Candlenut oil encapsulation with Hidroxypropyl Methylcellulose (HPMC) for body lotion application

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Abstract. Candlenut oil obtained by cold press extraction method was encapsulated in order to preserve the unsaturated fatty acids content from thermal oxidation. The encapsulation of candlenut oil was performed using hydroxypropyl methylcellulose (HPMC) and maltodextrin as the encapsulating agent due to their filming ability. The solution of wall materials was added with the candlenut oil and emulsified, followed by spray drying process. Three different ratios of oil to wall materials (2:3, 1:1, and 3:2) were evaluated. Based on the peroxide values, encapsulation efficiency, wettability, and moisture content of the microcapsules, the sample with a ratio of 1:1 had better preservation ability towards candlenut oil and therefore used as the main ingredient for body lotion development. Three different concentrations of encapsulated candlenut oil (10%, 6%, and 2%) within the lotion base were observed and analyzed in terms of antioxidant activity by DPPH assay, and overall physical likeness. The result suggested that the wall materials might hinder the reaction between candlenut oil and DPPH radical compounds. The body lotion sample with the lowest content of encapsulated candlenut oil showed the highest antioxidant activity (IC_{50} = 52.31 ppm). Furthermore, according to the ANOVA test result, there was no significant difference in overall physical likeness between the three samples (p-value ≥ 0.05).

1. Introduction
Vegetable oils are liquid vegetable fats that are frequently extracted from several diverse parts of vegetation, such as plant seedlings, fruits, or seeds, which contain high concentration of saturated and unsaturated fatty acids, especially triglycerides, that have beneficial influence especially on the skin. Due to this beneficial influence, unsaturated fatty acids are being one of the most important components in dermatology and cosmetology, and becoming frequently used in formulations of dermatological and cosmetic products [1].

As unsaturated fatty acids have numerous benefits, the demand for unsaturated fatty acids have been increasing. However, unsaturated fatty acids can only be found in certain fish and fish oils, and vegetable oils such as olive oil, which are relatively expensive in Indonesia as they are usually imported from another countries. Therefore, other sources of unsaturated fatty acids are needed in order to offer other lower cost alternatives and sustainable resources of natural unsaturated fatty acids, such as candlenut oil.

In Indonesia, candlenut is mainly used as spice to elevate the taste of food. Therefore, the economic value of candlenut is low due to its usage mainly as a cooking ingredient. However, based on the previous study regarding the extraction of candlenut oil done by Cendikiawan (2016), it has been proven that candlenut oil contains high concentration of unsaturated fatty acids which consist of 20.39%
Omega-3; 38.10% Omega-6 and 26.67% Omega-9 [2]. With this high concentration of unsaturated fatty acids, candlenut oil can be applied as a natural source of antioxidant and a main ingredient in various food and beverages, cosmetics, and health care products, thus increasing the economic value of candlenut in Indonesia.

Although candlenut oil is a good source of fatty acids, the thermal sensitivity of candlenut oil degrades the quality of fatty acids that it contains and therefore reduces its concentration [3]. According to Tjhin (2014), the concentration of Omega-3 significantly decreases at the temperature of 220°C with a mixing time of more than 3 hours while it is given various concentration of oxygen sparged. Due to this characteristic, it is going to be difficult to apply candlenut oil as one of the ingredients during production process as most of the processes might involve heat. Based on the study of formulating Omega-3-rich baby rusk from candlenut oil done by Apelia (2013), the concentration of Omega-3 after the baking process reduces significantly compared to the concentration before the baking process (from 2119 mg/100g to 18.15 mg/100g Omega-3) [4]. Another study done by Lukmanto (2013), shows that the concentration of unsaturated fatty acids decreases up to more than 90% during the production of ice cream inserted with candlenut oil due to the heat introduction during the process [5]. Thus, in order to overcome this problem, encapsulation could be the alternative solution for the thermal instability of candlenut oil. Hydroxypropyl methylcellulose (HPMC) is chosen as the encapsulation material for candlenut oil, in regard to its nature as essential oil and moisturizer, as they have the ability to encapsulate fatty acids. HPMC has good water-solubility, emulsifying-ability, and film-forming-ability, which is already used as microencapsulating agent [6]. Thus, HPMC might be compatible as coating material of candlenut oil.

