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Formulation of Spray Gel Hand Sanitizer from *Citronella Oil* Using Gelling Agent Carbopol 940 and Humectant Propylene Glycol

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ABSTRACT

Citronella (*Cymbopogon winterianus*) oil contains various phytochemical compounds which have antibacterial activity such as citronellal, citronellol, geraniol, and citral. These compounds can inhibit the growth of *Escherichia coli* and *Staphylococcus aureus,* two bacterias often found on the palm of hands. This study aimed to formulate citronella oil into a spray gel hand sanitizer. It used an experimental laboratory method. Spray gel hand sanitizers were developed from citronella oil using different concentration of gelling agent Carbopol 940 (0.2%; 0.3%) and humectant propylene glycol (10%; 15%). All formulas were evaluated in term of organoleptic, homogeneity, pH, viscosity, spraying patterns and stickiness. All of the obtained spray gel hand sanitizer presented a white viscous liquid having a distintive odor of citronella. Four formulas had pH in the range of 6.71 – 6.93 and viscosity in the range of 109.5±0.8 – 113.5±0.5 cps. The results showed that formulas 2 and 4 did not meet the requirements for spraying patterns and stickiness because the preparations clumped when sprayed from the spray applicator. Based on this study, it can be concluded that formula 1 had the best characteristics in term of organoleptic, homogeneity, pH, viscosity, spraying pattern and stickiness. The concentration of carbopol 940 affected the physical properties of spray gel hand sanitizer.

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ABSTRAK

Minyak sereh wangi (*Cymbopogon winterianus*) mengandung berbagai macam senyawa fitokimia yang memiliki aktivitas antibakteri diantaranya adalah citronellal, citronellol, geraniol, and citral. Senyawa tersebut diketahui dapat menghambat pertumbuhan bakteri *Escherichia coli* dan *Staphylococcus aureus* yang banyak ditemukan pada telapak tangan. Penelitian ini bertujuan untuk memformulasi minyak sereh wangi menjadi sediaan spray gel hand sanitizer. Metode yang digunakan dalam penelitian adalah eksperimental laboratorium. Spray gel hand sanitizers dibuat dengan bahan aktif minyak sereh wangi menggunakan konsentrasi carbopol 940 yang berbeda (0,2%; 0,3%) sebagai gelling agent dan propilen glikol sebagai humektan (10%; 15%). Sediaan kemudian dievaluasi karakteristiknya meliputi organoleptik, pH, viskositas, pola penyemprotan dan daya lekat. Hasil penelitian menunjukkan semua sediaan memiliki penampilan berupa cairan kental berwarna putih dengan bau khas minyak sereh wangi. Keempat formula memiliki pH pada rentang 6,71 – 6,93 dan viskositas pada rentang 109,5±0,8 - 113,5±0,5 cps. Formula 2 dan 4 tidak memenuhi persyaratan pola penyemprotan dan daya sebar karena sediaan menggumpal saat disemprotkan dari aplikator semprot. Berdasarkan hasil penelitian dapat disimpulkan bahwa Formula 1 memiliki karakteristik paling baik dari segi organoleptik, pH, viskositas, pola penyemprotan dan daya lekat. Konsentrasi carbopol 940 mempengaruhi karakteristik fisik dari sediaan spray gel hand sanitizer.

Kata Kunci: Carbopol 940; Minyak sereh wangi; Propilen glikol; Spray gel hand sanitizer

1. Introduction

Hands are used to carry out various activities so that they become the gateway for pathogenic microbes to entry the body. *Escherichia coli* and *Staphylococcus aureus* are bacteria often found on the hands. People use antibacterial soap or hand antiseptic (hand sanitizer) to maintain hands hygiene [1]. People prefer to use hand sanitizer because it can be applied anywhere and at any time without having to rinse it with water. One form of hand sanitizer preparation is spray gel. The spray gel preparation has several advantages due to low level of microbial contamination, more practical to use and has a longer drug contact time compared to other preparations form [2].

Hand sanitizer is a preparation containing alcohol with a percentage of 60-95%, therefore it has good bactericidal activity against gram-positive and gram-negative bacteria. According to the Food and Drug Administration (FDA), hand sanitizer can remove germs in less than 30 seconds. Besides, it can inhibit the growth of bacteria on hands, such as *Escherichia coli* and *Staphylococcus aureus* [3]. Unfortunately, high alcohol content in hand sanitizers will cause irritation to the skin when this preparation is used continuously. Natural ingredients having antibacterial activity can be used to replace alcohol content in hand sanitizer formula.

