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Oxidative Stability of Nano-Encapsulated Candlenut Oil

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Abstract.
Candlenut has a lot of potential pharmacological benefits including in hair care, in treating skin problems such as psoriasis, and in its use as an emollient. However, the high unsaturated fatty acid content of candlenut oil causes it to be easily degraded by oxidation due to exposure to air, light, and humidity. Nanoencapsulations were found to be an alternative in protecting the materials from degradation. The purpose of this study was to investigate the effect of polymer-based nanoencapsulations on the oxidative stability of candlenut oil. Polyvinyl alcohol (PVA) was used as a polymer wall material. A lower peroxide value indicates better oil stability against oxidation. Results showed that nano-encapsulated candlenut oil has a significantly lower average peroxide value of 0.41 meq/100g over 35 days storage time, compared to 3.8 meq/100g in pure candlenut oil. Furthermore, based on its physical characterization, it was found that the reduction of particle size with the optimal size of 172±12 nm was achieved by increasing the amount of PVA. A well-chosen and well-calculated amount of polymer material, therefore, plays an important role to obtain optimal nano-encapsulated candlenut oil.

Keywords: nanoencapsulation, candlenut oil, oxidative stability, polymeric materials

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1. INTRODUCTION

Aleurites moluccana, also known as candlenut, kukui nut (Hawaii), or kemiri (Indonesia), is widely distributed plant and variously used, especially in tropics. In East Africa, candlenut oil is used as an alternative fuel source [1]. In Southeast Asia, candlenut is a natural product that is often used as a spice in cooking and herbal medicine [2]. Empirically, candlenut oil is commonly used as natural hair growth and hair care products [3]. A study has confirmed the antioxidant potential of candlenut oil [2]. Other study also showed the potential of candlenut oil in the treatment of skin problems such as eczema, sunburn from chemotherapy and radiotherapy, and psoriasis [4]. Furthermore, low viscosity of candlenut oil making it possible to be applied as skin moisturizer for sensitive and dry skin [5].

Skin treatment potential of candlenut oil is closely related to its high unsaturated fatty acids content. The unsaturated fatty acid components in candlenut oil are ~ 15% oleic acid, ~ 40% linoleic acid, and ~ 30% linolenic acid [6]. Higher degree of unsaturated fatty acids promotes water permeability which directly causes the transepidermal water loss mechanism [7]. Skin protectant that both preventing drying out and allowing transpidermal water loss is necessary in the healing process of damaged skin [8].

However, high content of unsaturated fatty acids also promotes rapid degradation [9]. Oil degradation due to chemical instability may occur when exposed to oxygen, light, heat, and humidity [10]. Lipid oxidation shortens oil shelf life and creates unfavorable alteration in oil quality [9,11]. Several strategies, both physical and chemical approaches were studied to inhibit lipid oxidation [12]. Nanoparticle-based encapsulation, such as nanospheres and nanocapsules, were found to be well-performed to achieve this objective [13]. In matrix-system nanospheres, active substances may be embedded at the sphere wall of polymer matrix. While in vesicular-system nanocapsules, active substances are the inner core enclosed by polymeric wall materials [14,15].

Various wall materials used in nanoencapsulation of oils showed several interesting properties. A study showed that polysaccharide-protected nano-encapsulated chia seed oil has better oxidative stability compared to the

unencapsulated one during 28 days of storage [11]. Another study found that the use of gum Arabic as wall material significantly increased encapsulation efficiency of fish oil [16]. Utilization of several polymer-based wall materials were also found to be able in increasing oxidative stability [17-19]. Therefore, in this study polymer material was applied in nano-encapsulated candlenut oil formulation. Its oxidative stability was then compared to pure unencapsulated candlenut oil.

2. MATERIALS AND METHODS

2.1. Materials

Materials for candlenut oil nanoencapsulation include pure candlenut oil obtained from Lansida Group, Yogyakarta, ethanol 96%, sorbitan monooleate (Span 80), distilled water, polysorbate 80 (Tween 80), and polyvinyl alcohol (PVA). Materials for the determination of peroxide value include acetic acid, chloroform, potassium iodide, starch, and sodium thiosulfate.