Therefore, candlenut oil which contains a high concentration of unsaturated fatty acids, are going to be studied as the main ingredient for body lotion as one of the topical cosmetics in order to fulfil the increasing demand of natural fatty acids for cosmetics, to increase the economic value of candlenut in Indonesia, and possibly offer lower costs and provide a sustainable source of natural unsaturated fatty acids. This research aimed to obtain the best ratio between HPMC and candlenut oil in application to topical lotion. Several parameters, such as peroxide values, encapsulation efficiency, wettability, and moisture content of the microcapsules were evaluated to obtain the best ratio of encapsulation agents to candlenut oil. The best ratio of encapsulation agents to candlenut oil was used in the formulation of candlenut oil in the body lotion. The antioxidant activity and overall physical likeness were measured to obtain the best formulation of candlenut oil as a natural fatty acids ingredient in body lotion.

2. Materials and methods

2.1. Candlenut Oil Extraction & Unsaturated Fatty Acids Analysis
Candlenut oil was extracted from candlenut kernels using a cold press expeller under low temperature (≤ 15°C) to preserve the unsaturated fatty acids content. Then, the extracted candlenut oil was filtered using a filter cloth to remove any solid residual remain after the extraction. In order to obtain high purity of candlenut oil, the remaining solid residual that could not be filtered with filter cloth was settled by centrifugation and separated from the candlenut oil. The unsaturated fatty acids (UFAs) content was analyzed using gas chromatography (GC).

2.2. Encapsulation of Candlenut Oil
The candlenut oil was encapsulated with HPMC in which the ratio between HPMC and candlenut oil will be varied into three different concentrations as shown in Table 1. The sample was pulverized using spray dryer with inlet temperature 200°C and outlet temperature 80°C. All of the samples were then analyzed by measuring the percentage of encapsulation efficiency (%EE), thermal oxidative stability (PV), moisture content, and wettability of the samples.
Table 1. Concentration of HPMC, maltodextrin, and candlenut oil for encapsulation process.

| Sample | Candlenut Oil (mL) | HPMC (g) | Maltodextrin (g) | Soy Lecithin (g) | Distilled Water (mL) | Temperature (°C) |
|--------|--------------------|----------|------------------|-----------------|---------------------|------------------|
| A      | 30                 | 30       | 15               | 0.75            | 1000                | 5                |
| B      | 30                 | 15       | 15               | 0.75            | 1000                | 5                |
| C      | 45                 | 15       | 15               | 1.12            | 1000                | 5                |

2.3. Body Lotion Application
The best candlenut oil sample was used in the formulation of the body lotion. The formulation of the product was varied into three different concentrations as shown in Table 2.

Table 2. Concentration of encapsulated candlenut oil in body lotion.

| Sample | Encapsulated Candlenut Oil (%) | Lotion Base (g) |
|--------|--------------------------------|-----------------|
| A      | 10                             | 80              |
| B      | 6                              | 80              |
| C      | 2                              | 80              |

The antioxidant activity of each sample was measured in order to determine which sample has higher antioxidant activity. Sensory analysis was also conducted in order to ascertain the overall physical likeness of the products.

3. Result & Discussion

3.1. Candlenut Oil Extraction
The candlenut oil was extracted using cold press expeller machine to prevent any reduction of unsaturated fatty acids content within the candlenut kernels. The yields of extracted candlenut oil are shown in Table 3.

Table 3. Yield of extracted candlenut oil.

|                      | This research | Isnaeni (2017) | Cendikiawan (2016) |
|----------------------|---------------|----------------|--------------------|
| Weight of Candlenut Kernels (g) | 20,000        | 10,000         | 10,000             |
| Crude Candlenut Oil (mL)    | 6,500         | 2,250          | 3,928              |
| Refined Candlenut Oil (mL)  | 2,980         | 1,230          | 1,467              |
| Yield (mL oil / g kernels) | 0.149         | 0.123          | 0.150              |

The yield of extracted candlenut oil obtained in this research was comparably similar with the yield obtained by Cendikiawan (2016) [2], nevertheless the present yield was slightly higher compared with the yield obtained by Isnaeni (2017) [7]. The differences of yield obtained from the present and previous researches might be due to the freshness of candlenut kernels, leakage during the extraction process, and/or mishandling during the process that might decrease the amount of oil obtained.

3.2. Unsaturated Fatty Acids Content of Candlenut Oil
The unsaturated fatty acids content of candlenut oil was analyzed by using gas chromatography (GC). The result of the analysis is shown in Table 4. The total unsaturated fatty acids content of candlenut oil was 90.80% in which 25.58% Omega-3, 40.86% Omega-6, and 23.91% Omega-9 were contained. In comparison with the unsaturated fatty acids content from the previous researches, the Omega-3 content obtained from this research was higher. Additionally, the Omega-6 content was significantly higher compared with Isnaeni (2017) [7], though it was comparably similar to Cendikiawan (2016) [2].
Nevertheless, the content of Omega-9 content obtained in this research was the lowest compared to the previous researches. The differences in the omega content might be due to the difference in origin where the candlenuts were produced and the freshness of the candlenuts.

