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The Role of Carboxymethyl Cellulose (CMC) from Durian (*Durio zibethinus*) Rind in Enhancing Coconut Milk Stability Under Different Temperature Condition

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Abstract: The influence of carboxymethyl cellulose (CMC) concentration derived from durian rind (*Durio zibethinus*) and temperature variations on coconut milk quality was investigated through three stages. The first stage involved the isolation of α-cellulose from durian rind powder, which was analyzed using FTIR to confirm its identity as cellulose. In the second stage, a series of processes including alkalization with isopropanol and NaOH, carboxymethylation with NaMCA, and neutralization using 90% CH_3COOH and ethanol were conducted, followed by purification with distilled water, centrifugation, and the addition of acetone. This resulted in CMC, which showed positive qualitative analysis and FTIR peaks similar to those of commercial CMC. The final stage focused on coconut milk production, where CMC was incorporated at concentrations ranging from 0.5% to 0.7%, and temperature variations were set at 16ºC, 25ºC, and 80ºC. The quality of the resulting coconut milk was assessed for stability, pH, viscosity, moisture content, protein content, fat content, free fatty acid content, and organoleptic properties. The optimal results were achieved with the addition of 0.7% CMC at 16ºC, yielding a stability percentage of 34.78% with a shelf life of 9.5 hours, a pH of 6.21, viscosity of 12.6138 x 10^2 cP, moisture content of 45.81%, protein content of 7.57%, fat content of 43.51%, and free fatty acid content of 1.402%. Organoleptic tests indicated that panelists favored the coconut milk with 0.7% CMC for its color and aroma. The quality analysis results for the coconut milk with CMC addition and temperature variations met the existing SNI standards.

Keywords: Stabilizing agent, CMC, Durian rind, Cellulose, Coconut Milk.

INTRODUCTION

Durian (*Durio zibethinus*) is a well-established agricultural crop that has been recognized by communities primarily for its fruit. Literature indicates that durian is one of the tropical fruits indigenous to Indonesia (Rukmana, 1996). In urban settings, the consumption of durian is largely restricted to its flesh and seeds, which are incorporated into various culinary products, including dodol, kolak, jams, and other desserts. Conversely, the durian rind is often discarded, contributing to environmental pollution as it decays, emits foul odors, and attracts pests, ultimately becoming a source of disease (Prabowo, 2009). The edible portion of the fruit is relatively minimal, constituting only 20.52% of its total weight (Setiadi, 1996), while the rind is rich in cellulose, comprising approximately 50-60% of its composition (Hatta, 2007).

This substantial cellulose content presents an opportunity for alternative applications. Given its physical characteristics, the durian rind can be repurposed into various materials such as particle board, cement board, briquettes, activated charcoal, fillers, and even mosquito repellents (Sitepu, 2016). Traditionally, the waste from durian rinds has been utilized primarily for briquette production; however, the rich cellulose content suggests potential for synthesizing Carboxymethyl Cellulose (CMC). CMC is a significant compound due to its solubility in water, making it invaluable across multiple industries, including food, detergents, cosmetics, pharmaceuticals, textiles, paper, and ceramics (Hong, 2013).

The incorporation of CMC is particularly promising in the production of fresh coconut milk. Acting as a stabilizer, CMC helps maintain the uniform dispersion of solid particles within the emulsion, preventing sedimentation. Additionally, it can enhance the flavor, color, and consistency of coconut milk (Kamal, 2010). Coconut milk, an emulsion of oil in water obtained by pressing fresh coconut flesh, faces high spoilage rates due to its substantial water, fat, and protein content, necessitating effective preservation techniques (Sidik et al., 2013). The composition of coconut milk, which includes fat, protein, and water, relies on proteins as natural emulsifiers to keep lipid droplets suspended (Chiewchan et al., 2006 in Raghavendra, 2010).

One of the primary challenges with fresh coconut milk is the rapid separation of its components, occurring within approximately 3-4 hours. This separation results in three distinct layers: an upper cream layer rich in protein, a middle skim layer rich in fat, and a bottom sediment layer. The addition of CMC is crucial for maintaining the emulsion's stability, prolonging the shelf life of fresh coconut milk by increasing its viscosity and preventing separation.

Temperature also plays a vital role in the quality of coconut milk. Extreme temperatures can denature proteins, leading to physical separation of the emulsion. Previous research has shown that the use of various stabilizers can significantly enhance emulsion stability, with some achieving up to 100% stability over extended periods (Sidik, 2013). Studies by Pangastuti (2016) demonstrated that CMC concentrations of 0.5% provided optimal emulsion stability, while combinations with other stabilizers improved viscosity.