Citronella contains phytochemical compounds such as essential oils, saponins, polyphenols and flavonoids which can eradicate germs, viruses and bacteria [4]. The main components of citronella oil are citronellal and geraniol which have antibacterial properties [5].

There are two types of citronella oil, namely the Ceylon type (lenabatu) which is produced from *Cymbopogon nardus* and the Javanese type (mahapengiri) which is produced from *Cymbopogon winterianus*. *Cymbopogon winterianus* produces higher

content of citronellal and geraniol than *Cymbopogon nardus* so that the Javanese type has higher antibacterial activity [6].

Previous study conducted by Flora Yulen Pia Rumlus et al [7] proved that the Ceylon type of citronella oil could inhibit the growth of Escherichia coli and Staphylococcus aureus. Citronella oil with concencentration of 5% and 10% had medium category of antibacterial activity. Meanwhile, a concentration of 15% had strong category of antibacterial activity with an inhibitory zone diameter for *Escherichia coli* of 11.04 mm and *Staphylococcus aureus* of 11.81 mm.

Puspawati et al [8] reported the results of the antibacterial test of the Javanese type of citronella oil derived from leaves with a concentration of 100 ppm produced an inhibitory diameter of 10.25 mm in the strong category against *Escherichia coli* and 9.62 mm in the medium category against *Staphylococcus aureus*. The similar results was observed when using stem derived citronella oil at 100 ppm. It produced an inhibitory diameter of 10.62 mm in the strong category against *Escherichia coli* and 11.25 mm in the strong category against *Staphylococcus aureus*.

Hand sanitizer formulation generally contains gelling agents for generating the consistency of the preparation. Moreover, the formula also contains humectants which play a role in attracting water so that it can increase hydration in the stratum corneum of the skin. It is necessary to optimize the composition of these two components to obtain a spray gel with good physical properties [9]. Therefore, this study aimed to formulate spray gel hand sanitizer from citronella oil as natural antibacterial ingredient using carbopol 940 as gelling agent and propylene glycol as humectant.

2. Methods

Materials

Citronella oil was obtained from Triefta Aroma Nusantara, Indonesia. Carbopol 940, propylene glycol, polysorbate 80, methyl paraben, and triethanolamine were purchased from Dipa Prasada Husada, Indonesia.

Formulation of Spray Gel Hand Sanitizer

Four formulas of spray gel hand sanitizers were prepared from citronella oil (15%) using two different concentrations of Carbopol 940 (0.2% and 0.3%) and propylene glycol (10% and 15%). The formulations were presented on the Table 1.

Table 1. Formulation of spray gel hand sanitizer

Material	Function	Formula (% w/w)			
		F1	**F2**	**F3**	**F4**
Citronella oil	Active ingredient	15	15	15	15
Carbopol 940	Gelling agent	0,2	0,3	0,2	0,3
Propylene glykol	Humectant	10	10	15	15
Polysorbate 80	Surfactant	5	5	5	5
Methyl paraben	Preservative	0,1	0,1	0,1	0,1
Trietanolamine	Alkalizing agent	0,2	0,3	0,2	0,3
Aquadest	Solvent	69,5	69,5	69,5	69,5

First of all, carbopol 940 was dispersed into hot distilled water and stirred continuosly. Then, triethanolamine was added dropwise to adjust the pH to 6.0 hence a

clear gel was obtained. Methyl paraben was dissolved into propylene glycol in different beaker glass. The preservative solution was added into hydrogel and strirred homogeneously. Then, citronella oil and polysorbate 80 were poured into the mixture with gentle stirring. Finally the remaining distilled water was added up to 100 g.

Evaluation of Spray Gel Hand Sanitizer

Organoleptic test

The physical appearances of the spray gel hand sanitizer were observed visually in term of color, odor, consistency, clarity and separation [10].

Homogeneity test

Homogeneity testing is carried out to observe the presence of particles or substances that have not been mixed evenly. The test was conducted by spraying and spreading the spray gel preparation on a piece of transparent glass preparation [10].

pH test

The pH value of the preparations was determined using a pH meter (Mediatech). Then the pH value was compared with the range of pH requirements for topical preparations (4.5 - 7) [11].

Viscosity Test

The viscosity of the spray gel preparation was measured by a Brookfield viscometer using spindle No. 3. The viscosity value are recorded after the viscometer shows a stable number. Viscosity test was evaluated in triplicate [12]. A good spray gel viscosity value should be less than 150 cP [13].

Spraying Pattern

The preparations were sprayed on a sheet of mica plastic whose weight has been measured and numbered at a distance of 3, 5, 10, 15 and 20 cm. The spray formation pattern and the diameter of the spray pattern were observed. The preparation meets the acceptable criteria when it can be sprayed and the formed particles are small and evenly distributed [11].

