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Research article

# Reduction of the acidity and peroxide numbers of tengkawang butter (*Shorea stenoptera*) using thermal and acid activated bentonites

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ABSTRACT

Tengkawang fat (*Shorea stenoptera*), from an indigenous plant of the Kalimantan forest, has excellent potential as an alternative source of vegetable fat because it has a high level of fatty acids composition. Activated natural bentonite can be used as a bleaching agent to improve the quality of tengkawang fat. This research aims to reduce the acidity, peroxide number values and identify the physicochemical properties (fatty acid composition, nutrients, and thermal) of tengkawang butter. Initially, tengkawang samples from Nanga Yen and Sintang were pretreated using the degumming process with 1% phosphoric acid and the neutralization process with a 1 M NaOH 10% w/w solution. The results show that the acidity (mg NaOH/g) of the tengkawang fat samples was reduced from 11.00 to 3.36 when using bentonite activated at 200 °C. The bentonite activated with 0.5 M HCl reduced the acidity to 3.61. The peroxide number (meq $O_2$/kg) of the tengkawang fat samples was reduced from 9.45 to 4.84 and 3.47 by bleaching with thermal-activated and acid-activated bentonites, respectively. Peroxide value correlates with β-carotene content. The smaller of the β-carotene content, the smaller the peroxide value. The acidity, peroxide number, and iodine number values from tengkawang fat after treatment adhere to the SNI 2903: 2016 standard. The main content of fatty acids in tengkawang fat is palmitic acid, stearic acid, and oleic acid. These results show that both products are suitable for the food industry in terms of the acid and peroxide numbers. The application of this research results will assist local people in increasing the economic value of the product from tengkawang plant, which is an indigenous plant from Kalimantan.

## 1. Introduction

Indonesia's fat consumption needs are relatively high, so it must import 20,880,600 kg of solid vegetable fat to fulfil fat needs [1]. One of Indonesia's local plants used as alternative vegetable fat is tengkawang (*Shorea stenoptera*). Several plants in the *Dipterocarpaceae* family, such as *Shorea stenoptera, S. mecisopteryx, S. pinanga*, produce tengkawang fruit [2, 3]. Tengkawang fat, known as illipe butter, has excellent potential as an alternative source of vegetable fat because it has a high level of fatty acid composition and is widely used in the community to meet vegetable fat needs. Various ways of tengkawang fat being used as a material for household consumption. For example, tengkawang fat has been used by communities as a material for lipstick [4], wet noodles [5], and for making black butter rice [6]. Tengkawang butter can be used as a mixing agent for chocolate fat, margarine, and lip paint [7]. The properties of tengkawang fats are similar to cocoa fat; it is classified as a cocoa butter substitute and can be used in the cosmetics and food industries [8].

Tengkawang fat produced by individual community members has a quality below the Indonesian national standard (SNI), meaning that further processing is needed. Traditional tengkawang fat processing uses a device known as an "*apit*" with a production capacity of around 4–5 kg of fat in a single process [9]. Today's industrialized processing of fats and oils frequently includes degumming, neutralization, and bleaching. The degumming process removes impurities, such as phosphatides, from fats [10] and neutralization to reduce excess free fatty acids in oils and fats

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[11]. The bleaching process removes excessive dyes and impurities, using adsorbents such as bentonite [12].

Bentonite is primarily montmorillonite, and has a 2:1 form of mineral; the unit layer structure consists of one $Al^{3+}$ octahedral [13]. Modified reactions change bentonite's surface and structural characteristics by replacing interlayer cations (e.g., $Na^{+}$, $K^{+}$, $Ca^{2+}$) with particular species or sites [14]. Bentonite has reasonably good properties as an adsorbent because it can exchange ions on the surface. The silica layer's charge causes this phenomenon, while the interaction with its filling ions on interlayers such as Al, Ca, and Mg is not so strong that it tends to exchange ions with other cations (protonation) [15]. The cation exchange capacities (CEC) of calcium bentonite range from 40 to 70 mEq/100 g, while those of sodium bentonite is between 80 and 130 mEq/100 g [16]. The bentonite activation process is needed to obtain more optimal results. The activation of bentonite with acids can increase the surface area [17].

Modification of bleaching is needed to achieve the desired butter quality of tengkawang butter [18]. Various ways of modifying the bleaching process activate adsorbents, added chemicals, and heaters [18, 19, 20, 21]. Using adsorbent is the most commonly used because it is relatively useful and low-cost to bleach vegetable oil. One of the adsorbents often used in bleaching vegetable oils is bentonite/bleaching earth [12]. Bentonites/Bleaching Earth are proven to be an adsorbent for bleaching [12, 20, 22].

Research conducted by Hidayat et al on tengkawang fat processing with activated carbon obtained acidity values, peroxide numbers, and iodine numbers of 6.12 mg NaOH/g fat, 5.55 mEq $O_2$/kg fat, and 31.39 g $I_2$/100 g fat, respectively [23]. This research aims to reduce the acidity, peroxide number values and identify the physicochemical properties (fatty acid composition, nutrients, and thermal) of tengkawang butter. In such a way, this research will contribute to the increase of added value to the tengkawang fat products in Kalimantan by meeting the food industry standards.

## Materials and methods

### 2.1. Preparation of materials

Tengkawang fat for this study was obtained from the Nanga Yen and Sintang Villages, Kapuas Hulu Regency, West Kalimantan, Indonesia. Analytical-grade chemicals, such as NaOH, KI, $H_3PO_4$, ICl, and $KIO_3$, were obtained from Merck Millipore (Germany). Bentonite was obtained from Lampung Regency, Lampung Province. The tengkawang fat treatment process in this study involved degumming, neutralization, and bleaching. The bleaching process was carried out using bentonite, which was activated by thermal and acid methods. The pre-treatment process was carried out based on previous research using 20% phosphoric acid for degumming and 1 M NaOH in the neutralization process [23]. However, the bleaching process used thermal and acid activation methods.