Given these insights, this research aims to explore the utility of CMC derived from durian rind waste as a stabilizer or thickener in the production of fresh coconut milk, alongside varying temperature conditions. The objective is to produce high-quality coconut milk characterized by optimal pH, viscosity, moisture, fat, protein, free fatty acids, and organoleptic properties. This study seeks to address consumer preferences, particularly regarding the separation of cream and skim, which diminishes the product's market appeal.

METHOD

The study was conducted from October 2017 to May 2018. The addition of Carboxymethyl cellulose (CMC) as a stabilizer and temperature variation in coconut milk was carried out in the Biochemistry / Food Chemistry Laboratory, Polymer Chemistry Laboratory, FMIPA, University of North Sumatra. And the analysis of CMC functional groups was carried out in the Organic Chemistry Laboratory, FMIPA Gadjah Mada university.

RESULTS AND DISCUSSION

Results of Isolation of α-Cellulose from Durian Rind Waste

In the production of α-cellulose from durian rind waste, the process begins with sample preparation. The durian rind is first separated from its hard, thorny outer layer to obtain the inner white portion. This white part is thoroughly washed under running water to remove any dirt or impurities, then cut into small pieces and sun-dried until it turns brownish. Once dried, the material is crushed using a mortar and pestle, further ground using a blender, and then sieved to ensure a uniform particle size, which facilitates the subsequent isolation process.

The first step in isolating α-cellulose is delignification, which involves the removal of lignin from the durian rind powder using 3.5% HNO_3 in combination with $NaNO_2$. This process yields a by-product known as nitrolignin. Following delignification, a swelling process is conducted using a solution of 2% NaOH and 2% Na_2SO_3. This step causes the cellulose fibers to swell and opens the pores of the cellulose structure, allowing residual impurities to be removed. After swelling, the cellulose typically appears yellowish-brown in color. To achieve the desired white cellulose, a bleaching process is carried out using 17.5% NaOCl. These procedures are based on the method developed by Ohwoavworhua (2005) for the isolation of α-cellulose.

The cellulose was then separated according to its solubility in 17.5% NaOH, where α cellulose was insoluble in 17.5% NaOH while β-cellulose and γ-cellulose were soluble in 17.5% NaOH. The yellow α-cellulose precipitate was bleached again using H_2O_2 so that the α-cellulose turned white and dried in an oven at 60°C for the dehydration process of water still present in the α-cellulose.

Results of CMC Production from α-Cellulose

In the manufacture of CMC, the first alkalization process of α-cellulose was carried out using Isopropanol and 25% NaOH with stirring for 1 hour at room temperature. This process was carried out to make α-cellulose change into alkali cellulose in an alkaline atmosphere created by the solvent Isopropanol and the contribution of alkali Na^+ groups from the 25% NaOH used. The alkalization reaction of α-cellulose can be seen in Figure.

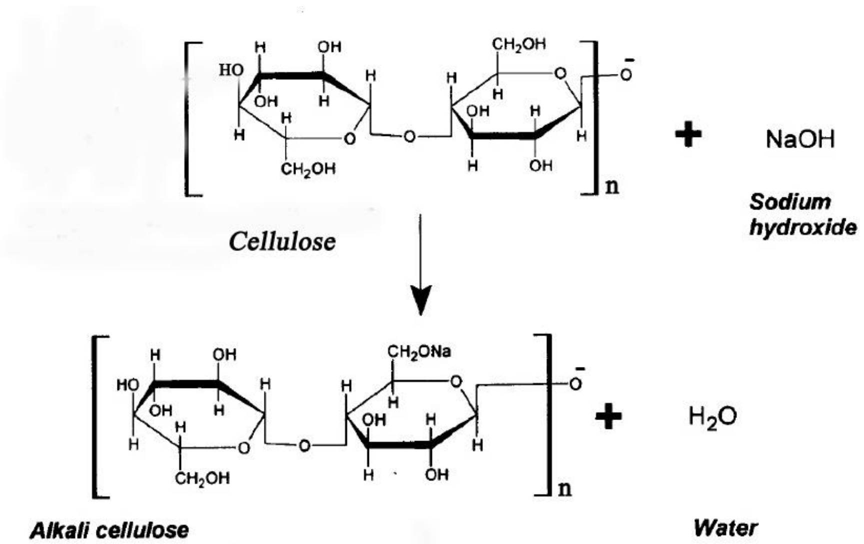


Image of α-Cellulose Alkalization Reaction Using Isopropanol and NaOH (Latif, A et al. 2007)

