Design and Development of Peel-off Mask Gel Formulation of Citronella Oil for Acne Vulgaris

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Keyword: Citronella Oil, Peel-Off Mask Gel, Stabilit, Antibacterial Activity

Abstract: Cymbopogon nardus is a traditional plant that has chemical content of citronella oil with potential antibacterial activity. Antibacterial peel-off mask is very easy to applicate on face. This research aim is to develop and characterize the antibacterial peel-off mask gel containing citronella oil. This study used an experimental method. The peel-off mask was formulated using 8%, 10%, and 12% of polyvinyl alcohol (PVA) as a gelling agent. Evaluation of peel-off mask including physical characteristics (organoleptic, homogeneity, viscosity, spreadability, film drying time), chemical characteristic (pH value), stability and antibacterial activity. The organoleptic properties showed that the citronella oil peel-off gel mask was white, aromatic and had a soft texture. The variation in PVA affects the pH value, viscosity and film drying time of the peel-off mask, significantly. Stability test showed that all peel-off mask was no significant changes physically in the freeze-thaw method, but there was some color change and phase separation in the real-time method. The peel-off mask containing citronella oil using 8% PVA has the highest inhibition against Propionibacterium acnes almost the same with the positive control (clindamycin gel). Variation concentration of PVA in the formula was affected on the physical and chemical characteristics and antibacterial activity of the citronella oil peel-off mask gel, significantly. The preparation using 8% PVA gives the best formula with optimal results.

1 INTRODUCTION

Acne vulgaris is one of the most common skin diseases worldwide where it diagnosed in 80% of all teenagers (Griffiths, C., Barker, J., Bleiker, T., Chalmers, R., Creamer, 2016). About 83% to 85% of adolescent girls and 95% to 100% of adolescent boys are afflicted (Shalita, Del Rosso and Webster, 2011). Several major factors influence acne pathogenesis, including *Propionibacterium acnes,* which secretes various biologically active molecules and plays a role in the initiation of the local inflammatory response (Zouboulis, Katsambas and Kligman, 2013).

Most people with acne will first try to self-treat and some of them using antibacterial masks as treatment (Draelos, 2016) and choosing use natural products. One of the natural herbs is citronella (*Cymbopogon nardus*). Citronella oils contain citronellal, citronellol, and geraniol as several major components. It has shown antibacterial and antifungal activities in vitro, as active as penicillin against certain Gram-positive bacteria (Ehab, no date). Due to these data, the citronella oil (*C. nardus*) has potential as an anti-acne agent for facial skin treatment.

Peel-off mask is applied as a liquid film that thinly spread with fingers on the face and peels-off as a thin plasticized film (Suhery and Anggraini, 2016) after complete drying without leaving any residues (Rosa *et al.*, 2013). It can be repairing refreshing and tightening facial skin (Husni and Dewi, 2019), provides deep pore cleansing and skin debris removal (Suhery and Anggraini, 2016). Peel-off can also provide slight moisturizing action and enhances the occlusive effect (Rosa *et al.*, 2013), improving blood circulation, stimulating the activity of skin cells, removing dead skin cells, softening the skin and providing nutrients on the skin (Husni and Dewi, 2019)(Kulkarni, Gupta and Bhawsar, 2019). Mostly, the peel-off mask is based on polyvinyl alcohol (PVA) or polyvinyl acetate (PVAc), which causes tensor and occlusion effects (Nilforoushzadeh *et al.*, 2018).

Rahmasari, D., Ermawati, D., Nugraheni, R., Putri, D. and Pratiwi, I.
Design and Development of Peel-off Mask Gel Formulation of Citronella Oil for Acne Vulgaris.
DOI: 10.5220/0009126101570163
In *Proceedings of the 2nd Health Science International Conference (HSIC 2019)*, pages 157-163
ISBN: 978-989-758-462-6

The objective of this study was to develop and formulate citronella oil peel-off mask by using various concentration of PVA and evaluate by several test methods, which possesses antibacterial activity as an alternative of facial skin care product.

2 METHODS

Instruments
pH meter, Brookfield viscometer, autoclave, petri dish, incubator, climatic chamber, refrigerator.