### 2.2. Pre-treatment of tengkawang fat

The degumming process was carried out to remove the gum and impurities found in tengkawang fat [23]. Tengkawang fat that had been melted was added to a 20% phosphoric acid solution for 1% (w/w). The mixture was then stirred with a magnetic stirrer for 30 min in a beaker glass. After 30 min, the tengkawang fat was separated from the impurities using warm water at 60–70 °C.

The neutralization process is carried out to remove the remaining free fatty acids in tengkawang fat using alkaline reagents [23]. 1 M NaOH of 10% (w/w) was added to degummed tengkawang fat. The mixture of tengkawang fat and base was stirred for 30 min at temperatures around 60–70 °C. The soap layer was separated from the oil layer using centrifugation. Warm water was used to dissolve the remaining soap contained in the component. Centrifugation was used to separate the oil (above) and the layer of water (below).

### 2.3. Bentonite activation

Two variations of the bentonite activation process were carried out: acid activation and thermal activation. The method used is based on the method used by Christidis et al. [24]. HCl was used as the chemical reactant in the acid activation method. HCl solution with two concentrations (0.5 N and 1 N) was added to the bentonite with bentonite to the HCl ratio of 1:5 (w/w). The mixture was then stirred with a magnetic stirrer for time variation (1 and 3 h) at 70 °C in a beaker glass. After stirring, the bentonite was washed with distilled water to a pH of about 6. The bentonite slurry was then filtered and separated using a vacuum filtration method and a Sartorius Stedim cellulose nitrate membrane with a porosity of 0.8 μm. The bentonite was dried in the oven at 100 °C for 3 h to evaporate the bentonite's water content.

The bentonite thermal activation was carried out at a variation temperature of 200 °C and 300 °C for 60 and 120 min.

The characterization of the bentonite before and after activation was conducted using a scanning electron microscope (SEM), brunauer-Emmett-teller (BET) surface area analysis, X-ray fluorescence (XRF), and X-ray diffraction (XRD).

### 2.4. Bleaching process

The bleaching process was conducted by adding 5% thermal-activated and acid-activated bentonite to the tengkawang butter. The mixture of pre-treatment tengkawang and bentonite was stirred for 30 min at a temperature of 60–70 °C in a beaker glass. After the stirring process was complete, the bentonite was separated from the tengkawang fat using a vacuum filtration method and a Sartorius Stedim cellulose nitrate membrane with a porosity of 0.8 μm.

### 2.5. Chemical analysis of tengkawang butter

The composition of tengkawang fats was characterized using a Gas Chromatography (GC) instrument to determine the fatty acid profile. Samples of up to 1 g of tengkawang fat from Nanga Yen and Sintang were prepared and dissolved in a 100 mL heptane solvent. For each fat sample, 1 ml was injected into the GC column. Before the fat sample was injected, it had to be confirmed that the mobile phase was running well. The mobile phase used in this study was helium gas. After being heated to a temperature of 300 °C, the evaporating component was detected by an FID detector. The detector produced a separation chromatogram of the sample component.

Thermal testing of fat samples was conducted using DSC instruments to determine the thermal properties of the fat. A total of 1 g of sample was put into the DSC container. In this study, samples were heated to a temperature of 70–80 °C with a 1 °C/min heating rate and allowed to stand for 30 min. Data generated from the thermogram were compared to determine changes in the thermal properties.

#### 2.5.1. Analysis of heavy metal contaminant

Analysis of heavy metals contaminants in tengkawang fat was carried out according to the AOCS 999.11 method [25]. Flame Absorbance Spectroscopy (FAAS) instrument was used to test the heavy metal content with a mixture of acetylene gas - air as a burner. Hollow cathode for heavy metals element (Pb, Hg, As, Cd, Sn, Cu). Concentrated HCl and $HNO_3$ for diluting heavy metals in the sample. Heavy Metals (Pb, Hg, As, Cu, Sn, and Cd) Standard solution. The sample was drying for 10–20 min in the oven at 100 °C. After the drying process, the sample was placed in the furnace. The temperature was increased at a maximum of 50 °C/h to 450 °C for 8 h or overnight. After the sample turning into ash, dilute sample residue with 10–30 mL 0.1 M $HNO_3$ and homogenize. The calibration curve was made with a heavy metal standard. The sample was

tested with a specific hollow cathode for each heavy metal elements in the AAS instrument.

### 2.6. Physical characterization of bentonite

The type of XRD (X-Ray Diffraction) instrument used in this study was the PANalytical: X'Pert PRO with Cu K – alpha 1.54 Å as the radiation source. XRD characterization was carried out at angles of 2θ.5°–90°. Characterization was carried out in both the initial bentonite and after activation (acid and thermal).

The type of XRF (X-ray fluorescence) instrument used in this study was the PANalytical: Epsilon1 with Ag. XRF is the source of radiation. XRF was performed to determine the metal and non-metal content in the bentonite samples. The XRD and XRF instruments belong to the Laboratory of Research Department of Physics, Faculty of Mathematics and Natural Sciences, Universitas Indonesia.

An SEM test was performed to determine the bentonite microstructure's characteristics at each stage of the process; the test produced each sample's surface image in three dimensions. The instrument used was an SEM (Scanning Electron Microscopy) JEOL JSM 6510 LA belonging to the Scanning Electron Microscope Laboratory of the Department of Metallurgy and Materials, Faculty of Engineering, Universitas Indonesia.

The Brunauer-Emmett-Teller (BET) test was performed to determine the pores' surface area in the bentonite. This study's type of BET instrument was Quantachrome Quadrasorb-Evo Surface Area & Pore Size Analyzer. The bentonite samples were characterized using a BET analyzer and $N_2$ gas sorption to determine their surface areas. The BET instrument belongs to the Integrated Laboratory and Research Centre, Universitas Indonesia.