The next stage is the carboxymethylation (etherification) process using Monochloro Acetate with stirring for 1 hour at room temperature. In this process, there is a substitution of groups between the Na^+ group in alkali cellulose with the $CH2COONa^+$ group in Monochloro Acetate. The carboxymethylation (etherification) reaction of alkali cellulose into carboxymethyl cellulose can be seen in Figure https://gr

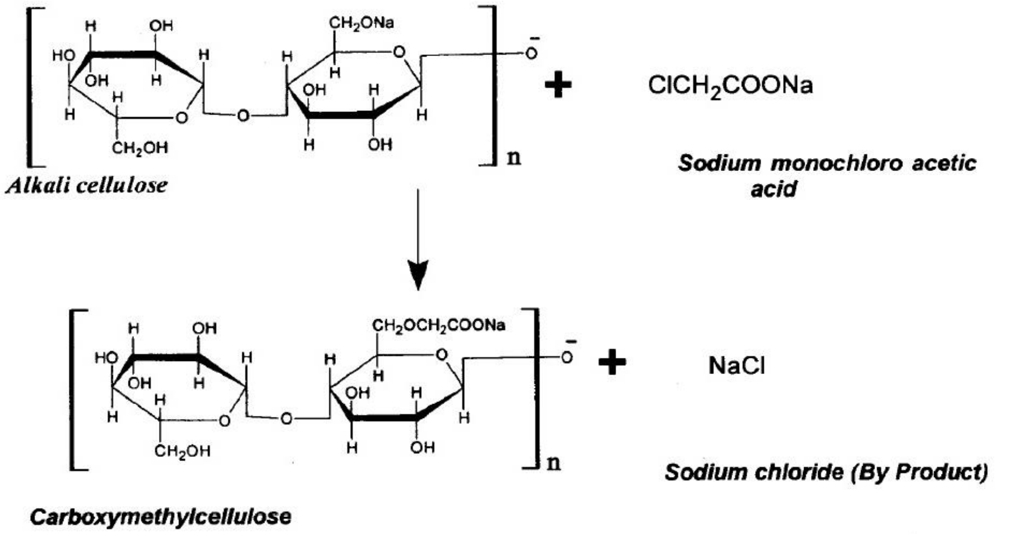


Image of Alkali Cellulose Carboxymethylation (Etherification) Reaction to Carboxymethyl Cellulose (Latif, A et al. 2007)

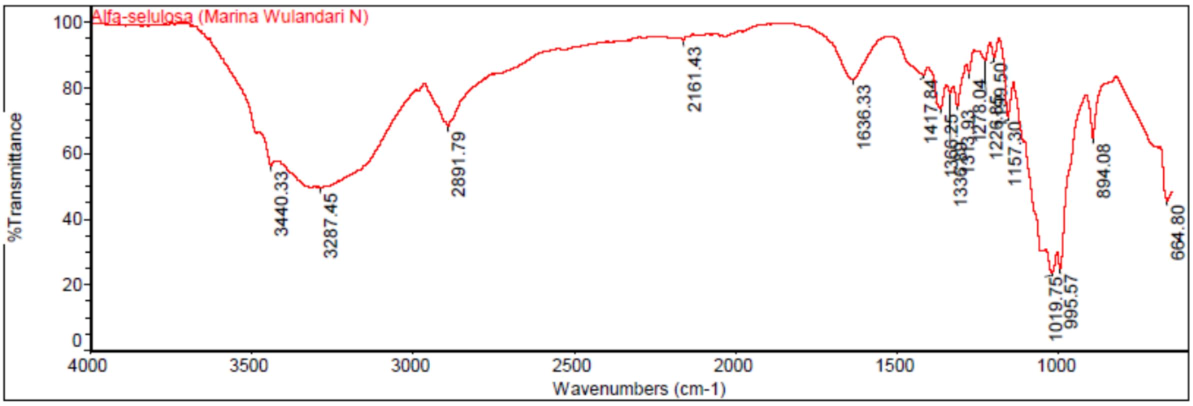
The presence of OH^- groups and various additions, starting from isopropanol and NaOH, causes the CMC mixture to become more alkaline so that neutralization is needed using 90% CH_3COOH until pH = 6 - 8, then filtering is carried out to separate the precipitate formed with the filtrate. The precipitate is then washed with ethanol to remove unwanted by-products in the form of NaCl which is still attached to the CMC so that the resulting CMC is purer (Latif, A et al. 2007).