2.2. Methods

2.2.1. Nanoencapsulation of Candlenut Oil

The nanoencapsulation method used in this study was a modification of the method used by Rigo et al. [17]. The formulation consists of an organic phase and a water phase. The organic phase was prepared by mixing pure candlenut oil (5 g), ethanol (40 mL), and sorbitan monooleate (3.75 mL) followed by magnetic stirring at 600 rpm for 20 minutes. The water phase was a mixture of distilled water (49 mL), polysorbate 80 (1.25 mL), and varied polyvinyl alcohol concentration (0.5, 1, 1.5, 2 g), which then heated at 40 °C. The organic phase was slowly poured into the water phase while stirred using a homogenizer at 5000 rpm for 20 minutes. The mixture was then evaporated to reduce the solvent. The formulation is shown in table 1 below.

Table 1: Formulation of nano-encapsulated candlenut oil.

Components	**F1**	**F2**	**F3**	**F4**
Candlenut oil (g)	5	5	5	5
Ethanol 96% (mL)	40	40	40	40
Span 80 (mL)	3.75	3.75	3.75	3.75
Tween 80 (mL)	1.25	1.25	1.25	1.25
PVA (g)	0.5	1	1.5	2
Distilled Water (mL)	49	49	49	49

2.2.2. Physical Characterization of Nano-Encapsulated Candlenut Oil

Average particle size, distribution of particle size (polydispersity index), and zeta potential of nano-encapsulated candlenut oil were measured using dynamic light scattering (DLS) particle size analyzer.

2.2.3. Evaluation of Oxidative Stability

Oxidative stability evaluation aims to determine the resistance of nano-encapsulated candlenut oil and pure candlenut oil against oxidation by measuring their peroxide values. Peroxide value is an oxidation level experienced by a sample. Measurement of peroxide value was carried out based on the method used by Singh et al. [18]. 30 mL of acetic acid and 20 mL of chloroform were added to 5 g of oil. 0.5 mL of saturated potassium iodide solution was then added towards the mixture followed by occasional shaking for 1 minute. After that, 30 mL of distilled water and 0.5 mL of starch solution was added to the mixture. The final mixture was then titrated using 0.01 N sodium thiosulfate. Measurement of peroxide values were carried out both on pure and nano-encapsulated candlenut oil at 5 days interval for 35 days. Peroxide value is given by following equation.

$$\text{peroxide value} = \frac{(S - B) \times N \times 1000}{weight of sample} \tag{1}$$

Where S is the sample titration volume, B is the blank titration volume, and N is the normality of sodium thiosulfate.

3. RESULTS AND DISCUSSION

3.1. Nano-Encapsulated Candlenut Oil

Organoleptic Evaluation of Nano-Encapsulated Candlenut Oil

Organoleptic evaluation includes color, odor, and consistence are shown in table 2 below.

Table 2: Organoleptic evaluation of nano-encapsulated candlenut oil.

Formulation	**Organoleptic Properties**		
	Color	**Odor**	**Consistence**
F1	Milky white	Neutral	Phase Separation
F2	Milky white	Neutral	Few Sedimentation
F3	Milky white	Neutral	Homogeneous
F4	Milky white	Neutral	Homogeneous

Due to biphasic nature of nano-encapsulated oil, a continuous, homogeneous, and stable system were expected. Sufficient amount of each constituent therefore contributes a significant role to satisfy this objective. Phase separation and sedimentation were occurred at 0.5 g and 1 g of PVA, respectively. This showed that lower amount of polymeric wall material was insufficient to perform proper polymerization. Polymerization mechanism played an important role in the process of encapsulation [20].

3.1.1. Physical Characterization of Nano-Encapsulated Candlenut Oil

Physical characteristics includes average particle size, distribution of particle size (polydispersity index), and zeta potential values are shown in table 3 below.

Table 3: Physical characterization of nano-encapsulated candlenut oil.

Sample	Average Particle Size (nm)[a]	Polydispersity Index[a]	Zeta Potential (mV)[a]
F1	1907±60	1.15±0.07	-63.7±6.7
F2	1358±55	0.92±0.04	-78.5±0.7
F3	658±123	0.85±0.04	-57.5±6.5
F4	172±12	0.58±0.05	-72.5±0.2

[a] Average values and standard deviation were resulted from three measurements.