### 2.7. Analysis of acidity, peroxide, and iodine value of tengkawang butter

The analysis of acidity, peroxide, and iodine value was using the SNI method: 01-3555-1998 [26]. Analysis of acidity was using the method of base titration with 0.1 M NaOH as the titrant. The calculation of acidity uses the following equation:

$$Acidity = \frac{V \; x \; T \; x \; 40}{m} \quad (1)$$

where:

V = volume of NaOH as the titrant (mL)
T = NaOH Normality
40 = Mass molecule of NaOH (gram/mol)
m = mass of sample (gram)

Iodine number analysis was performed using the titrimetric method with Wijs solution as the reagent, sodium thiosulfate solution as the titrant, and starch as an indicator. The calculation of iodine numbers uses the following equation:

$$Iodine\ Number = \frac{12.69 \; x \; T \; x \; (V_3 - V_4)}{m} \quad (2)$$

where:

T = Normality of sodium thiosulfate solution (0.1 N)
$V_3$ = Volume of sodium thiosulfate solution 0.1 N for blank titration (mL)
$V_4$ = Volume of sodium thiosulfate solution 0.1 N for sample titration (mL)
M = mass of sample (gram)

Peroxide number analysis was performed using the titrimetric sodium thiosulfate solution as titrant and starch as an indicator. The equation for calculating peroxide numbers as follows:

$$Peroxide\ number = \frac{(V_0 - V_1) \; x \; T}{} \; x \; 1000 \quad (3)$$

where:

Vo = Volume of sodium thiosulfate solution 0.1 N for blank titration (mL).
$V_1$ = Volume of sodium thiosulfate solution 0.1 N for sample titration (mL)
T = Normality of sodium thiosulfate solution (0.1 N)
m: the mass of the sample (gram)

### 2.8. Statistical data analysis

Analysis of variance testing (ANOVA) was carried out in this study. If there is a change in the treatment process, further testing is carried out using Duncan's New Multiple Range Test (DMRT) at the 5% level [27].

## 3. Results and discussion

### 3.1. Physicochemical properties of bentonite

XRD (X-Ray Diffraction) characterization to determine the bentonite's crystal composition and structure. Comparing the XRD diffractogram peaks with the database can determine the type of crystal in the material.

In Figure 1(A and B), the bentonite has a montmorillonite crystal structure, shown at the peaks of 2θ 5.0°, 20.0°, and 35.0°. Other minerals, such as quartz and kaolinite, have peaks at 2θ 27.0° and 35.0°. The intensity of montmorillonite in natural bentonite was reduced from 2096 to 1763 in acid activated bentonite. There is a slight decrease in the intensity of montmorillonite peaks after acid activation. This phenomenon

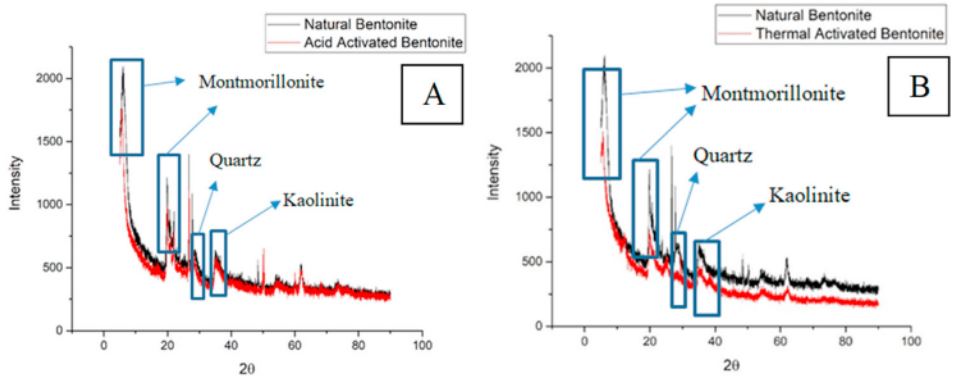



**Figure 1.** XRD graph of natural bentonite with (A) acid activated, and (B) thermal activated bentonite.

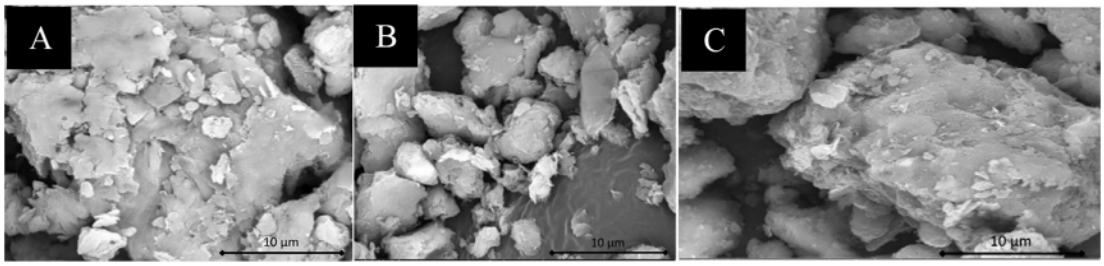



**Figure 2.** Morphology structure of natural bentonite (A), acid-activated bentonite (B), and thermal-activated bentonite (C) magnitude 10.000 x.

as also seen in the study conducted by Temuujin et al. (2004), which used HCl as an acid for activation with reduced montmorillonite peak results [17]. There are still quartz peaks, which indicate that quartz tends to be resistant to acid changes compared to montmorillonite. In a study conducted by Amari et al. (2018), quartz peaks also remained after a leaching process with sulfuric acid [28].