Ingredients
Citronella (*Cymbopogon nardus L.*) oil, Polyvinyl Alcohol (PVA), Polyethylene glycol 400, Propylene glycol, Butylated Hydroxyl Toluene (BHT), Methyl Paraben, Ethyl Paraben, Disodium EDTA, Glycerin, Sweet orange oil, polysorbate 80 (Tween 80), and aquadest. *Propionibacterium acne* was used for antibacterial activity test and clindamycin gel was used as the positive control.

Peel-off mask gel preparation of citronella oil
The formulation of a peel-off mask gel of citronella oil was made according to the formula presented in Table 1. PVA was dispersed in 80% of the distilled water (85□ - 90□) in a water bath (mixture 1). PEG 400 was dissolved in the water (mixture 2). Methyl paraben and disodium EDTA was dispersed into the propylene glycol and glycerin then stirred until homogenous and added with Tween 80 and the remaining amount of water (20%) (mixture 3). Propyl paraben and BHT were dispersed into the citronella oil (mixture 4). Then added mixture 3 into mixture 2 homogenously and added again into mixture 1. Lastly, added some fragrance into mixture 1 and poured with mixture 4 homogenously until peel-off masks are formed.

Physicochemical evaluation of citronella oil peel-off mask gel
The physicochemical evaluation involved was organoleptic, homogeneity test, determination of pH value, viscosity, gel spreadability, and film drying time. Organoleptic was observed as its odor, color, and consistency visually. The homogeneity test was observed by applying the gel on transparent glass. pH value was measured by using pH meter and viscosity was determined by using Viscometer. Gel spreadability was measured by applying gel on transparent glass plates then loading by some loads. Film drying time was observed by applying the amount of gel on hands and wait until it formed a dry and elastic layer.

Stability evaluation
The stability evaluation was held using two methods, real-time and accelerated stability method. The real-time stability method evaluation was observed by storing the peel-off mask gel under the following storage conditions: 30±2°C (room temperature) and 40±2°C (climatic chamber). Each sample was analyzed for one month (28 days) according to the following parameters: organoleptic characteristic and pH value. The accelerated stability method evaluation was performed to see the stability of formulations at low (4ºC ± 2ºC) and high temperature (40ºC ± 2ºC) for 24 hours respectively. It was counted as one cycle and was analyzed for 6 cycles according to the several parameters: organoleptic characteristic, phase separation and pH value.

Table 1: Formulation of Citronella oil peel-off mask gel.

Ingredients	F1	F2	F3
Citronella Oil	0.5 %	0.5 %	0.5 %
PVA	8 %	10 %	12 %
PEG 400	2 %	2 %	2 %
BHT	0.7 %	0.7 %	0.7 %
Propylene glycol	2 %	2 %	2 %
Glycerin	2 %	2 %	2 %
Methyl paraben	0.1 %	0.1 %	0.1 %
Ethyl paraben	0.3 %	0.3 %	0.3 %
Tween 80	1 %	1 %	1 %
Disodium EDTA	0.02 %	0.02 %	0.02 %
Sweet orange oil	0.1 %	0.1 %	0.1 %
Aquadest	until 100%	until 100%	until 100%

Antibacterial activity test on citronella oil peel-off mask gel

The disc diffusion method was used to determine the zone of inhibition against *Propionibacterium acnes*. Antibacterial activity of citronella oil peel-off mask gel was tested by a modified well in agar method (Ilavenil, S, B. Kaleeswaran, B. and Ravikumar, 2010). Paper discs (6mm in diameter) were placed on MHA (Mueller Hinton Agar) media surface that been inoculated with bacterial suspension (Budiman *et al.*, 2017). Bacteria that were suspended with 0.9% NaCl equalized using nephelometer (BD Pheonix) with a standard of 0.5 Mc Farland (estimated 1.5 x 10^8 bacterial cells / mL) (Azrifitria; Aziz, 2010). Then poured the citronella oil peel-off mask gel into each well about 50µm using micropipette. Each gel was assayed in triplicate. The antibiotic (Clindamycin gel) was used as a positive control, while the negative control contained peel-off mask gel without citronella oil. Petri dishes were incubated at 37□ for 24 hours (Ilavenil, S, B. Kaleeswaran, B. and Ravikumar, 2010).

