity and pH of liquid hand soap of citronella oil

Formula	Homogeneity	Viskosity (cPs)	pH
F1	Homogeneous	565.8±0.1	7.89±0.11
F2	Homogeneous	549.7±0.1	7.59±0.14
F3	Homogeneous	550.1±0.2	7.39±0.15

Table 4 shows the summary of the foam height and stability of liquid hand soap which discovered that the preparations had the foam height in the range of 74±5.13 to 85±12.89 mm and foam stability 63.52 - 68.91 %.

Table 4. The height and stability foam of liquid hand soap of citronella oil

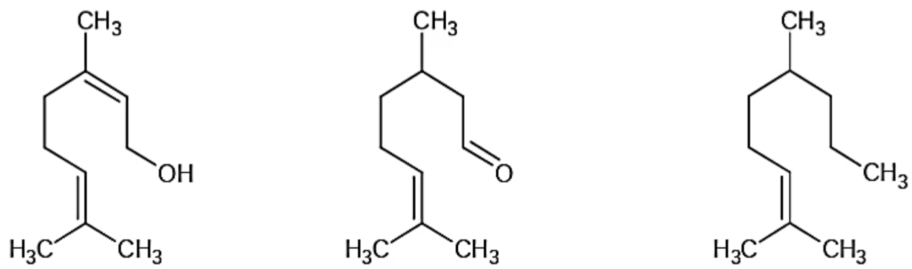
Formula	Ho (mm)	H (mm)	Foam Stability (%)
F1	74±5.13	51±5.50	68.91
F2	85±12.89	54±5.29	63.52
F3	76±6.11	49±5.03	64.47

DISCUSSION

This study used Java citronella oil as an active antibacterial ingredient for liquid hand soap. Java citronella oil, an essential oil, is an aromatic and volatile oily liquid derived from *Cymbopogon winterianus* Jowwit. Essential oils consist of various compounds that are chemically derived from terpenes and their oxygenated form (terpenoids) (Chouhan et al., 2017). A previous study by Shintawaty, (2020) found that the inhibitory activity of citronella oil against *Bacillus cereus*, *Staphylococcus aureus*, and *Escherichia coli* was categorized as strong, with the inhibition zone diameters being 18.80 mm, 13.07 mm, and 18.36 mm, respectively (Shintawati et al., 2020).

The major components of Java citronella oil were citronellal, geraniol, and citronellol. These compounds are terpene groups, which largely contribute to the antibacterial activity of citronella oil. Essential oils such as citronella oil have an antibacterial mechanism of action by increasing the permeability and disruption of bacterial membrane cells. The partition of terpene molecules into the lipid layer of the bacteria membrane changes the structural integrity of the bacteria cell membrane which leads to the leakage of various intracellular components such as protein, reducing sugar, ATP, DNA, and electrolyte.

Furthermore, the hydroxyl group of terpenes in essential oil has the capacity to bind and inhibit proteins such as ATPase so that they will inhibit the production of energy (ATP) (Álvarez-Martínez et al., 2021; Chouhan et al., 2017). Alcohol compounds of citronella oil, such as citronellol, geraniol, and nerol at low concentrations, cause dehydration of bacteria. Meanwhile, they will denature proteins, rupture cytoplasmic membranes, and damage bacterial cell walls at high concentrations (Shintawati et al., 2020).



(a) (b) (c)

Figure 1. Chemical structure of major component in Java citronella oil. (a) Geraniol, (b) citronellal, (c) citronellol (Lestari, 2022)

Based on Indonesian National Standard SNI 06-3953-1995, citronella oil should meet the following quality parameters: have a pale yellow to brownish yellow colour, a specific gravity of 0.880-0.922, a refractive index of 1.466-1.475, and a total citronellal more than 35%. Citronella oil purchased from Triefta Aroma Nusantara had a specification that complies with SNI 06-3953-1995 criteria with the content of citronellal of 35.67% (Sulaswatty & Adilina, 2019). Soap, a household health preparation, is used for washing by emulsifying dirt and impurities.

Liquid hand soap is commonly used Surface Active Agent (surfactant) that reduces the surface tension of skin and emulsifies unwanted lipophilic materials into finer particles then washing away skin impurities from the skin surface (Mijaljica et al., 2022; Navare, 2019). Surfactants, amphiphilic molecules, have hydrophilic and lipophilic structure that adsorps at the interfaces and stabilize foam film. Consumers generally prefer cleansing product that produce good foam to identify the effectiveness of washing or cleaning quality (Bureiko et al., 2015).

Soap preparations on the market generally contain SLS as a surfactant which functions to enhance foam production. SLS, an anionic surfactant, can irritate and lead the skin dryness because it removes the beneficial intercellular lipid component of the epidermal barrier and damages the keratinocytes. The previous study proved that SLS increased the disruption of structural protein in the skin. Anionic surfactant forms smaller micelles that easily penetrate the stratum corneum of the skin.

However, these micelles tend to break down into monomers rapidly due to electrostatic repulsion of polar moieties in the molecules. Then, the monomers bind and denature the cutaneous protein on the skin (Klimaszewska et al., 2022; Sari & Ferdinan, 2017). The binding of protein and surfactant also decreases the ability of protein to bind to water, leading to skin dryness (Draelos, 2018).

SLS is widely used in topical cleansing preparation due to high capacity of cleansing and foaming. Besides, it is cheaper that the other surfactants. To decrease the negative effect of SLS, a combination of two different surfactants, an anionic surfactant and nonionic surfactant has been developed. In this research, SLS was combined with lauryl glucoside, which is an non-ionic surfactant. Lauryl glucoside is a glycoside formed from mixing glucose and lauryl alcohol.

