Microemulsion Stability of Virgin Coconut Oil Based on Tradition of Melala Sumbawa's Society

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Abbreviation: virgin coconut oil VCO

Abstract: The Melala is a tradition of the Sumbawa people who use coconut milk to obtain Sumbawa oil for traditional medicine. Coconut milk is a colloidal system of stable oil in water (O/W) microemulsion. Within a particular time, the emulsion will split to produce oil (VCO), protein, and water due to the colloid equilibrium on the stability of the coconut milk emulsion. The purpose of this study was to compare the microemulsion stability of the coconut milk colloid system. The VCO was isolated by heating, enzymatically, acidifying, and adding whitening methods for comparison. The physical stability of the O/W microemulsion was measured by the volume of VCO produced from various isolation methods in simple laboratory experiments. The results showed that the physical stability of the O/W microemulsion on VCO isolation by enzymatic method using papain enzyme was the least.

Keywords: Melala Tradition, Virgin Coconut Oil, Physical Stability of Microemulsion M/A

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INTRODUCTION

Melala tradition is a tradition of processing coconut milk as the basic ingredient for making Sumbawa oil which has been carried out for generations [1]. Coconut oil from melala is used as cooking oil to meet food needs in daily life [2]. Coconut oil is obtained from the raw material extract of dried coconut flesh [3]-[4]-[5]. Coconut oil has many benefits for humans. It can be used as industrial raw material, namely as cooking oil. In addition, it can be used to maintain health and cure various diseases such as cancer, cholesterol, diabetes, heart [6]. It is partly because of the high lauric acid content in coconut oil [7]-[8]-[9].

Coconut milk in the Melala tradition is a colloidal system of oil-in-water microemulsions. A microemulsion is a thermodynamically stable and transparent system.

It is a dispersion of oil and water which is stabilized by a thin layer (film) of amphiphilic molecules (surfactant and cosurfactant) [10]. Protein acts as an amphiphilic molecule in coconut milk solution. Oil is soluble in water due to proteins that coat the oil so that the oil is trapped in the water solvent. The interaction between oil droplets depends on the quantity and quality of the protein [11]. The coconut milk colloid system is a surface tension balance between protein and water. The voltage balance determines the amount of coconut oil produced [12]-[13]. Coconut milk colloids are also affected by the pressure equilibrium of the oil-water molecular homogeneity. Small oil droplet size in coconut milk emulsion is achieved at high homogeneity pressure. [14]-[15]. The type of fatty acid or components in the oil phase can affect the stability of the microemulsion [16]. The colloidal condition of coconut

milk can be maintained by maintaining a protein-oil and protein-water surface tension balance and an oil-water homogeneity pressure balance.

The colloidal balance of coconut milk is the focus of attention in producing coconut oil. The high demand for coconut oil requires an effective method to break the colloidal balance of coconut milk. A disturbed colloid system will create three layers: oil, blondo, and water [17]-[18]. On the traditional to modern production scale, various methods are applied to separate the oil and water in coconut milk. Starting from heating, the use of enzymatic, acidification, and the addition of whiting. Each method used will give a different effect and quality of the oil. Production costs are also a consideration in selecting oil and water separation methods [19]-[20].

The method used plays a role in disrupting the stability of the coconut milk colloid system [21]. Of the four methods, it is necessary to estimate the VCO isolation method with a large volume and good quality. Estimation was conducted by designing experiments using heating methods, enzymatic, acidifying, and adding whiting to coconut milk colloids. Not only volume and quality, but the time of solution separation is also a priority in estimating the isolation method. Thus, the estimation results can find an effective method in producing VCO oil by disrupting the physical stability of oil-in-water microemulsions to reduce community production costs.

MATERIALS AND METHODS

Research

Materials and Tools

The materials used in this study were grated old coconuts obtained from Lombok traditional markets, papaya, lime, whiting, water, pH indicator paper, and label paper. The tools used in this study were 600 mL plastic bottles, grater, coconut filter, filter cloth, scales, measuring cup, dropper, spoon, thermometer, stirrer, and hot plate.

Work Procedures

The procedure in this study has several stages, namely the preparation of raw materials, treatment for each method, and observation.