3 RESULTS AND DISCUSSION

Physicochemical evaluation of citronella oil peel-off mask gel

Organoleptic test was carried out to see the physical appearance of the preparation by observing the color, smell, consistency, and homogeneity of the preparation. The all peel-off masks gel are white. It had a distinctive smell of citronella oil and a hint of orange scent. These gels had a semi-solid consistency, soft and smooth, easy to apply, light to spread, thin and get thicker with the addition of PVA. Homogeneity of the preparation was shown by the absence of coarse particles (Ditjen POM, 1985), has a flat color and showed no phase separation (between citronella oil and peel-off mask gel base) in the preparation after its application to a transparent glass.

The pH value should be in the broad range of skin pH (from 4.0 to 7.0) to avoid any irritation to the skin (Ali and Yosipovitch, 2013) and should be lower than 5.0 for better skin condition (Lambers *et al.*, 2006). The result of pH value measurement was done in triplicate and average values calculated (Table 2). PVA is a water-soluble synthetic polymer that has a pH value of 5.0 – 8.0 (Rowe, Sheskey and Cook, 2009). The variation of PVA concentration in the citronella oil peel-off mask gel will affect the pH value of resulting preparation. The greater concentration of PVA, the higher the pH value of the preparation obtained. The pH of peel-off mask gel preparations was within the normal pH range of the skin.

A viscosity test was carried out to determine the thickness of a preparation and was measured using Brookfield viscometer. As shown in Table 2, the greater the concentration of PVA as a film-forming or gelling agent, the higher viscosity of the preparation was obtained. Viscosity in the peel-off mask gel is influenced by an increase in gelling agent concentration (Yuliani, 2010). PVA has substantial flexibility, tensile strength and hardness (Gaaz *et al.*, 2015). This will cause a greater binding and retention of the fluid by the gelling agent, therefore, the viscosity of the preparation increases.

A gel spreadability test was performed to examine the ability of citronella oil peel-off mask gel to spread when applied on the skin. The rate of spreading depends on the rate and time of shear produced upon smearing and also depends on the viscosity of the formulation (Garg *et al.*, 2002). In the topical preparation, spread is inversely proportional to its viscosity. Based on the results of testing the spreadability of peel-off mask gel, it was concluded that the increasing of PVA concentration affected the gel spreadability. The lower the viscosity, the higher the dispersal power (Chandira, R. M. Pradeep; Pasupathi, A.; Bhowmik, D. Chiranjib, Jayakar and K., K. Tripathi; Kumar, 2010). A good peel-off mask gel has a dispersion of 5 – 7cm (Rahmawanty, Dina; Yulianti, Nita; Fitriana, 2015) (Table 2).

Film Drying Time was carried out to find out how long the peel-off mask gel preparation will dry on the surface of the skin and form a layer of film. Ideally, the peel-off mask gel should dry between 15 – 30 min (Vieira *et al.*, 2009). As shown in Table 2, the drying test of the film showed that F3 dried faster than F1 and F2, this might be caused by the high concentration of PVA in the formula.

Table 2: Results of the physicochemical evaluation.

Parameter	F1	F2	F3
pH	4,80 ± 0,23	5,28 ± 0,58	6,11 ± 0,32
Viscosity (cps)	500 ± 0	1467 ± 115,47	3800 ± 1609,35
Spreadability (cm)	7,18 ± 0,32	6,67 ± 0,51	6,22 ± 1,15
Film Drying Time (min)	23,38 ± 0,08	21,18 ± 0,25	16,66 ± 0,30

Stability evaluation

Freeze-thaw stability evaluation was evaluated in 6 cycles (12 days) to verify signs of instability in the formulations under extreme conditions. The results are shown in Table 3 and Figure 1. It was verified that the optimized formulations stored at extreme temperature (4ºC ± 2ºC to 40ºC ± 2ºC for 6 cycles) did not show signs of instability, both in organoleptic parameters or in pH value.

Nevertheless, the storage at real-time method (28 days) with exposure in room temperature (30ºC ± 2ºC) and in climatic chamber (40ºC ± 2ºC) led to a sign of instability in phase separation. The citronella oil peel-off mask gel showed a visual phase separation in 3rd weeks on storage in room temperature and in 2nd weeks on storage in the climatic chamber (Table 4). The sedimentation phenomenon on phase separation led to a distribution gradient where larger particles were concentrated at the bottom of the glass bottle (Beringhs *et al.*, 2013).

Table 3: Result of the freeze-thaw stability evaluation.