Stickiness

The stickiness test of the spray gel preparation was carried out on the surface of the skin by spraying the preparation onto the upper arm from a distance of 3 cm. After the spray gel preparation was sprayed, it was counted for 10 seconds and observed the attachment of the preparation to the skin surface [14].

3. Results and Discussion

This study used citronella oil as active compound for spray gel hand sanitizer. Citronella oil purchased from Triefta Aroma Nusantara has specification that comply with Indonesian National Standard SNI 06-3953-1995. It has pale yellow to brownish yellow color, specific gravity of 0.880-0922, refractive index of 1.466 – 1.475, and minimum content citronellal of 35%.

The Javanese citronella oil derived from citronella grasses (*Cymbopogon winterianus)* was chosen to be formulated into spray gel hand sanitizer. The major components of the Javanese citronella oil were citronellal (32-45%), geraniol (11-13%), and citronellol. The Java type contains more rich consentrations of citronellal and geraniol than the other type derived from *Cymbopogon nardus.* Besides, the Java type also contains citronellyl acetate, L-limonene, ellemol and other sesquiterpene alcohols [15][16].

Several studies had proved antibacterial activity of citronella oil. The hydrophobic structure of citronella oil allows this compound to partition into lipid layer of bacterial cell membrane so that it generates disorganized cell membrane. The disrupted structural integrity of cell membrane leaks various ions and other cellular moleculs from the bacterial cell [17].

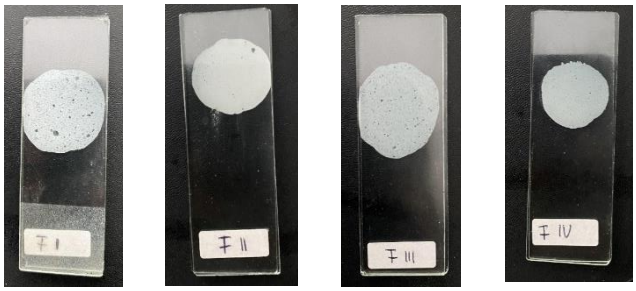
Carbopol 940, the gelling agent of the spray gel hand sanitizer of citronella oil, is an acrylic acid polymer. It is used widely as gelling agent in many topical preparations due to nontoxic and nonirritant propierties. Besides, it yields high viscosity at low concentration, has wide range of viscosity, compatible with many substances, and has low termosensitivity. This polymer has acidic properties which turn into gel by organic amines as neutralizing agent. Carbopol gel is formed by the association and entanglement of networks depended on the pH value. The dispersion of carbopol in aqueous medium is in the free acid form. The increasing of pH in hydration medium caused the ionization of carboxylic group on polymer chain to acquire negative charge so that it uncoil and expand the conformation [18].

Table 2. Organoleptic test of spray gel hand sanitizer

Formula	Color	Odor	Form
F1	White	Distinctive odor of citronella oil	Viscous liquid
F2	White	Distinctive odor of citronella oil	Viscous liquid
F3	White	Distinctive odor of citronella oil	Viscous liquid
F4	White	Distinctive odor of citronella oil	Viscous liquid

The distilled water was used as dispersion medium in the formulation of the spray gel hand sanitizer. The preparation is difficult to stabilize because there are two phases, the oil phase and the gel phase. Therefore to produce a stable preparation a surfactant must be added. This study used polysorbate 80 5% as surfactant. Initially, without surfactant, the hand sanitizer preparation was clear and non-homogeneous gel, however, after adding surfactant the gel preparation turned into white color and dispersed homogeneously. The addition of polysorbate 80 surfactant apparently not only changes the color of the preparation but also changes the form of the preparation into an emulgel because there are globules in the gel.

The obtained preparations were evaluated on the physical and chemical characteristics including organoleptics, homogeneity, pH, viscosity, spray pattern and sticky spreadability. The organoleptic test results of spray gel hand sanitizer were shown in Table 2.



(a) **(b)** **(c)** **(d)**

Figure 1. Homogeneity test of spray gel hans sanitizer. (a) F1; (b) F2; (c) F3; (d) F4

All formulas had similar organoleptic appearance as follow white viscous liquid with distictive odor of citronella oil. The aromatic odor of the preparations was produced by various compounds of citronella oil mainly citronellal, geraniol and citronellol. Citronellal, a major monoterpenoid fraction, is responsible for giving lemon odor. Geraniol also found in essential oil of ginger, rose, palmarosa gives a pleasant odor of roses [15]. Citronella oil is an essential oil so that it has high volatile compounds which giving a characteristic odor [17].