Furthermore, a centrifugation process is carried out for 1 minute at a speed of 400 rpm to neutralize and remove the remaining acid in the previous process and acetone is added to evaporate the remaining water attached to the CMC then dried in an oven at a temperature of 60^oC (Hong, 2013).

Results of Functional Group Analysis Using FTIR Spectroscopy

In the analysis of functional groups using FTIR for both durian rind α-cellulose spectra and commercial α-cellulose showed that there was no striking difference between the durian rind α-cellulose bands and commercial α-cellulose. This is because both come from cellulose. From the FTIR spectra there is a broad band in the absorption region of 3410.15 and 3440.19 cm-1 which indicates the presence of O-H stretching vibrations from alcohol in the cellulose molecule, followed by C-H stretching vibrations from the alkane chain in the absorption region of 2900.94 and 2891.95 cm-1 (Khalil et al. 2001). In addition, vibration peaks are also seen in the absorption areas of 1319.31, 1313.99 and 1056.99 and 1019.89 cm-1 which indicate the presence of stretching (C-O) in the cellulose ring (Nacos et al. 2006; Garside et al. 2003). While the C-H swing vibration of cellulose is found in the absorption area of 894 cm-1 which indicates the presence of β-glycoside bonds in the structure (Alemdar et al. 2008). From the results of the FTIR analysis, it can be concluded that durian rind α-cellulose and commercial α-cellulose are indeed cellulose compounds.

In the FTIR spectrum of durian rind carboxymethyl cellulose, and commercial carboxymethyl cellulose, changes in absorption peaks are seen. In the absorption area of 1635.64 and 1585.97 cm-1 indicate the presence of stretching vibrations (C = O) of carbonyl originating from sodium monochloroacetate then supported by absorption peaks at wave numbers 1026.13 and 1053.36 indicating vibrations of the symmetrical (C-O) group and absorption peaks at wave numbers 1334.74 and 1372.93 indicating vibrations of the (C-O) stretching group. The absorption peaks at wave numbers 1427.32 and 1413.10 indicate vibrations of the methylene group (-CH2-) from the addition of sodium monochloroacetate. According to Pescok, Shields and McWilliam (1976), the carboxyl group and its salts show two absorption peaks at wave numbers between 1600 - 1640 cm-1 and 1400 - 1450 cm-1 which indicate the presence of carboxymethyl substituents. This shows the characteristics of carboxymethyl cellulose compounds containing carboxyl groups resulting from substitution between monochloro acetate and cellulose compounds. So based on the results of the FTIR analysis, it can be concluded that durian rind carboxymethyl cellulose and commercial carboxymethyl cellulose are indeed carboxymethyl cellulose compounds.



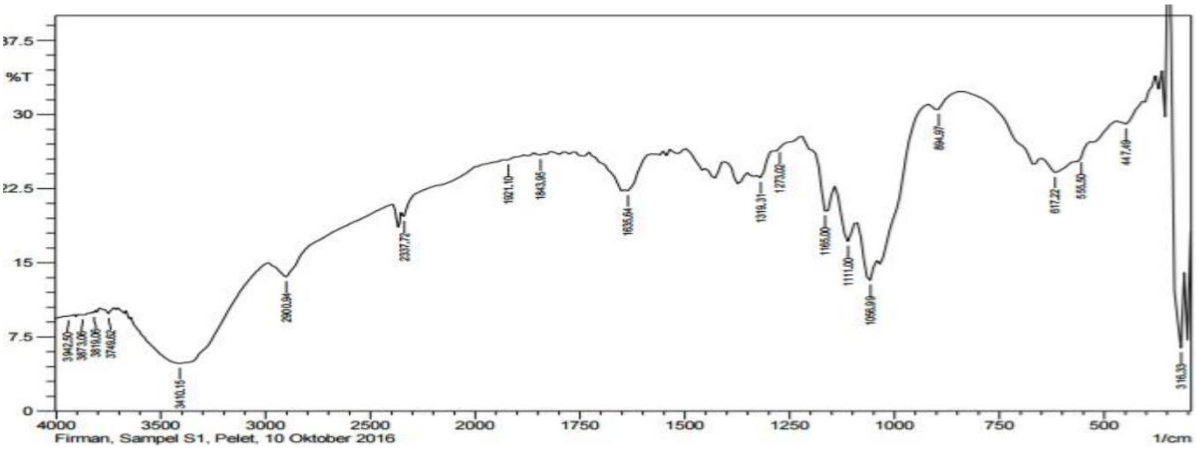


Image of FTIR Spectrum of α-Cellulose Isolated from Durian Rind Compared to Commercial α-Cellulose.