Significantly different average particle size observed by dynamic light scattering particle size analyzer showed that increasing the amount of PVA influenced the reduction of particle size in nano-encapsulated system. This is due to the role of polymeric materials as stabilizing agent in a dispersed system. High polymer concentration may improve kinetic stability by restricting the movement of particles. At sufficient polymer concentration, constituent particles would also be adequately coated. Therefore, irreversible agglomeration due to van der Waals interaction may be restricted [21]. Stable nanocapsule system at higher amount of PVA also promotes more homogeneous particle size which indicated by lower polydispersity index value.

From zeta potential measurement, it is shown that generally all formulations possess similar results. Greater zeta potential values (more than ±61 mV) imply excellent electrokinetic stability of colloidal systems [22]. As shown in table 3, nano-encapsulated candlenut oils have high zeta potential, ranged from -57.5 to -78.5 mV, indicating good stability. In addition, negative zeta potential values from the formulation were possibly due to the presence of negatively charged hydroxyl group from PVA which developed electrostatic repulsion with fatty acids of candlenut oil [23].

3.2. Evaluation of Oxidative Stability

Oxidative stability was evaluated in order to compare the resistance of pure candlenut oil and nano-encapsulated candlenut oil against oxidation during storage time. Pure and nano-encapsulated candlenut oil were stored under controlled temperature and humidity condition. Peroxide value (PV) of all samples were measured at an interval of 5 days for 35 days and results are summarized in Figure 1.

At 5-day storage time, an immediately steep PV increase in pure oil was occurred which amounted to 3.65 meq/100g. While significantly more shallow increases in nano-encapsulated oil were ranged from 0.32 to 0.59 meq/100g. During 35 days evaluation, both pure and nano-encapsulated candlenut oil showed combination of increasing and

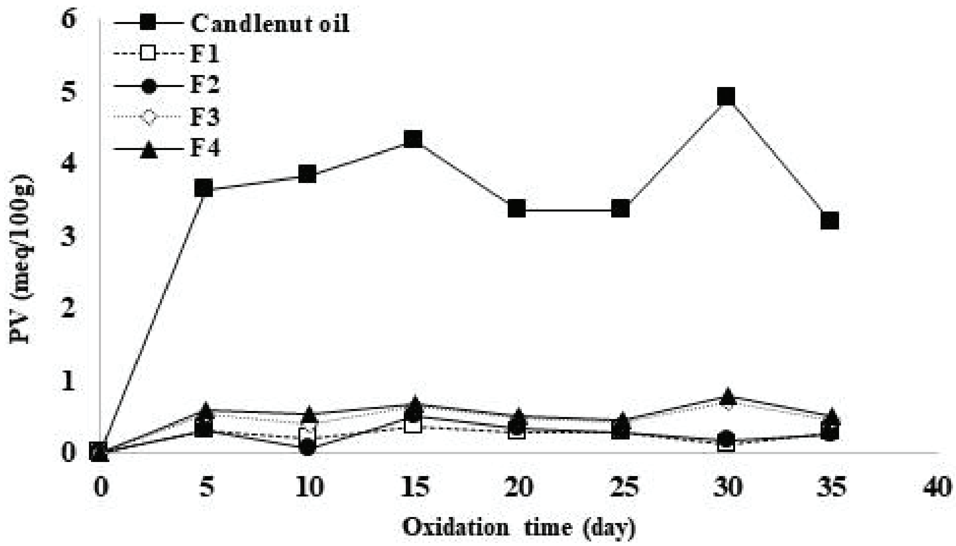

Figure 1: Changes of peroxide value of pure and nano-encapsulated candlenut oil during storage time.

decreasing slopes. This is possibly due to conversion of primary to secondary oxidation products at certain evaluation points [9]. It is also noteworthy that until 25 days of storage, all nano-encapsulated oil formulations followed similar trends in terms of curve gradient, indicating similar oxidation rates. In addition, varying the amount of PVA only slightly affecting the change of peroxide value. Nevertheless, the overall peroxide value of nano- encapsulated candlenut oil was dramatically lower than pure candlenut oil, confirming the ability of polymeric wall materials in protecting oil against oxidation.

4. CONCLUSION

Particle size of nano-encapsulated candlenut oil were reduced when the amount of PVA were increased. It was also observed that nano-encapsulated candlenut oil produced significantly lower peroxide value during storage time compared to pure candlenut oil. This implies that nanoencapsulation treatment is a potential strategy in improving oxidative stability of candlenut oil.

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