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Liquid Smoke from Coconut Shell Pyrolysis Process on Palm Surfactant Based Liquid Hand Soap

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ABSTRACT

The use of synthetic antibacterial liquid hand soap such as triclosan has begun to be avoided. Therefore it is necessary to find an antibacterial alternative that is safe for the skin and friendly to the environment. One of the environmentally friendly antibacterial alternatives is liquid smoke resulting from the pyrolysis process from coconut shells. The purpose of this study was to obtain the right concentration of liquid smoke for liquid hand soap made from palm MES surfactants and glycerol. The stages of the research were raw material analysis, liquid soap formulation (surfactant methyl ester sulfonate 7.5%, surfactant diethanolamide 5%, palm glycerol 9%, sodium chloride 1%, liquid smoke grade I, and distilled water). The treatments in this study were the addition of 1, 3, and 5% grade I liquid smoke. The next stage is an analysis of the physicochemical properties of the resulting liquid soap product, quality test was carried out based on SNI 2588: 2017, and a product effectiveness test. Liquid soap with the addition of 1% liquid smoke showed the best results with a density value of 1.037 g cm^{-3}, specific gravity 1.04, viscosity 11,560 cP, surface tension 29.08 dyne cm^{-1}, pH 7.2, free fatty acids 0.27%, ingredients insoluble in ethanol 0.14%, the total active ingredient is 12.52%, the number of plates is 990 CFU g^{-1}, the colony reduction is 61.13%, and has the minimal pungent aroma.

Key words: *antibacterial soap, diethanolamide, methyl ester sulfonate, triclosan*

INTRODUCTION

Maintaining hand hygiene is one of the most important things to prevent bacteria, germ, and virus transmission. Hand hygiene can be maintained by washing hands or by using hand sanitizers, but washing hands with soap is more effective in preventing germs and viruses (Nakoe *et al.* 2020). However, there are side effects to using excess hand soap such as dry skin, skin epidermis damage, irritation, and other dermatological symptoms caused by frequent hand contact with water and synthetic chemicals (Beiu *et al.* 2020). The soap used for washing hands has a big effect on the effectiveness of bacteria and virus prevention and skin health.

Soap is a product that dissolves in water and has the ability to remove dirt from the surface of objects such as the surface of the skin. Soap is generally

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divided into two types, namely liquid soap and solid soap. Liquid soap is currently more widely used because more practical and more hygienic than solid soap (Kholil 2020). Commercial liquid soap is made from a mixture of synthetic surfactants to clean dirt and achieve the desired physicochemical properties, such as pH, viscosity, stability, and foam properties. Unfortunately, synthetic surfactants are toxic to the skin and the environment. A study on the most widely used surfactant in hygiene products, sodium lauryl sulfate, showed skin irritation at a concentration of 0.5% in hand soap use. In addition to toxicity to the skin, toxicity to ecology and the environment was also found (Nazdrajic and Bratovcic 2019). Other ingredients that are commonly added to commercial liquid soaps, especially liquid hand soap, are antibacterial agents. Triclosan ($C_{12}H_7Cl_3O_2$) is the most commonly used antibacterial agent in liquid hand soap which has endocrine damaging properties that can cause genetic mutations in pathogens. Excessive use of antibacterial agents can cause antimicrobial resistance. Several pathogens such as *Escherichia coli* and *Bacillus cereus* have shown resistance to the antimicrobial benzalkonium chloride (Daverey and Dutta 2021).

Plant-based surfactants can be an alternative in the production of liquid hand soap that is environmentally friendly, biodegradable, and non-toxic to humans. methyl ester sulfonate (MES) is an anionic surfactant derived from palm oil which has a good surface tension lowering ability and good detergency compared to oil-based surfactants such as linear alkylbenzene sulfonates (LAS) (Ishak *et al.* 2017). Diethanolamide is a nonionic surfactant derived from palm oil which is easily soluble and resistant to hard water compared to anionic surfactants but is less effective in cleaning dirt than anionic surfactants. Soap usually consists of a mixture of several types of surfactants such as anionic and non-ionic surfactants to obtain the desired properties (Cheng *et al.* 2020).

Coconut shell liquid smoke is the result of the coconut shell pyrolysis process. Liquid smoke contains phenolic compounds, carbonyl compounds, and acidic compounds that have antimicrobial and antioxidant properties. Liquid smoke has been widely used as a natural food preservative because of its ability to inhibit bacterial growth. Liquid smoke is also used as an organic pesticide that is safe for the environment (Rizal *et al.* 2019). Liquid smoke can be an alternative to synthetic antimicrobial agents commonly used in liquid hand soap products. The study aims to obtain the characteristics, quality, and performance of three different concentrations of coconut shell liquid smoke in liquid hand soap, determine the best concentration of coconut shell liquid smoke, and analyze the effect of coconut shell liquid smoke addition on the palm surfactant-based liquid hand soap.