This surfactant is produced by combining corn sugar with palm oil or coconut oil. Lauryl glucoside has excellent foaming capacity and non-irritating properties. The foaming capacity correlates with the effectiveness of washing and rinsing off dirt. Although lauryl glucoside produces hard foam, the combination of this material with anionic surfactant will produce soft foam suitable for hand washing. A previous study found that the combination of anionic and nonionic surfactants produced bigger size micelles with less irritancy effect (Takagi et al., 2014). This study also used Tween 80, a nonionic surfactant, to increase the solubility level of citronella oil.

The liquid hand soap was prepared from 15% citronella oil using three different weight ratios of SLS and lauryl glucoside: F1 (1:2), F2 (1:1), and F3 (2:1). Then, the obtained preparations was evaluated in term of organoleptics, homogeneity, pH, viscosity, the foam height and foam stability. The organoleptics test presented on Table 2 show that all formulations were viscous liquid possesing clear color and distinctive odor of citronella oil. The distinctive odor of this preparations was attributed by the major component of citronella oil such as citronellal, geraniol and citronella.

The citrus like odor is attributed by citronellal component. Citronellol is responsible for giving the herbaceous rose with fuity citrus scent. Furthermore, geraniol contributes for the sweet floral rose aroma (Eden et al., 2018). The results obtained in this research in accordance with the criteria of SNI 2588:2017 for liquid hand soap. Organoleptic testing is carried out to determine the sensory quality of the preparation in terms of shape, color and odor by observing visually. Organoleptic examination needs to be carried out because it is related to comfort in use.

All developed liquid hand soap were homogeneous as represented on Table 3. Good homogeneity of a preparation indicates that the active substance is evenly distributed in the soap base. This result is comply with the homogeneity requirements, that the liquid soap preparation should have a homogeneous composition and have no clumped and visible coarse particles.

The pH value for the prepared liquid hand soap were 7.89±0.11, 7.59±0.14, and 7.39±0.15 for F1, F2 and F3 respectively. According to SNI 2588:2017, liquid hand soap should have pH value in the range of 4 – 10. All prepared liquid hand soap have pH that tends to be neutral. The increase of the SLS concentration decrease the pH value. SLS has a pH value between 6-8, so soap formulations with a greater SLS concentration will have a smaller pH (Chasani et al., 2022).

The nature of the skin has a slighty acidic pH for about 4 – 6. The preparations which are in the range skin pH result lower disruption of skin barrier and native microflora. A natural slighty acidic pH of the skin keep attachment of the microflora resident such as *Staphylococcus epidermidis* which is beneficial for skin healthyness. The alkaline pH will destabilize lipid bilayer of the skin due to swelling of skin protein and ionization of the fatty acid molecules. Moreover, an alkaline pH permit pathogenic bacteria such as *Staphylococcus aureus* to attach the skin (Mijaljica et al., 2022).

The viscosity test aims to determine the consistency of the preparation, which will later influence the application of the preparation. The liquid hand soap should be easy to pour or flow but not immediately spill from the hand. Table 3 shows that all of the liquid hand soap had viscosity in the range of 549.7±0.1 to 565.8±0.1 cps. These results indicated that all formulas met the required viscosity criteria 400 – 4,000 cPs. HPMC in liquid hand soap was used for thickening the preparations.

HPMC, a semi-synthetic methylcellulose derivate, is a hydrophilic polimer forming clear, odorless and stable hydrogel so that it affect the physical properties of liquid soap preparations, mainly the viscosity of the topical preparations. HPMC gel was formed by three dimensional polymeric network having capacity to hold high water content (Alyahya et al., 2023; Pan et al., 2023). Several advantages of HPMC as follow: has a cooling effect, nonirritating, nontoxic and generally regarded as safe (GRAS) properties (Muţ et al., 2018). Good hydrogels are very suitable for use as a base for topical preparations (Laksana et al., 2017). HPMC in liquid soap formulas also plays a role to stabilize the preparations by reducing the frequency of particles collision (Mahayuni et al., 2023).

One important parameter needs to be considered in determining the quality of soap is the amount of foam. A good preparation should meet the foam height requirements of 13 – 220 mm and should be able to maintain 60-70% of the initial foam height for 5 minutes. Table 4 shows that all formulations met the requirements with the foam stability in the range of 63.52 to 68.91 % which means the foam remains stable. The good foam stability of the preparation is caused by the surfactant content, namely sodium lauryl sulfate and lauryl glucoside. The higher surfactant concentration produced the higher amount of foam (Rohmani et al., 2022).

Foam is generated from dispersion of gas bubbles in surfactant solution. The adsorbed molecules of surfactant at the gas – liquid interface decrease the interfacial tension which then promote the liquid to foam. The drainage of liquid phase and the puncture of the bubble lamellae degrade foam so that they affect the foam stability (Nunes et al., 2020). The foam of F1 was the most stable due to the higher concentration of lauryl glucoside. Foam relates to the ability to remove dirt and instability (Takagi et al., 2014).

This study proved that the combination of SLS and lauryl glucoside surfactant had the potential to produce good characteristics of hand soap formulation. Lauryl glucoside, as a sugar-based, non-ionic surfactant, is a promising ingredient intended for sensitive skin preparation due to its mild properties. This surfactant also complies with the consumer demands nowadays that prefer to use naturally derived products (Nunes et al., 2020). The products using this eco-friendly natural surfactant can be considered to replace SLS-based commercially available products.

CONCLUSION

The liquid hand soap of citronella essential oil was successfully formulated using a combination of sodium lauryl sulfate and lauryl glucoside as a foaming agent. All formulas had good characteristics and met the criteria set by SNI 2588:2017 in terms of organoleptics, homogeneity, viscosity, pH, foam height, and foam stability. F1 was the optimal formula due to producing higher foam stability.

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