Thermal activation reduces the intensity of montmorillonite crystals in bentonite. The intensity of montmorillonite in natural bentonite reduced from 2096 to 1510 for thermal-activated bentonite. Taher et al. (2018) have reported that thermal activation showed a decrease in montmorillonite crystals' intensity at 2θ 5° in their study [21]. This phenomenon indicates a dehydration process or loss of water molecules between the crystal sheet and distortion of the crystalline structure due to the high temperatures used in thermal activation [29]. From the graph, the activation causes a decrease in quartz crystal peaks and kaolinite. This phenomenon occurs because kaolinite and quartz begin to experience anhydrous decomposition at 200 °C; the heating temperature causes damage in the crystal structure [30].

Figure 2 shows the difference in morphological structure between natural, acid-activated, and thermal-activated bentonite. Acid-activated bentonite has a smooth surface due to acid leaching. Hydrogen ions from acids dissolve impurities on bentonite's surface, increasing the surface area [31]. Morphologically, thermal-activated bentonite has more pores when compared to natural bentonite. This phenomenon is due to the effect of the activation temperature, which evaporates the bentonite's water content so that the pores that were previously still bound to water become open [32]. Rough surfaces show swelling as a result of the interactions between the montmorillonite crystals in bentonite and water. Rough surfaces occur because thermal activation makes bentonite more porous [33, 34]. Previous studies have shown that heating at 250 °C for 90 min tends to make bentonite surfaces rougher [35].

Table 1 shows BET testing on thermal- and acid-activated bentonite to determine changes in the bentonite samples' surface area. The thermal and acid-activated bentonite samples' surface area values were 131.08 and 230.82 $m^2/g$, respectively. Christidis et al. (1997) conducted a study using a 1:15 ratio of bentonite: HCl and variations in the concentration of HCl (0.5–8 N) and contact time (1–6 h) [24]. In conditions of 0.5 N and 1 h, Christidis et al. obtained surface area values of 100 $m^2/g$ of 50 $m^2/g$ and 70 $m^2/g$ of 60 $m^2/g$. Results in this study are consistent with research conducted by Toor and Jin (2012), which found that the activation of bentonite with acid (hydrochloric acid) resulted in a change in surface area from 25.0 $m^2/g$ to 75.5 $m^2/g$ [36]. Viera et al. (2010) reported a calcination process of bentonite at a temperature of 500 °C, increasing the surface area from 78.9 to 89.0 $m^2/g$ [37]. The acid-activated bentonite's surface area in this study is much higher compared to those of natural and thermal-activated bentonite. This phenomenon was because of the reduced impurity of minerals found on the bentonite's surface that increased the surface area and later served as an adsorbent [38].

Table 2 shows the composition of natural bentonite, acid-activated bentonite, and thermal-activated bentonite. In thermal activation, there was a decrease in the silicate levels and the Si/Al ratio. This phenomenon

**Table 1.** Pore size of bentonite after activation.

| Treatment | Before Activation | After Activation | Reference |
|---|---|---|---|
| | Surface Area ($m^2/g$) | Surface Area ($m^2/g$) | |
| Thermal + Acid ($H_2SO4$) Activation | 55.63 | 75.67 | [29] |
| Thermal + Acid (HCl) Activation | 63.00 | 226.00 | [69] |
| Thermal Activation | 78.90 | 89.00 | [37] |
| Thermal Activation | 82.70 | 131.08 | This study |
| Acid Activation ($H_2SO_4$) | 94.36 | 160.70 | [39] |
| Acid Activation (HCl) | 82.70 | 230.82 | This study |
| Acid Activation ($H_2SO_4$) | 92.00 | 294.00 | [70] |

**Table 2.** Comparison of various bentonite activation.

| Treatment | Before Activation | | After Activation | | Reference |
|---|---|---|---|---|---|
| | $SiO_2$ | $Al_2O_3$ | $SiO_2$ | $Al_2O_3$ | |
| $H_2SO_4$ | 49.87 | 14.27 | 66.94 | 7.94 | [39] |
| $H_2SO_4$ | 47.70 | 15.40 | 55.2 | 14.50 | [70] |
| $H_2SO_4$ | 61.68 | 13.91 | 70.69 | 11.98 | [71] |
| HCl | 62.19 | 12.93 | 73.87 | 9.39 | [40] |
| HCl | 56.28 | 13.92 | 60.24 | 14.48 | This study |
| $HNO_3$ | 67.72 | 11.89 | 70.95 | 12.53 | [42] |
| Thermal | 56.28 | 13.92 | 50.07 | 18.91 | This study |

was likely due to damage to the bentonite structure, especially the silicate bonds. The decreasing Si/Al ratio data reinforces data from the XRD, where the montmorillonite crystal peaks were smaller than in the natural bentonite. The activation of bentonite with acids caused an increase in the silicate levels ($SiO_2$) due to the octahedral cations' remobilization presented in the aluminosilicate group [39, 40]. The displaced octahedral cation was composed of silicate structures due to the bentonite composition's high silicon content [41]. Some studies have also proved that acid activation increases silica levels in bentonite and reduce the impurities. Salem et al. (2015) have reported that the process of activating bentonite with acid ($HNO_3$) caused $SiO_2$ and $Al_2O_3$ levels (%) to rise from 67.72 to 70.95 and from 11.89 to 12.53, respectively [42]. Cagla et al. (2013) have reported that the activation of bentonite with $H_2SO_4$ increased $SiO_2$ and $Al_2O_3$ levels (%) from 62.87 to 76.75 and 24.37 to 15.09, respectively [43].