Parameter	F1						F2						F3					
	Time (Cycle)																	
	1	2	3	4	5	6	1	2	3	4	5	6	1	2	3	4	5	6
Color	N	N	N	N	N	N	N	N	N	N	N	N	N	N	N	N	N	N
Odor	N	N	N	N	N	N	N	N	N	N	N	N	N	N	N	N	N	N
Consistency	N	N	N	N	N	N	N	N	N	N	N	N	N	N	N	N	N	N
Phase Separation	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS

N Normal, *NS* No Separation

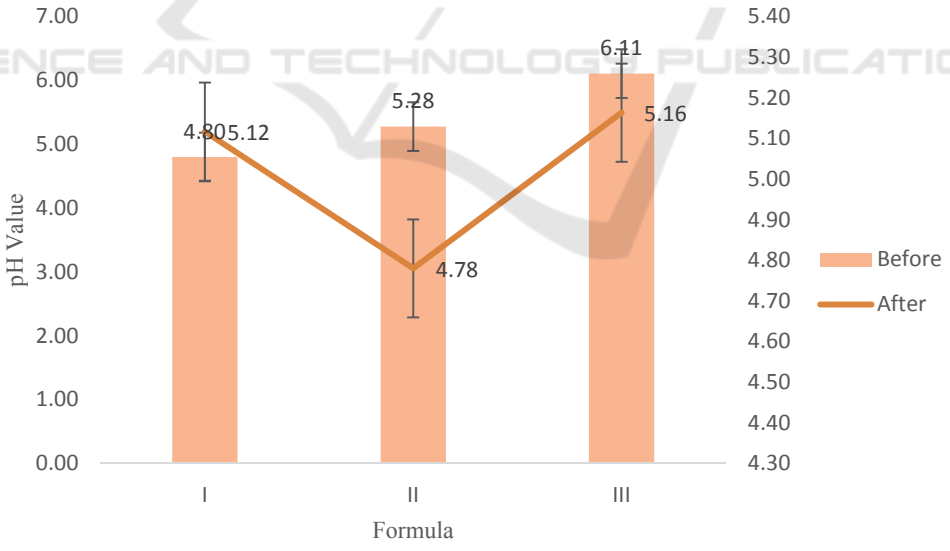

Figure 1: Variation of pH value on freeze-thaw stability method evaluation

Table 4: Result of the real-time stability evaluation

Parameter	F1				F2				F3			
	Time (Week)				Time (Week)				Time (Week)			
	1	2	3	4	1	2	3	4	1	2	3	4
	30±2°C				30±2°C				30±2°C			
Color	N	N	N	N	N	N	N	N	N	N	N	N
Odor	N	N	N	N	N	N	N	N	N	N	N	N
Consistency	N	N	N	N	N	N	N	N	N	N	N	N
Phase Separation	NS	NS	S	S	NS	NS	S	S	NS	NS	S	S
	40±2°C				40±2°C				40±2°C			
Color	N	N	N	N	N	N	N	N	N	N	N	N
Odor	N	N	N	N	N	N	N	N	N	N	N	N
Consistency	N	N	N	N	N	N	N	N	N	N	N	N
Phase Separation	NS	S	S	S	NS	S	S	S	NS	S	S	S

N Normal, *NS* No Separation, *S* Separation

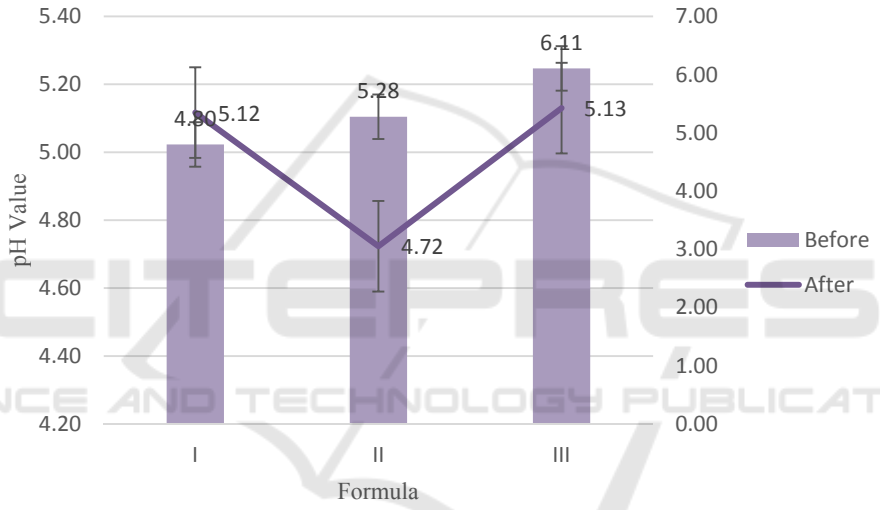

Figure 2: Variation of pH value on real time (30±2°C) stability method evaluation