MATERIAL AND METHODE

This research conducted by producing three different formulations of liquid hand soap from palm surfactant MES and DEA, palm glycerol, and liquid smoke grade I as an antibacterial agent (Table 1). The liquid smokes concentrations used are 1, 3, 5% (w/w) of the total mass of liquid hand soap produced.

In the liquid hand soap production, MES surfactant, DEA surfactant, palm glycerol, and distillate water were mixed and heated in a 500 mL beaker glass using a magnetic stirrer at 60 °C for 30 minutes until homogeneous. Then the mixture cooled until the temperature reach 30 °C.

Table 1 Liquid hand soap formulation

Material	Formula		
	1	2	3
MES (%) w/w	7.5	7.5	7.5
DEA (%) w/w	5.0	5.0	5.0
Glycerol (%) w/w	9.0	9.0	9.0
Grade I liquid smoke (%) w/w	1.0	3.0	5.0
NaCl (%) w/w	1.0	1.0	1.0
Distilled water (%) w/w	76.5	74.5	72.5
Total	100.0	100.0	100.0

Then NaCl and antibacterial ingredients (liquid smoke or triclosan 0.3%) were added and mixed to the mixture using a magnetic stirrer at 30 °C for 30 minutes until homogeneous. The treatment used in this study was the concentration of liquid smoke i.e. 1, 3, and 5%. Liquid hand soap with triclosan 0.3% and base soap were used as a comparison sample.

Analysis of samples is observed on the appearance, characteristics, quality, and performance parameters. Characteristic parameters include density, specific gravity, viscosity, and surface tension. Quality analysis was conducted based on SNI 2588: 2017, including analysis of pH, total active ingredients, ethanol insoluble ingredients, free fatty acid levels, and microbial contamination (TPC method). The performance of liquid hand soap produced was analyzed using a colony reduction test and compared with comparison samples.

The experimental design used in this study is a completely randomized design with three different treatments of liquid smoke and three replications. the experimental design has a mathematical model as follows.

$$Yij = \mu + \sigma i + \varepsilon ij$$

Where:
Yij, the value of the j-th observation of the effect of the liquid smoke i-treatment; μ, general average; σi, effect of liquid smoke treatment-i; εij, effect of error; I, number of treatments of liquid smoke (1, 3, 5%); j, deuteronomy.

The data obtained were then analyzed using the ANOVA method. If between treatments shows significantly different results, a further test is carried out using the Duncan's Multiple Range Test with a 95% confidence level. The entire data analysis process was carried out using the software Microsoft Office Excel and SPSS for windows version 15.0.

RESULT AND DISCUSSION

The appearance of three different concentrations of liquid smoke liquid hand soap was observed in the shape, homogeneity, color, and aroma (Table 2). The appearance of a liquid hand soap product is one of the critical characteristics that should be considered because it affects s consumer acceptance of the product (Rochima *et al.* 2015).

Characteristics of the liquid hand soap was analysed with parameter of densiti, specific gravity, viscosity, and surface tension. The density of hand soap is the ratio of hand soap mass with its volume. Meanwhile, specific gravity is a measure of hand soap density with the density of pure water. Viscosity indicates the ease with which the soap flows. Surface tension is the condition of the surface of a liquid that tends to tighten and is influenced by the cohesion force between water molecules. Soap is one of the ingredients that can lower the surface tension of water. All of the samples have a density value ranging from 1.037 – 1.038 g cm^{-3} and specific gravity of 1.040. Specific gravity values are met the requirement. The addition of liquid smoke did not significantly affect the density and specific gravity of liquid hand soap (Table 3).

Table 2 The appearance of three different concentrations of liquid smoke hand soap

Parameter	Concentration of liquid smoke hand soap (%)		
	1	3	5
Shape	Liquid	Liquid	Liquid
Homogenity	Homogeneous	Homogeneous	Homogeneous
Color	Transparent, yellowish	Transparent, slightly yellow	Transparent, yellow
Aroma	Slightly pungent	Pungent	Very pungent

In most cases, the values can change by sugar, sodium lactate, or glycerin added in the formulation of liquid hand soap. This analysis can determine whether a material can be dissolved with other substances or not in the soap formulation (Uzwatania *et al.* 2020). In the viscosity test, all samples are meet the requirement but there is a decrease in the liquid hand soap viscosity with the increase of liquid smoke concentration added. This parameter affects the filling of soap into the packaging container and its application. The higher the viscosity of soap, the more difficult it is to pour (Christiani 2015). In the surface tension analysis, the lowest result is 1% Liquid Smoke formulation. According to Bhattarai *et al.* (2020), the lower the surfaces tension of the soap, the higher the cleaning power and vice versa. This means the addition of liquid smoke increase the surface tension of water, the liquid smoke may affect the cohesion force between the water molecules.