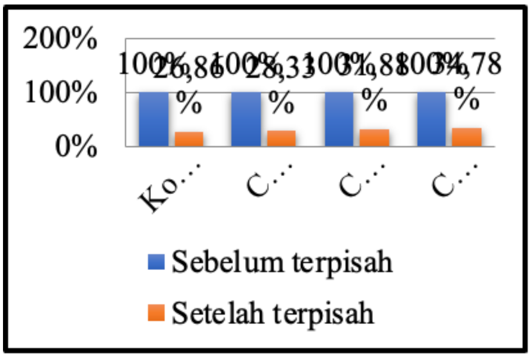
Qualitative Analysis Results of Carboxymethyl Cellulose (CMC)

Carboxymethyl Cellulose (CMC) results obtained were subjected to qualitative analysis aimed at ensuring that the compound made was indeed a positive test for the Carboxymethyl Cellulose (CMC) compound. This is in accordance with the literature from (COEI-1-CMC: 2009) which shows that the addition of other chemical reagents such as acetone, 1.2 N $CuSO_4$ and aquadest + 1 naphthol + $H_2SO_{4(p)}$ will provide reaction results with the desired changes. Heating in this qualitative analysis was carried out at a temperature of 60 - 70 with the aim of completely dissolving CMC into water so that when tested with the existing reagents it immediately provides the appropriate results.

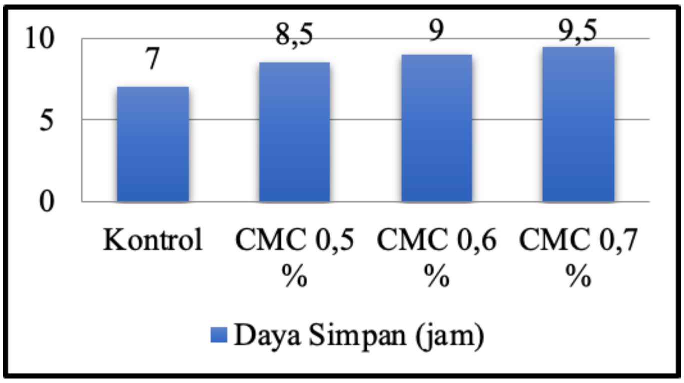
Results of Making Coconut Milk

Coconut milk production with the addition of CMC from α-cellulose from durian rind waste as a stabilizer or emulsifier. Coconut milk begins to separate during storage, making it unstable and not durable. Using CMC as a stabilizer will make coconut milk last longer. The separation of coconut milk into two parts makes the quality of coconut milk less good. According to Fennema (1996), a stabilizer that reduces the separation of coconut milk and as a water binding agent by increasing the hydrophilic properties of the protein. In this study, several research limitations were used, with variations in temperature and concentration. The temperatures used were 16ºC, 25ºC and 80ºC and the concentrations used were 0.5%, 0.6% and 0.7%. According to Iswanto (2009), the best CMC concentration is around 0.6%. Excessive use of stabilizers can cause effects on the texture and appearance of coconut milk that is rough and lumpy. A temperature of 80 ºC is the temperature at which protein will denature.

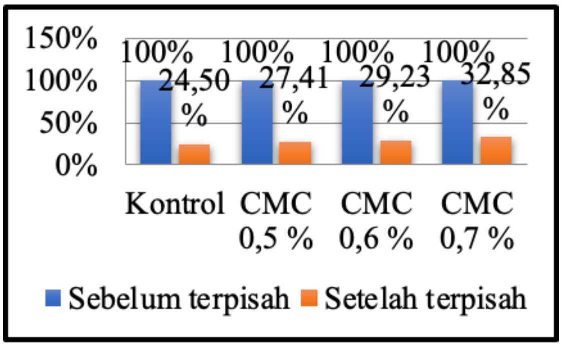
Stability Test



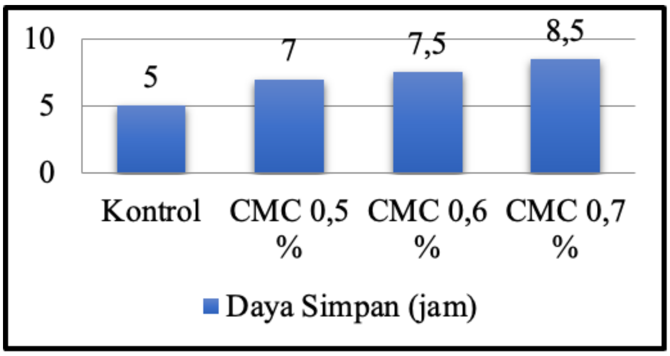
Graphic image of the effect of adding CMC at a temperature of 16 ºC on the % stability