Based on the data in Figure 1(A and B) and Table 2, there is a correlation between changes in crystal intensity with the composition of acid-activated and thermal-activated bentonite. In figure 1(A and B), there is a decrease in the intensity of montmorillonite, quartz and kaolinite crystals. Crystal intensity in thermal activation bentonite reinforcing the data in Table 2 that reduces the Si/Al ratio composition in bentonite. In acid-activated bentonite, the intensity of montmorillonite crystals decreased slightly, but quartz and kaolinite tended to remain. However, because in the acid activation process, there is a leaching process which reduces the impurity cations. There is cation remobilization which increases the Si/Al content ratio in bentonite after the leaching process. Due to the addition of acid in the activation process, there will be $H^+$ cations in the bentonite composition. $H^+$ cations correlate with an increase in acidity in the system, which results in increase acidity of fatty products in the bleaching process.

Based on the data in Table 1 and Figure 2, the thermal and acid activation processes increase the surface area of bentonite. The larger surface area can absorb more impurities in the oil bleaching process. The impurities such as coloured compounds, organic hydroperoxides and FFA can be absorbed into the pores on the surface of the bentonite. The reduced impurity content will improve the quality of the oil. Therefore, the activation process that increases the surface of bentonite can have a good effect on oil processing.

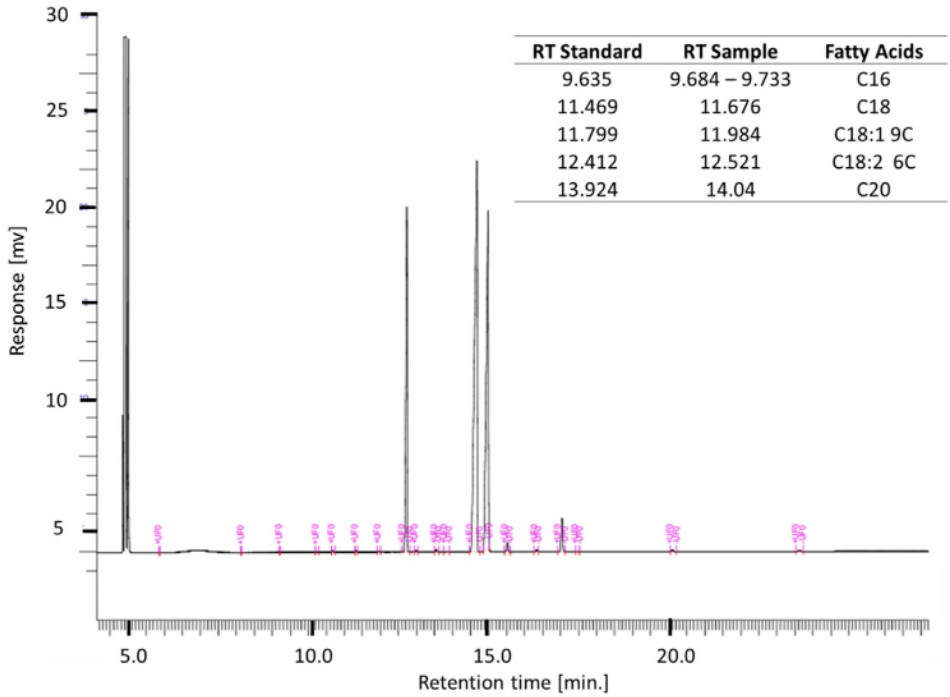
### 3.2. Fatty acid composition of tengkawang butter

Fatty acid profiles of the tengkawang butter samples from Nanga Yen and Sintang were analyzed to determine the fatty acid composition. The fatty acids contained in the tengkawang fat were predominantly palmitic acid, stearic acid, and oleic acid.

Figure 3 is a GC chromatogram for analysis of the fatty acid content of tengkawang fat. Based on the chromatogram, the clear peaks are fatty acids C16 (palmitic acid), C18 (stearic acid), C18: 1 9c (oleic acid), C18: 2 6c (linoleic acid), C20 (arachidonic acid) with RT values of 9.635, 11.469, 11.799, 12.412 and 13.924, respectively. Comparing the retention time (RT) of the sample with the standard retention time (RT) can determine the type of fatty acids [44]. The standard RT can be compared with a specific RT value to determine components in a separated column. Retention time for standard C16, C18, C18:1, C18:2 and C20 are 9.684, 11.676, 11.984, 12.521 and 14.040, respectively.

Table 3 shows the main fatty acid composition (%) of each Nanga Yen and Sintang tengkawang fat sample consisted of stearic acid (44.267 and 42.600); oleic acid (31.894 and 31.290), and palmitic acid (19.710 and 20.220). The differences in the tengkawang growth environment and the seed extraction method influence the various tengkawang fat [45]. For the tengkawang fat from Nanga Yen, the method is the traditional steaming/heating process at high temperatures (100–150 °C). In Sintang, in contrast, the fogging process was used to process the tengkawang fat. Both processes yield different results regarding the quality of the tengkawang fat produced, seen in the differing acidity and peroxide number values. Apart from the growth sites, the different types of tengkawang also have different fatty acid contents. Based on research conducted by Gusti and Zulnely (2015) using *Shorea pinanga* and *Shorea mecisopteryx,* different types of tengkawang have different contents [46]; *Shorea pinanga* contains stearic acid, oleic acid, palmitic acid, and linoleic acid in quantities of 1.56, 42.79, 11.78, and 22.04, respectively, while *Shorea mecisopteryx* contains these acids in quantities of 0.80, 31.28, 14.51, and 27.05, respectively.