Table 4. Omega content of candlenut oil.

| Parameter | Unit       | This research | Isnaeni (2017) | Cendikiawan (2016) |
|-----------|------------|---------------|----------------|-------------------|
| Omega-3   | mg/100g    | 25,583.5      | 21,742.1       | 20,390.00         |
| Omega-6   | mg/100g    | 40,864.6      | 26,009.5       | 38,100.00         |
| Omega-9   | mg/100g    | 23,906.8      | 42,767.5       | 26,670.00         |

3.3. Encapsulation of Candlenut Oil

The candlenut oil was encapsulated using HPMC and maltodextrin as the wall materials. HPMC is modified cellulose that has the ability to emulsify and formed a layer which has already been utilized as a coating material for drugs [6,8]. Three different formulations of candlenut oil encapsulation were observed in this research to ascertain which formulation had better encapsulation ability towards candlenut oil. The formulation differed in the ratio of candlenut oil to wall materials in which 2:3 for sample A, 1:1 for sample B, and 3:2 for sample C.

3.4. Encapsulated Candlenut Oil Samples Analysis

Peroxide Value. Lipid oxidation occurs when unsaturated fatty acids react with oxygen which resulting in fatty acid hydroperoxides as the primary product [9]. The concentration of hydroperoxides from the primary oxidation of oil can be measured by Peroxide Value (PV) calculation. Therefore, PV was calculated in order to ascertain the encapsulation ability of HPMC to prevent the candlenut oil from rapid oxidation when expose to heat.

As shown in Figure 1 below, the peroxide values of all of the samples were fluctuated except for the non-encapsulated candlenut oil. At day zero, all of the samples have zero peroxide values as they have not been exposed to heat. When the oxidation process begin, the peroxide values of all of the samples were slowly increasing over time which occurs due to the formation of hydroperoxides from the oxidation of unsaturated fatty acids [10]. At day four, the peroxide values of all of the encapsulated candlenut oil samples decreased unexpectedly due to the formation of the secondary products of the lipid oxidation. The hydroperoxides formed during lipid oxidation are broken down into a variety of volatile and non-volatile compounds, such as aldehydes and ketones [10,11].

![Figure 1. Peroxide value of samples over time.](image-url)
From the graph, it can be seen that the slope of non-encapsulated candlenut oil sample was higher compared with all of the encapsulated candlenut oil samples, which means that lipid oxidation occurs more rapidly in non-encapsulated candlenut oil sample compared with the encapsulated candlenut oil samples. Thus, it can be concluded that the encapsulation process was able to prevent the oxidation of candlenut oil. Among the three samples, sample B proves to be more proficient in preventing the oxidation of candlenut oil because the hydroperoxides formed during the oxidation process was the lowest.

3.5. Encapsulation Efficiency (%EE)
In this research, the encapsulation efficiency indicates the capability of HPMC to encapsulate candlenut oil. The measurement of encapsulation efficiency was done by extracting the free oil on the surface of the encapsulated samples with organic solvent (hexane). The result is shown in Table 5 below.

| Sample | Encapsulation Efficiency (%) |
|--------|------------------------------|
| A      | 40.05 ± 0.0040              |
| B      | 54.23 ± 0.0057              |
| C      | 34.92 ± 0.0105              |

Comparing the three samples, sample B has the highest encapsulation efficiency of 54.23% compared to sample A and sample C with encapsulation efficiency of 40.05% and 34.92% respectively. The increase of encapsulation efficiency is affected by the size of droplets emulsion, which means the higher the stability of the emulsion will result in higher retention time of the oil [12-14]. This theory can be applied for sample C that contained higher oil concentration than wall materials causing the decrease of emulsion stability and thinning out the wall materials leading to a poor retention of oil and rapid lipid oxidation [6,15].

3.6. Wettability
Wettability is a measure of the ability of microcapsules to rehydrate [16]. The wettability of each sample was displayed in Table 6.

| Sample | Time (minutes) |
|--------|----------------|
| A      | 18.19 ± 2.9769 |
| B      | 4.32 ± 1.2940  |
| C      | 2.02 ± 0.7212  |

Comparing the result of each sample, sample A took the longest time to become entirely wet, which was 18.19 minutes while sample B and sample C took 4.32 minutes and 2.02 minutes respectively. This result is similar with Karim et al. (2016) [17] which indicates that lesser polymer content (HPMC) will result in shorter wetting time of the sample as it will facilitate the accessibility and penetration of water into the powder samples. Furthermore, the short amount of time that sample B and sample C took might also be due to the agglomeration of the powders which could increase the penetration of liquid through the pores of the powder particles [17].