Preparation of Coconut Milk (Santan)

The coconut used is ready, the flesh is taken, and then grated. Freshly grated coconut is weighed as much as 2500 grams, then water is added in a ratio of 1: 1, then squeezed, and coconut milk is produced (virgin coconut). The coconut milk was put into 5 plastic bottles of 500 mL each and labeled with numbers 1 to 5. Bottle label 1 as a control contained 500 mL of coconut milk, while bottles labeled 2, 3, 4, 5 were given the method treatment.

Heating Method

Coconut milk in bottle label 2 was heated using a hot plate at a temperature of 60°C–70°C for 20 minutes, then allowed to stand for 24 hours.

Papain Enzyme Method

Fresh young papaya peel was grated and weighed as much as 50 grams, then added water in a ratio of 1: 1, then stirred, squeezed, and filtered. The papain enzyme extract was measured as much as 10 mL, put into bottle label 3, then stirred and allowed to stand for 24 hours.

Acidification Method

Used fresh lime cut and squeezed water then measured the pH of the solution with pH indicator paper. The acid solution obtained was measured as much as 10 mL, added to bottle label 4, then stirred and allowed to stand for 24 hours.

Calcium Hydroxide (Whiting) Method

The whiting powder is weighed as much as 50 grams and added water in a ratio of 1:2. Then the solution is stirred and filtered. The whiting solution obtained was measured as much as 10 mL, added to bottle label 5, then stirred and allowed to stand for 24 hours.

Observation Parameter

The parameters observed in this study included the physical stability of the microemulsion of the coconut milk colloid system. The stability of the oil-in-water microemulsion was determined based on the volume of VCO obtained against the time of the oil-water separation process. Then the results of the microemulsion stability obtained were analyzed. It is descriptively based on the % yield in each method and the physical properties of the isolated VCO oil.

RESULTS AND DISCUSSION

The experimental results of the isolation of *virgin coconut* oil with various traditional production methods are presented in Table 1, Figure 1, and Figure 2. Based on Table 1 and Figure 2, it can be seen that the highest volume of VCO isolation was obtained by the enzymatic method, namely 88.5 mL with a % yield of 17.7%. Yield is the ratio between the volume of VCO produced and the volume of thick coconut milk used. According to the research of Onsaard et al., the yield of oil content (VCO) in 1 liter of coconut milk is a maximum of 35.2% [22]. However, compared with the highest yield obtained in this study, which is 17.7% in 500 ml of coconut milk, it is almost close to the maximum VCO results. It is because the holding time is carried out for 24 hours obtained can be more than that.

Table 1. Results of VCO Isolation After Isolation for 24 Hours

Method	VCO Volume	% Yield	Physical Properties
Control	22 mL	4,4%	The color of the oil is slightly cloudy, slightly runny, the smell tends to be fresh coconut, and the amount of blondo (protein) is medium.
Heating Method	0 mL	0 %	The amount of blondo (protein) is pure white.
Enzymatic (Papain Enzyme)	88,5 mL	17,7 %	The color of the oil is yellow, slightly thick, the smell tends to be fresh coconut, and the amount of blondo (protein) is a little Acidic.
Acidification (Lime)	25,2 mL	5,04 %	The oil color is clear (colorless), slightly runny, the smell tends to be fresh coconut, and the amount of blondo (protein) is medium.
Calcium Hydroxide (Whiting)	1,2 mL	0,24 %	The oil color is clear (colorless), slightly runny, the smell tends to be fresh coconut, and the amount of blondo (protein) is medium.

Figure 1. The volume of VCO for each method **Figure 2.** Volume of VCO Isolation for each Method

In addition, the more solvent, the more oil is extracted [23]. In this study, the volume control of coconut milk was used, namely 500 mL, so that it affected the volume and yield of VCO obtained

Based on Figure 1, it can be seen that four plastic cups show the results of VCO insulation with different colors. The physical properties of each VCO isolation method used also show variations.

Control

The test results for bottle label 1, used as a control (no treatment) containing 500 mL of coconut milk, were allowed to stand for 24 hours. Three layers were formed, namely, on the top layer, there was a blondo (protein) layer, then the middle layer was oil, and the bottom layer was water. The test results for the control bottle can be seen in Figure 3.