Table 3. pH and viscosity test of spray gel hand sanitizer

Formula	pH	Viscosity(cps)
F1	6.75±0.11	110.2±0.5
F2	6.82±0.26	113.2±0.1
F3	6.93±0.12	109.5±0.8
F4	6.71±0.13	113.5±0.5

The homogeneity test in Figure 1 showed that the four formulas were homogeneous. There were no solid coarse particles in the gel. The particle distributed homogeneously in the preparation.

The pH evaluation is carried out to ensure that the preparation will have the pH value in the range of 4.5 – 7 so that it does not cause irritation to the skin. As presented on table 3, the pH values of spray gel hand sanitizer were found to comply the acceptable pH of skin. Human skin has a pH range of 4 – 6 due to the production of fatty acids, sebum and amino acids by the sebaceous glands. This slightly acidic skin condition acts as a protective barrier preventing the growth of bacteria. The pH value also plays a role in maintaining the integrity of the stratum corneum. The formation of the lipid barrier in the stratum corneum involves enzymes whose activity is optimal at slightly acidic pH [19]. Ali & Yosipovitch [20] recommend that the ideal pH for products applied to the skin is 4.5 – 6.5. Alkaline preparations can cause skin irritation.

Table 4. Spraying pattern of spray gel handsanitizer

Formula	**Distance (cm)**	**Diameter (cm)**
F1	3	6.4 ± 0.5
	5	9.0± 0.2
	10	15.2± 0.3
	15	23.1 ± 0.8
	20	26.4± 1.1
F2	3	5.7 ± 0.3
	5	7.1±0.2
	10	8.7 ± 0.3
	15	11.1 ± 0.3
	20	10.8± 0.3
F3	3	7.5 ± 0.3
	5	10.2 ± 0.0
	10	14.9 ± 0.2
	15	20.3± 0.3

	20	24.3 ± 1.0
F4	3	5.3 ± 0.3
	5	6.4 ± 0.5
	10	8.9 ± 0.2
	15	10.0± 0.3
	20	10.1 ± 0.1

The viscosity test aim to measure whether the gel can be easily delivered via the spray applicator. The viscosity test was carried out using a Brookfield viscometer at a speed of 60 rpm and spindle number 1. Table 3 revealed that all formulas had viscosity less than 150 cPs. The viscosity value of F1 and F3 were relatively similar because they contained similar concentration of Carbopol 940 (0.2%). Likewise, F2 and F4 had the same carbopol concentration (0.3%) so their viscosity is similar. The pH value affects viscosity of carbopol gel. The acidic medium tend to form a low viscosity of carbopol gel. Neutralization carbopol dispersion with trethanolamine will generate a high viscosity gel [21].

F1 F2 F3 F4

Figure 2. The stickiness test of spray gel hand sanitizer

The results of the spraying pattern vary for each formula. This is caused by the spraying distance and the viscosity of the preparation. As shown in Table 4, it can be observed that the spray distance is directly correlated with the diameter, the greater the spray distance, the greater the diameter of spray pattern. The spray pattern of F1 and F3 tend to produce evenly distributed circular patterns with small particles due to the smaller concentration of carbopol 940. Meanwhile, the spraying pattern of F2 and F4 tends to be unevenly distributed, only accumulating at one point. The increasing concentration of carbopol enhance the viscosity so that the preparation is more difficult to spray from the spray applicator. Booq et al [22] reported that the viscosity of preparations was the main parameter affecting the gel spreadability, in which the higher viscous gel has lower spreadability.

Furthermore, the results of stickiness test of the spray gel hand sanitizer is presented in Figure 2. The results found that the preparations sticked to the skin surface during the test time. The spray gel has good stickiness when it remains attached to the skin for 10 seconds [13]. F1 and F3 had an even distribution, while F2 and F4 had an uneven and lumpy distribution. The viscosity of the prepations gives a parallel correlation with the spray pattern and stickiness results. If the viscosity is low then the resulting spray pattern is spread out with small particles, whereas if the viscosity is high the resulting spray pattern is lumpy. The ability to stick to the skin shows that the

preparation can retain the active substance on the skin thereby increasing its effectiveness [11].

4. Conclusion

Citronella oil was successfully formulated into spray gel hand sanitizer using Carbopol 940 and propylene glycol. The study showed that the optimal formulas was F1 containing concentration of carbopol 940 of 0.2% and propylene glycol of 10%. F1 was a a white viscous liquid with the distinctive odor of citronella oil, had pH of 6.75 ± 0.11, and viscosity of 110.2 ± 0.5 cps. Moreover, the spraying pattern and stickiness of F1 were the best distributed compared to the others formulas.

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