The characteristics test result of liquid hand soap products with three different concentrations of liquid smoke is presented in Table 4. The quality of liquid hand soap was analysed based on SNI 2588: 2017. pH parameter in soap products affects the skin power adsorption. pH should be in a range of 4.0-10.0 for safetiness. Free fatty acids are fatty acids in soap that are not bound as sodium compounds or triglyceride compounds. The high level of free fatty acids in soap will reduce its cleaning power (Simanjuntak 2018). Materials that are insoluble in ethanol can be left on the skin in the rinsing process because they are not soluble with water or polar (Lestari *et al.* 2020). The amount of fatty acids is the total amount of fatty acids in soap and free fatty acids (Rohman *et al.* 2020). The total plate count of the product calculates the presence of microbiological contamination contained in a product (Uswah *et al.* 2019).

There are a decreased of pH on the addition of liquid smoke (Table 4). The addition of low pH liquid smoke causes the pH of the liquid hand soap to decrease as the liquid smoke concentration increases. A low pH value of liquid smoke indicates good quality because it is related to its use as an antibacterial (Kausa 2012). There are an increase in free fatty acid levels with an increase in the concentration of liquid smoke. From the result, liquid smoke 5% was not meet the

Table 3 The characteristics of three different concentrations of liquid smoke hand soap

Characteristics	Formulation (%)			Requirement
	1	3	5	
Density (g cm^{-3})	1.037	1.037	1.037	-
Specific gravity	1.040	1.040	1.040	1.010 - 1.100
Viscosity (cP)	11,560	6,806	5,626	500 - 20,000
Surface tension (dyne cm^{-3})	29.080	29.260	29.480	The lowest

Table 4 The quality of three different concentrations of liquid smoke hand soap

Parameter	Formulation (%)			Requirement
	1	3	5	
pH	7.20	7.10	6.80	4.00 - 10.00
Free fatty acid (%)	0.27	0.67	1.24	Max 1.00
Ethanol insoluble material (%)	0.14	0.11	0.09	Max 0.50
Total active ingredients (%)	12.52	12.53	13.92	Min 10.00
Total plate count (CFU mL^{-1})	990	350	280	Max 1 x 10^3

requirement. This means, liquid smoke has fatty acid that not bound as a triglyceride compounds and can affect to its cleaning power. There are no significance differences on the ethanol insoluble material of liquid hand soap. The analysis results show when the rinsing process, all of soap formulation able to clean dirt and didn't leave a resideu. The result of total active ingredients also didn't have any significance differences. For the total plate count test, there are a decrease of bacteria in the addition of liquid smoke. This means liquid smoke has an antibacterial properties to prevent the growth of bacteria. The total plate count results still in the range of requirement.

Product effectiveness was calculated by observing the decrease in colony growth before and after using soap products. There is an increase in the percentage of colony reduction from 1% to 5% liquid smoke. The highest colony reduction was found in the soap formulation with 5% liquid smoke. The comparison soap sample in the form of 0.3% triclosan and control had results below 5% liquid smoke, namely 68.57% and 27.71% (Figure 1). This shows that the addition of liquid smoke concentration into liquid soap products can increase the effectiveness of soap in inhibiting bacterial growth.

The best liquid smoke concentration was determined based on the parameters of the characteristics, quality, and performance of the liquid hand soap. Of the three parameters, the quality parameter is the most important parameter to consider because it relates to the requirements of the Indonesian national standard (SNI 2588: 2017) so that liquid hand soap produced must comply with the established standards. The soap performance parameter shows the ability related to the function of liquid hand soap, namely eliminating or inhibiting the growth of bacteria on the hands. The appearance parameter on