Figure 3. Control Bottle Residual Results

Based on the results of standing for 24 hours, 22 ml of VCO isolation was obtained in the control bottle with a yield of 4.4%. The color of the VCO produced is a bit cloudy, slightly runny, smells like fresh coconut, and the amount of blondo (protein) is medium when compared to the other four bottles.

Oktaviana's research on the coconut milk emulsion microscope structure in Figure 4 [24] shows that the emulsion particles in coconut milk are denser and smaller. It indicates that the physical stability of bottle 1 coconut milk emulsion is quite large.

Figure 4. Microscopic Overview of Untreated Coconut Milk Emulsion (45x Magnification) [24]

Heating Method

The heating method resulted in 0 mL of VCO isolation (no oil separation). It is shown in Figure 5 that two layers are formed, namely the blondo (protein) and water layers. In this study, heating was carried out at 60°C - 70°C for 20 minutes. According to Nely et al. in their research, it was stated that the higher the temperature, the higher the temperature, the higher the emulsion stability, and the lower the yield of VCO obtained [23]. And suppose you look at the microscopic structure of the coconut milk emulsion with the heating method as shown in Figure 6. In that

case, it shows that the emulsion particles are large and loose [24], which offers high emulsion stability. However, this is not by the results obtained by the researchers because the heating method is not carried out continuously but for 20 minutes so that the breaking of the bond between oil and water in the coconut milk emulsion does not occur.

Figure 5. Results of Heating Method

Figure 6. Microscopic Overview of Coconut Milk Emulsion Heating Method (45x Magnification) [24]

In the heating process of coconut milk, the boiling point increases. The colligative properties of the coconut milk emulsion in the heating process depend on the presence of dissolved particles and their number. The mass of the coconut milk solution is 500 mL in 500 g of grated coconut. The molecular weight of the coconut oil fatty acids is 205 g/mol with the price of $K_b = 0,52^\circ\text{C} / \text{molal}$ [25]. So the price is calculated boiling point elevation (ΔT_b) gained by $2,53^\circ\text{C}$.

Enzymatic Method

In the enzymatic method used papaya fruit skin. Papaya peel contains the enzyme papain, namely proteinase. The nature of the enzyme can decompose the protein that covers the oil, which can cause the bonds between the oil and protein to separate. So it can speed up the process of making VCO. The following are the results of standing for 24 hours in a bottle containing 500 mL of coconut milk and 10 mL of papain enzyme extract in Figure 7.

Based on Figure 7, three layers are formed, namely, on the top layer, there is a blondo (protein) layer, then the middle layer is oil, and the bottom layer is water. The highest volume of VCO is obtained compared to other methods, namely 88.5 mL and % yield of 17.7%. The color of the oil is yellow, slightly thick, the smell tends to be fresh coconut, and the

amount of protein is small. The color of the VCO isolation is yellow in the enzymatic method. It is due to the influence of yellowish-green pigments (chlorophyll and carotene) contained in the skin of papaya fruit which is soluble in oil. According to Suirta and Astitiasih's research, the chlorophyll ($C_{55}H_{72}O_5N_4Mg$) contained in papaya leaf extract was more distributed in the fraction, as seen from the color of the fraction, which was still green [26].

Figure 7. Results of the Enzymatic Method

Figure 8. Microscopic Overview of Coconut Milk Emulsion using the Enzymatic Method [24]

Based on **Figure 8**, it can be seen that the microscopic structure of the coconut milk emulsion using the enzymatic method [24] shows that the emulsion particles are very loose and large compared to other methods. It shows that the stability of the emulsion is very small so that the breaking of the bond between oil and water is very large and accelerates the formation of VCO isolation as the results obtained in the study, namely the volume of VCO as much as 88.5 mL.

Acidification Method

The acidification method manufactures VCO by creating an emulsion atmosphere in coconut milk in an acidic state. Acid can break the bond between oil and protein. Oil will form on the top layer because the density is the lightest, blondo (protein) in the second layer, and water will form on the bottom layer.