3.7. Moisture Content
Moisture content of the samples was measured in order to determine the physical properties of the powders. Moisture content of the samples may ascertain the quality of the samples.
Table 7. Moisture content of samples

| Sample | Moisture Content (%) |
|--------|----------------------|
| A      | 8.32 ± 0.2656        |
| B      | 6.61 ± 0.1028        |
| C      | 5.67 ± 0.3846        |

It can be seen from Table 7 that sample A has the highest value of moisture content compared to sample B and C. Moisture content of the powders is affected by the viscosity of the emulsion which is highly depended on total solids within the emulsion [18]. Higher total solids within the emulsion will produce higher viscosity emulsion. Higher moisture content of powders will also affect the quality of the sample as moisture might enhance lipid oxidation of the oil resulting in the formation of off-flavors molecules and degradation of samples [17].

3.8. Development of Body Lotion with Encapsulated candlenut Oil

Based on the results of encapsulated candlenut oil analyses, sample B was chosen as the main ingredient for the development of body lotion because sample B had better encapsulation efficiency and low peroxide value.

In this research, three different formulations were applied for the development of body lotion in order to select which sample had better antioxidant activity and overall physical acceptance. The formulation differed in the concentration of encapsulated candlenut oil within the lotion base in which 10% of encapsulated candlenut oil for sample A, 6% for sample B, and 2% for sample C. By considering the final physical texture and fragrance of the products, the respective concentration of encapsulated candlenut oil within the lotion base was chosen.

3.9. Sensory Analysis

The sensory analysis of the products was done in order to ascertain the overall physical likeness of the consumers towards the products. Hedonic test of certain attributes of the products was performed by 30 untrained panelists. The assessed attributes were: texture, spreadability, oiliness, and fragrance. The result of sensory analysis of the sample is shown in Figure 2.

![Figure 2. Quantitative result of sensory analysis.](image)

The result of the sensory test was then analyzed using ANOVA in order to determine whether there was any significant difference between the samples. According to the ANOVA test result, there was no significant difference between each sample (p ≥ 0.05) meaning that despite the difference in encapsulated candlenut oil concentration, the physical attributes of the samples produced similar results.
This result showed similar scores for each sample in physical attributes and overall acceptance. The similar results and insignificant difference between each sample might be due to the use of commercial lotion base in this formulation, as the concentrations of the encapsulated candlenut oil were probably not high enough to alter any physical attributes of the products. Furthermore, as HPMC was used as the wall material for the encapsulation of candlenut oil, the products produced an unfavorable smell in which, despite the difference in concentration of encapsulated candlenut oil, the smell still can be strongly detected from each sample.

3.10. Antioxidant Activity
Antioxidant activity of the body lotion sample was measured using DPPH assay method. Based on the Table 9 below, the IC$_{50}$ of the blank sample (lotion base only) was the highest among the other samples. The high value of IC$_{50}$ of the blank sample might be due to the lack of antioxidant compounds within the lotion base. However, among the samples that contained encapsulated candlenut oil, sample C with the lowest concentration of encapsulated candlenut oil had the lowest IC$_{50}$ value compared to the other samples. According to Rakmai et al. (2017), wall materials might prevent the antioxidant compounds within the core material reacting with the DPPH radical compounds [19]. Therefore, the high IC$_{50}$ value of sample A and sample B might be due to the wall materials blocking the reaction between the antioxidant compounds and DPPH radical compounds. Nevertheless, these results indicated that the addition of encapsulated candlenut oil into the lotion base presumably improved the antioxidant activity of the lotion as the IC$_{50}$ values of each samples were lower than the blank sample.

| Sample | IC$_{50}$ (ppm) |
|--------|----------------|
| A      | 53.70          |
| B      | 64.27          |
| C      | 52.31          |
| Blank  | 91.47          |

4. Conclusion
The encapsulation process of candlenut oil using HPMC and maltodextrin was successful. From the three different concentrations, sample B with 1:1 ratio of wall materials to core material, was proven to be the most effective to preserve the quality of candlenut oil in terms of peroxide value and encapsulation efficiency. The development of the body lotion using encapsulated candlenut oil was varied in three different concentrations which were 10%; 6%; and 2%. The antioxidant activity of the samples was higher than the commercial lotion base. Based on the sensory analysis, none of the samples had better overall physical likeness towards the untrained panelists.

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