In the tengkawang butter samples from Nanga Yen and Sintang, the type of tengkawang used was *Shorea stenoptera*, which has the dominant fatty acid components of stearic acid, oleic acid, and palmitic acid. Significant fatty acid components in *S. pinanga* and *S. mecisopteryx* are oleic




| RT Standard | RT Sample | Fatty Acids |
|---|---|---|
| 9.635 | 9.684 – 9.733 | C16 |
| 11.469 | 11.676 | C18 |
| 11.799 | 11.984 | C18:1 9C |
| 12.412 | 12.521 | C18:2 6C |
| 13.924 | 14.04 | C20 |

**Figure 3.** GC chromatogram analysis of tengkawang butter (Column DB FastFame, FID Detector, 1.0 μL Injection volume).

**Table 3.** Comparison of fatty acid composition in various of fat resources.

| Fat Resource | Fatty Acid Composition | | | | | | | | Ref. |
|---|---|---|---|---|---|---|---|---|---|
| | C12:0 | C14:0 | C16:0 | C18:0 | C18:1 | C18:2 | C18:3 | C20:0 | |
| *S. pinanga* | - | 0.030 | 11.780 | 1.560 | 42.790 | 22.040 | - | - | [46] |
| *S. mecisopteryx* | - | 0.040 | 14.510 | 0.800 | 31.280 | 27.050 | - | - | [46] |
| Cocoa butter | - | - | 25.600 | 36.600 | 32.700 | 2.800 | - | - | [72] |
| CBE | - | - | 27.000 | 33.000 | 35.000 | 3.000 | - | 2.000 | [73] |
| *Shea butter* | 0.090 | 0.020 | 3.560 | 43.500 | 44.470 | 6.110 | 0.150 | 1.440 | [74] |
| Rambutan fat | - | - | 6.100 | 7.100 | 40.300 | - | - | 34.500 | [75] |
| Nanga Yen TB[#] | 0.009[e] | 0.044[e] | 19.710[c] | 44.267[a] | 31.894[b] | 0.526[e] | 0.135[e] | 2.182[d] | This Study |
| Sintang TB[#] | 0.150[e] | 0.080[e] | 20.220[e] | 42.600[a] | 31.290[b] | 1.010[e] | 0.450[e] | 3.530[d] | This Study |

[#] According to DMRT 5%, there is no significant difference in the same letters.

acid and linoleate acid. High oleic acid and linoleate acid contents in fat indicate that the fat is susceptible to oxidative degradation reactions triggered by the influence of high temperatures and light [46].

Tengkawang butter has potential as a cocoa butter equivalent (CBE) because it has a fatty acid composition that treats chocolate fat. CBE are vegetable fats with similar physical and chemical characteristics to cocoa butter (CB) [47]. The primary fatty acids in CBEs are palmitic acid, stearic acid, and oleic acid, similar to those found in CB [48]. Gunstone (2011) have reported that the percentage of palmitic, stearic, and oleic acid in illipe butter makes it a useful component of CBE blends and allows it to be used directly without further processing [49].

### 3.3. Health issue of tengkawang butter

Research on health issues from tengkawang fat is still limited. That is because there is not much research data on nutrient content contained in tengkawang. In this study, we analyzed the nutrient content and metal contamination of tengkawang butter.

Table 4 is the nutrient content found in tengkawang fat. The total fat content of tengkawang is very high, which is above 99%. Tengkawang fat has more calories than chocolate fat and shea fat. Consumption of foods with high calories can increase body weight and body fat mass [50]. Therefore, the intake of tengkawang, chocolate, and shea fats should be controlled not to cause obesity.

The vitamin composition of tengkawang fat is also deficient and insignificant. This condition is probably due to the processing at high temperatures and oxidation, which destroys the tengkawang fat vitamins. Vitamins are easily damaged when exposed to hot temperatures and interact with oxidizing agents such as oxygen in the atmosphere [51]. When using tengkawang fat as a food ingredient, it is necessary to add vitamins to increase vitamin levels.

The dominant fatty acid content in tengkawang fat is saturated fatty acids (SFA) such as palmitic acid and stearic acid and mono-unsaturated fatty acids (MUFA) such as oleic acid. The results of the analysis did not show any trans fatty acids in tengkawang fat. Trans fat increases low-density lipoproteins (LDL), triglycerides, and insulin levels and reduces beneficial high-density lipoproteins (HDL) [52]. High serum LDL levels, such as cholesterol, will increase the risk of atherosclerosis or coronary heart disease. Although it does not contain trans fat, tengkawang fat is high in SFA. Consumption of saturated fatty acids too high will also increase the LDL content in the body. Joint FAO/WHO recommended total intake of SFA less than 10% of total energy intake [53]. Therefore, consumption of tengkawang fat must be controlled not to increase LDL levels in the body.

**Table 4.** Nutritions of tengkawang butter and several fat sources.

| Parameter | unit | Nanga Yen TB[#] | Sintang TB[#] | Beef [76] | Cocoa Butter | Shea Butter [77] |
|---|---|---|---|---|---|---|
| Carbohydrates | % | 0.01 | 0.01 | - | 12[a] | - |
| Protein | % | 0 | 0 | 8.21 | 1[a] | - |
| Total Fat | % | 99.8 | 99.8 | 70.9 | 60[a] | 100 |
| Energy | cal/100 g | 898 | 898 | 674 | 884[a] | 884 |
| Cholesterol/phytosterol | mg/100 g | 1.38 | 0.49 | 99 | 1[b] | - |
| Vitamin A | IU/100 g | 0.1 | 0.1 | - | 33[a] | 0 |
| Vitamin E | µg/100 g | 0.005 | 0.005 | - | 0.1[a] | 0 |
| Vitamin D | mg/100 g | 0.1 | 0.1 | 11 | - | 0 |
| trans-fat | % | 0 | 0 | - | - | 0 |
| Saturated Fats* | % | 66.327 | 66.698 | 29.5 | 55.2–73.50[b] | 46.6 |
| Mono-unsaturated Fats* | % | 32.199 | 31.804 | 30.9 | 31–35.2[b] | 44 |
| Polyunsaturated Fats* | % | 0.793 | 0.792 | 2.56 | 1.5–4.2[b] | 5.2 |
| Linoleic Acid | % | 0.135 | 0.134 | | | 4.9 |
| Omega-3 | % | 0.266 | 0.265 | | | |
| EPA | % | 0.131 | 0.131 | | | |
| Omega-6 | % | 0.526 | 0.526 | | | |
| Omega-9 | % | 32.093 | 31.695 | | | |

[*] based on the percentage of total fat.
[a] reference from [78].
[b] reference from [79].
[#] According to DMRT 5%, there is no significantly different.