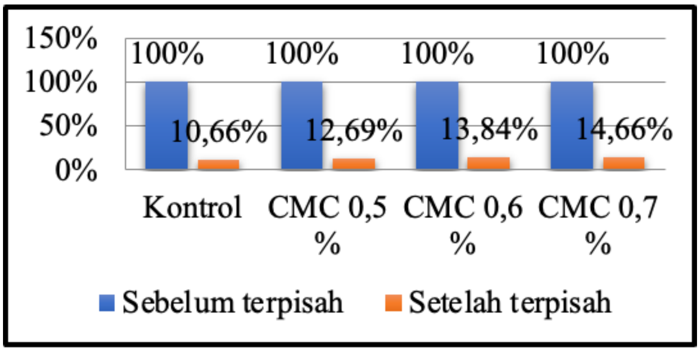
Graphic image of the effect of adding CMC at a temperature of 16 ºC on shelf life



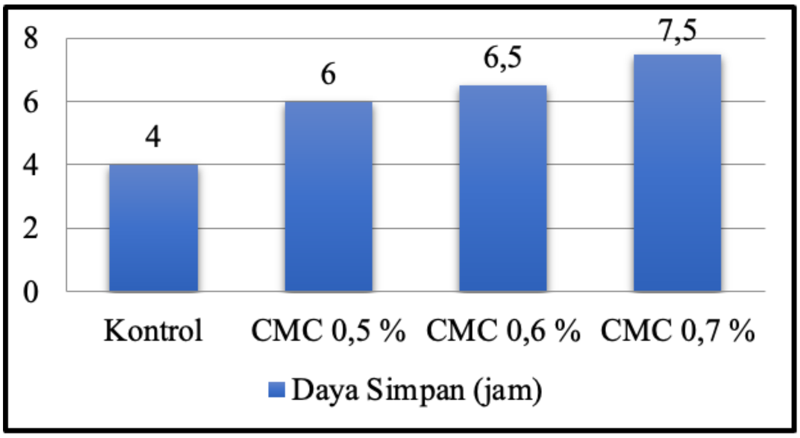
Graphic image of the effect of adding CMC at a temperature of 25ºC on the % stability



Graphic image of the effect of adding CMC at a temperature of 25ºC on shelf life



Graphic image of the effect of adding CMC at a temperature of 80ºC on the % stability



Graphical Image Effect of adding CMC at a temperature of 80 ºC on shelf life

From the graph above, it can be concluded that temperature and variations in CMC concentration greatly affect the shelf life of coconut milk. With a temperature of 25ºC and variations in the concentration of CMC added, it can be seen that the shelf life increases by several hours. This is because CMC works as a stabilizer that binds water by increasing the hydrophilic properties of the protein. At a temperature of 80 ºC, the shelf life and stability of coconut milk are low because high temperatures will reduce the stability of the coconut milk emulsion. At a temperature of 16 ºC, the longest shelf life is obtained because cold temperatures will slow down the activity of microorganisms that grow in coconut milk to damage the quality of coconut milk.

CONCLUSION

1. Carboxymethyl cellulose (CMC) from durian rind cellulose affects the quality of coconut milk, which can be seen from the difference in quality tests for each addition of CMC concentration and temperature variations in making the coconut milk. The results of the study obtained the best results in the addition of 0.7% CMC concentration with a temperature treatment of 16 ºC with quality test results of % stability of 34.78%, pH value of 6.21, viscosity of 12.6138 x 102 cP, water content of 45.81%, protein content of 7.57%, fat content of 43.51%, free fatty acid content of 2.103%.
2. The temperature variation carried out affects the quality of coconut milk, which can be seen from the results of the quality tests carried out.
3. Carboxymethyl cellulose (CMC) from α-cellulose durian rind can be used as a stabilizer because it can improve the quality of coconut milk.

Acknowledgment

It is expected that from the results of the research that has been conducted, the researcher suggests that further researchers test the total mold and bacteria in coconut milk and use temperature variations below 0 ºC.

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