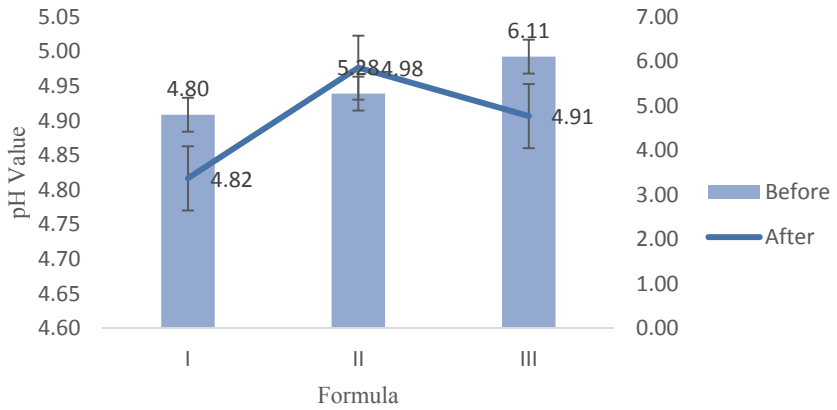

Figure 3: Variation of pH value on real time (40±2°C) stability method evaluation.

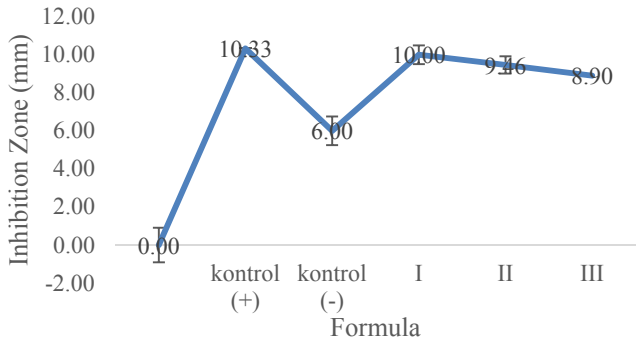

Figure 4: Results of antibacterial activity test Citronella oil peel-off mask gel

Antibacterial activity test of citronella oil peel-off mask gel

The result of the antibacterial activity test of citronella oil peel-off mask gel can be seen in Figure 4. The results of the antibacterial activity test showed that the citronella oil peel-off mask gel can inhibit the growth of bacteria *Propionibacterium acnes*. Most of the antibacterial activity from natural plants identified as phenolic and terpenoids. The compounds include citronellal, citronellol, and geraniol, which grouped as terpenoids (Bota, Welmince; Martosupono, Martanto ;Rondonuwu, 2015).

Terpenoids were considered as fast-acting compounds, since they inactivated organism in a short period, with mechanism of action should be related by the dysfunction or rupture of the cell membrane of bacteria (Guimarães *et al.*, 2019). The inhibition zone in the peel-off mask gel is influenced by an increase in gelling agent concentration. The test results demonstrate that the greater concentration of PVA as a gelling agent, the lower was the inhibition zone of the preparation. This can occur because of the greater trapping active compound by the gelling agent, therefore, the antibacterial activity of the preparation decreases.

4 CONCLUSIONS

The citronella oil peel-off mask gel with PVA has the best physical properties in Formulation 1 caused the formulation showed the greatest pH value, viscosity, spreadability and film drying time on human skin. Nevertheless, it was showed instability in 30±2°C and 40±2°C storage. The Formulation 1 of citronella oil peel-off mask gel has antibacterial activity against *Propionibacterium acnes* (MIC 10,00mm ± 0,7550) almost the same with the positive control (MIC 10,33 ± 0,9074). The result demonstrated that variation concentration on PVA was affected on physicochemical characteristics and antibacterial activity of citronella oil peel-off mask gel, significantly.

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