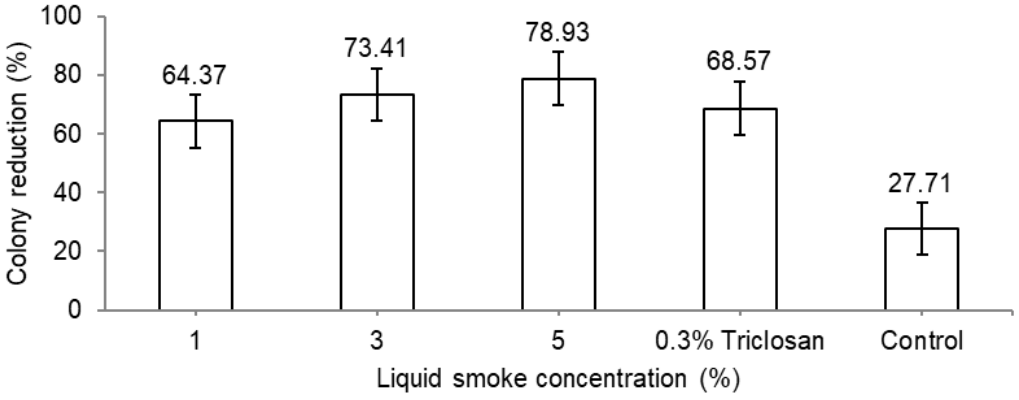

Figure 1 Liquid smoke concentration on the colony reduction of bacteria.

the aroma of liquid hand soap needs to be considered because liquid smoke has an effect on the pungent aroma of the soap. The characteristic parameter of surface tension is related to the way soap works to reduce the surface tension of water and clean dirt. Meanwhile, characteristic parameters such as density, specific gravity, and viscosity are related to consumer preferences.

The resulting product needs to meet the criteria set out in the Indonesian national standard. This is intended to monitor and guarantee the quality of a product. Products that meet standards can be commercialized while products that do not meet standards can indicate that the product is not safe to use. In the analysis of the quality of liquid hand soap based on Indonesian national standards, liquid smoke soap 5% does not meet the free fatty acid criteria because its content exceeds 1%. Meanwhile, soap 1% and liquid smoke 3% have met all the requirements. Based on product quality, liquid smoke 1% and 3% can be considered the best concentrations.

The next parameter to consider is the liquid hand soap performance parameter in reducing the number of bacterial colonies on the hands when using soap. This parameter needs to be considered because it correlates with the function of antibacterial liquid hand soap. Based on the results of the analysis, the colony reduction in liquid smoke 1% was 61.13%, and liquid smoke 3% was 75.08%. Compared to plain soap which has a colony reduction of 27.71% and triclosan soap of 68.57%, soap with the addition of liquid smoke can compete with antibacterial soaps on the market.

The aroma parameter is a critical parameter that needs to be considered. Liquid smoke has an acid smoky aroma that is not commonly used in cleansing and beauty products. The pungent aroma can make it difficult for consumers to accept liquid smoke hand soap products. The aroma of liquid smoke is a parameter that wants to be minimized in liquid hand soap products. Based on observations, liquid smoke 1% has a slightly pungent aroma compared to liquid smoke 3%. It can be concluded that on the aroma parameter, liquid smoke 1% is the dominant consideration in the production of liquid hand soap.

In surface tension analysis, liquid smoke 1% has the lowest surface tension result so that it can lower the surface tension of the water better and clean the dirt better. This reinforces the consideration of liquid smoke 1% as the best concentration. Meanwhile in the analysis of density, specific gravity, and viscosity because all of the concentrations have a good result, are not significantly different, and meet the requirement. With these considerations, it can be concluded that liquid smoke 1% is the best concentration because it meets the standard, has competitive antibacterial performance, and has the least pungent aroma.

CONCLUSION

The formulation of liquid hand soap is using palm oil-derived surfactants, namely methyl ester sulfonate surfactant with a concentration of 7.5% and diethanolamide surfactant with a concentration of 5% to replace oil-based surfactants which are not environmentally friendly. The antibacterial agent used in the formulation of liquid hand soap is a liquid smoke grade I resulting from the pyrolysis process of coconut shells. Palm glycerol 9%, NaCl 1%, and aquades are also used in the formulation of liquid hand soap. The treatments used in this study were liquid smoke with concentrations of 1, 3, and 5%. The results showed that of the three concentrations, liquid hand soap with the

addition of liquid smoke 1% produced the best test results. The best liquid hand soap has a density value of 1.037 g cm^{-3}, specific gravity 1.040, viscosity 11,560 cP, surface tension 29.08 dyne cm^{-1}, pH 7.2, free fatty acids 0.27%, ingredients insoluble in ethanol 0.14%, the total active ingredient is 12.52%, the number of plates is 990 CFU g^{-1}, the colony reduction is 61.13%, and has the least pungent aroma.

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