**Table 5.** Heavy metal contaminant of tengkawang butter.

| Metal | unit | Threshold [26] | Nanga Yen TB | Sintang TB |
|---|---|---|---|---|
| Lead (Pb) | mg/kg | max 0.10 | <0.031 | <0.031 |
| Cadmium (Cd) | mg/kg | max 0.10 | <0.004 | <0.004 |
| Mercury (Hg) | mg/kg | max 0.05 | <0.005 | <0.005 |
| Arsenic (As) | mg/kg | max 0.10 | <0.013 | <0.013 |
| Tin (Sn) | mg/kg | max 40.00 | <0.800 | <0.800 |
| Copper (Cu) | mg/kg | max 20.00 | <0.004 | <0.004 |

Table 5 are metal contaminants found in tengkawang fat. Heavy metals can harm human health. Cd is a dangerous element that can damage membranes and DNA and accumulate in the body's metabolism for 16–33 years [54]. Cd's daily intake is 0.008 mg, which is about 10.7% of the Reference Dose (RfD), equivalent to 0.001 mg.kg-1 of body weight per day [55]. Pb toxic effects on several organs, such as the liver, kidney, spleen, and lungs, leading to reduced organ function [54]. Pb's daily intake is estimated to be up to 0.012 mg equivalent to 0.245 mg per day for a 70 kg weight [55]. Arsenic is an element with a high risk of various complications, including dermatological effects, cardiovascular effects, pulmonary disorders, reproductive effects, and neurological effects [25, 56, 57, 58, 59]. Mercury is an environmental pollutant with a high risk to health due to the high toxicity and mobility of mercury in the ecosystem [60, 61]. The Joint FAO (Food and Agriculture Organization of the United National)/WHO Expert Committee on Food Additives (JECFA) in 2004 set a limit for the concentration of mercury at 1.6 mg/kg of body weight [62].

In the contaminant analysis of tengkawang butter, the content of heavy metal contaminants (Cd, Hg, As, Pb, Sn) was below the threshold for heavy metal content in food raw materials. Therefore, tengkawang fat in this study is free from the risk of heavy metal contaminants. Tengkawang Nanga Yen and Sintang were obtained from rural areas far from industry and the risk of pollution.

### 3.4. Thermal analysis of tengkawang butter

Thermal characterization uses DSC to determine the thermal properties of a material. The tengkawang fats from Nanga Yen and Sintang have different characteristics due to their different fatty acid contents.

The DSC of the tengkawang fat samples in Figure 4 shows different peaks for the tengkawang fats from Sintang and Nanga Yen. In the raw tengkawang fat from Nanga Yen, the onset point occurred at 37.7 °C and the offset point at 48.4 °C, whereas in the raw tengkawang fat from Sintang, the onset point was at 39.1 °C, and the offset point was at 48.1 °C. There was also a big difference in heat flow between the tengkawang fat samples from Nanga Yen and Sintang. For the Nanga Yen tengkawang fat sample, the peak of heat flow occurred at -28,921 mW, while for the Sintang tengkawang fat sample, the peak of heat flow occurred at -3,156 mW. The enthalpy value of the raw tengkawang from Nanga Yen was 119.34 J/g, while that of the tengkawang from Sintang was only 98.99 J/g. This phenomenon was probably due to differences in the fat compositions of the tengkawang samples from Nanga Yen and Sintang. The tengkawang sample from Nanga Yen has a larger component of saturated fatty acids, requiring more energy than the samples from Sintang. There are thermogram changes in raw and refined tengkawang fat. Refined tengkawang has a shorter peak than raw tengkawang and confirms the fatty acid profile data for both fats. In refined tengkawang fat, there is a slight increase in unsaturated fatty oleic acid and a decrease in saturated palmitic acid; due to the change in composition, the peak shifts to a lower temperature with a smaller peak height. The energy value needed to melt tengkawang fat before and after purification also changed, from 119 J/g to 91 J/g for the tengkawang from Nanga Yen and 98 J/g to 86 J/g for the tengkawang from Sintang. The enthalpy and temperature values of tengkawang fat are comparable to those of other vegetable fats.

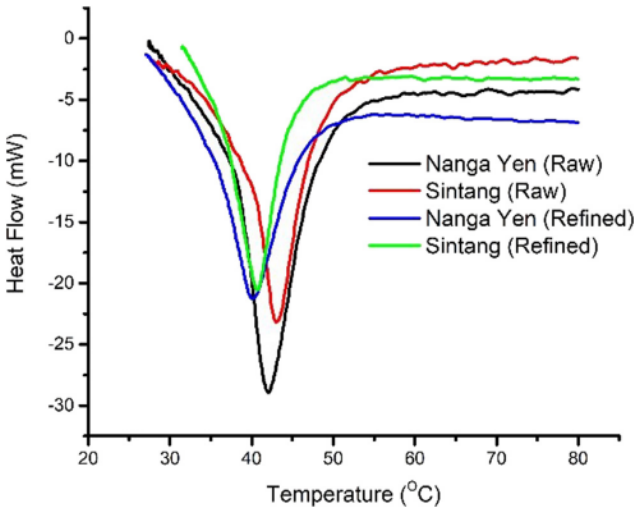



**Figure 4.** DSC thermogram of tengkawang butter Nanga Yen and Sintang raw and refined.

From the results shown in Table 6, the enthalpy value of tengkawang fat (from both Nanga Yen and Sintang) is almost similar to dark chocolate and chocolate fats. This phenomenon is probably due to fatty acids' compositions, which are almost the same for tengkawang butter as for chocolate fat.