In this study, the acid from lime fruit was used by mixing it into coconut milk. The results showed that three layers were formed, as shown in Figure 9, with a volume of 25.2 mL of VCO isolation and 5.04% yield %. The volume in the acidification method is caused by the amount of water content in the coconut milk volume. The color of the oil (VCO) produced is clear (colorless), slightly runny, the smell tends to be fresh

coconut, and the amount of blondo (protein) is medium.

In the acidification method, the most optimal pH is a mixture of coconut milk and acid, pH 4.3 [27]. However, in this study, a pH of 1 was used so that the amount of VCO obtained was slightly higher than the control.

Cream of coconut milk emulates the type O/A (oil in water), and protein acts as a stabilizing agent in the emulsion (O/W) process. The equilibrium colloidal O/A is affected by pH. The O/A equilibrium occurs at pH 4.3. The addition of H^+ will lower the pH so that the M/A balance is disturbed (separated).

Figure 9. Residual Results of the Acidification Method

Figure 10. Microscopic Overview of Coconut Milk Emulsion on Acidification Method [24]

Calcium Hydroxide (Whiting) Method

The volume of VCO isolation in the whiting method was 1.2 ml, and the % yield was 0.24%. The volume of VCO obtained was less than the control, but by using the whiting method, the color of the VCO was clearer (colorless), slightly watery, the smell tended to be fresh coconut, and the amount of blondo (protein) was medium. In contrast, in control, the VCO color was more cloudy. Betel lime ($Ca(OH)_2$) contains strong alkaline properties that can produce a more transparent VCO color than the control. Proteins in coconut milk emulsions can react to form complexes by adding metal ions from salt ($CaSO_4$, $MgSO_4$). Where in whiting with Ca ions can disrupt the stability between oil and water (O/A) [28].

Figure 11. Results of the Calcium Hydroxide (Whiting) Method

O/W Emulsion Stability (Gibbs Free Energy)

In the "Melala tradition" coconut cream is a colloidal equilibrium of oil-in-water emulsion assisted by protein as a stabilizing agent. Coconut milk colloid has been quite stable for a long time. The equilibrium reaction for an oil-in-water emulsion is as follows:

The equilibrium reaction occurs due to the role of the stabilizing protein in the oil-in-water emulsion. The protein structure has two distinct polar properties. The functional groups are polar, and the straight-chain carbon groups are non-polar. The polar group binds to the water molecule (polar-polar), while the carbon group binds to the oil (nonpolar-nonpolar). This event causes the oil to be trapped in the water. The protein that coats the oil cannot last long, reducing its role as a stabilizer between oil and water. The process causes the separation of oil and water.

The oil-in-water emulsion system depends on Gibbs's free energy. Emulsion equilibrium occurs when Gibbs's free energy is zero. The O/W emulsion also results in the interaction of the surface tension of the protein with water and the surface tension of the protein with the oil. The surface tension maintains the system. The following equation can determine the Gibbs free energy:

$$G = \partial d A d + \partial e A e$$

Information:

∂d = protein-oil interfacial tension

∂e = protein-water interfacial tension

A = surface area

Total Gibbs free energy

$$G = g p \cdot m + g p \cdot a$$

At equilibrium $G = 0$, then

$$G = G^* + RT \ln K$$

$$G^* = -RT \ln K$$

$$G^* = -RT \ln \frac{\partial e}{\partial d}$$

G^* = standard Gibbs free energy

In the emulsion system in the presence of protein (zantap) where at equilibrium, the Gibbs's free energy is determined by the protein-oil interfacial tension and protein-water interfacial tension. Bao, Wang, and Li suggested the optimal conditions for preparing a sterilized coconut milk drink as follows: coconut: water ratio 1:10, pH 6.5, sugar 4%, homogenization at 20-25 MPa, and sterilization at 121°C for 20 min. Surface-active stabilizer (whey protein isolate (WPI), sodium caseinate [29]. The addition of surfactant reduced the homogeneous tension to 10 Mpa (emulsion stabilized (0.4 m oil drop by sodium caseinate stabilizer)) [30].

CONCLUSION

Colloidal equilibrium occurred between stress protein-oil and protein-water tension in coconut milk solution. The physical stability of the microemulsion O/W in various VCO isolation methods is the smallest by using the enzymatic method with papain enzymes. It is evidenced by the results of the VCO isolation method at most 88.5 mL.

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