### 3.5. Acidity and peroxide number of tengkawang butter

Table 7 uses a complete randomized design of the first-factor factorial group Nanga Yen and Sintang consisting of thermally activated treatments (300 °C - 120 min; 300 °C - 60 min; 200 °C - 120 min; and 200 °C - 60 min). The second factor Nanga Yen and Sintang consisted of acid-activated treatment (HCl 0.5 N (1 h); HCl 0.5 N (3 h); HCl 1 N (1 h); and HCl 1 N (3 h).

Table 7 shows the results from a statistical analysis of the acidity, peroxide, and iodine number of tengkawang fat. Numbers in the same acidity columns in thermal treatment of Nanga Yen dan Sintang tengkawang butter indicate that interaction was not significantly different according to Duncan's Multiple Range Test (DMRT) 5%. Meanwhile, the acid treatment shows different letters, which indicate the interaction significantly different. Numbers in columns of peroxide number in thermal treatment for Nanga Yen tengkawang butter there are significantly different for 300 °C-120 min and 200 °C–60 min, and not significantly different for Sintang tengkawang butter. For iodine columns, there are no significantly different for some data and significantly different for the others.

The quality of tengkawang fat's analysis parameters is acidity, peroxide number, and iodine number. The acidity describes how much free fatty acid is in the fat sample. The acidity significantly influences the acidity of the solution needed to determine the effect of bentonite on the fat sample. The peroxide number is an essential indicator of the rancidity of a sample. The higher the peroxide number's value, the easier it is for fat

**Table 6.** Comparison of enthalpy from various of fat resources.

| Fat Resources | Temperature (°C) | | Enthalpy (J/g) | Reference |
|---|---|---|---|---|
| | Onset | Offset | | |
| CB | 25.97 | 37.70 | 116.20 | [74] |
| Rambutan fat | 34.87 | 38.95 | 113.70 | [80] |
| Dark chocolate | 12.54 | 32.32 | 121.52 | [81] |
| CBE | 26.20 | 34.60 | 30.70 | [81] |
| Shea butter | 7.13 | 17.82 | 58.93 | [82] |
| Nanga Yen TB | 37.70 | 48.40 | 119.34 | This study |
| Sintang TB | 39.10 | 48.40 | 98.99 | This study |

Abbreviation used: CB: cocoa butter; CBE: cocoa butter equivalent; TB: tengkawang butter.

**Table 7.** Acidity, peroxide, and iodin numbers from tengkawang butter.

| Tengkawang | Treatment | Acidity (mg NaOH/g) | Peroxide (meq $O_2$/kg) | Iodine ($I_2$/100 g) |
|---|---|---|---|---|
| Nanga Yen | 300 °C - 120 min | $3.808^{ab}$ | $7.357^{abc}$ | $30.815^{b}$ |
| | 300 °C - 60 min | $3.725^{ab}$ | $5.524^{cd}$ | $31.649^{a}$ |
| | 200 °C - 120 min | $3.545^{bc}$ | $6.089^{bcd}$ | $30.303^{d}$ |
| | 200 °C - 60 min | $3.368^{c}$ | $4.846^{d}$ | $30.756^{bc}$ |
| Sintang | 300 °C - 120 min | $1.846^{d}$ | $8.794^{a}$ | $30.697^{c}$ |
| | 300 °C - 60 min | $1.820^{d}$ | $7.388^{a}$ | $30.070^{e}$ |
| | 200 °C - 120 min | $1.794^{d}$ | $8.230^{abc}$ | $29.466^{f}$ |
| | 200 °C - 60 min | $1.680^{d}$ | $7.262^{abc}$ | $30.012^{e}$ |
| Nanga Yen | HCl 0,5 N (1 h) | $3.614^{d}$ | $4.829^{ab}$ | $30.993^{b}$ |
| | HCl 0,5 N (3 h) | $3.824^{c}$ | $3.470^{c}$ | $31.589^{a}$ |
| | HCl 1 N (1 h) | $4.366^{b}$ | $3.225^{cd}$ | $30.128^{e}$ |
| | HCl 1 N (3 h) | $4.804^{a}$ | $2.450^{d}$ | $30.815^{c}$ |
| Sintang | HCl 0,5 N (1 h) | $2.030^{h}$ | $5.735^{a}$ | $30.697^{d}$ |
| | HCl 0,5 N (3 h) | $2.144^{g}$ | $4.626^{b}$ | $29.954^{f}$ |
| | HCl 1 N (1 h) | $2.284^{f}$ | $4.925^{ab}$ | $29.466^{f}$ |
| | HCl 1 N (3 h) | $2.345^{e}$ | $2.403^{d}$ | $29.954^{f}$ |

According to Duncan's Multiple Range Test (DMRT) 5% the same column's number followed by the same letter is not significantly different.

to oxidize or turn rancid. The purpose of the bleaching process is to reduce the peroxide value to below ten meq $O_2$/kg. The iodine number is a value that shows how many double bonds are in fat.

Table 8 shows the comparison of acidity, peroxide, and iodine number with SNI 2903: 2016 of tengkawang fat as raw material. The acidity value of tengkawang fat after acid-activated bleaching treatment has a higher value than after thermal-activated bleaching. In the activation of bentonite, a hydronium ion ($H^+$) fills the cation vacancy in the activation process and increases the acidity value. The higher the HCl concentration in bentonite activation, the more acidity in the tengkawang sample increased. This phenomenon is because bentonite acidity is higher from thermal bentonite, and it releases more Hydronium ions ($H^+$) and increases in acidity.

The iodine value in the tengkawang sample varied based on the type of bleaching performed. However, the iodine value change still enters the

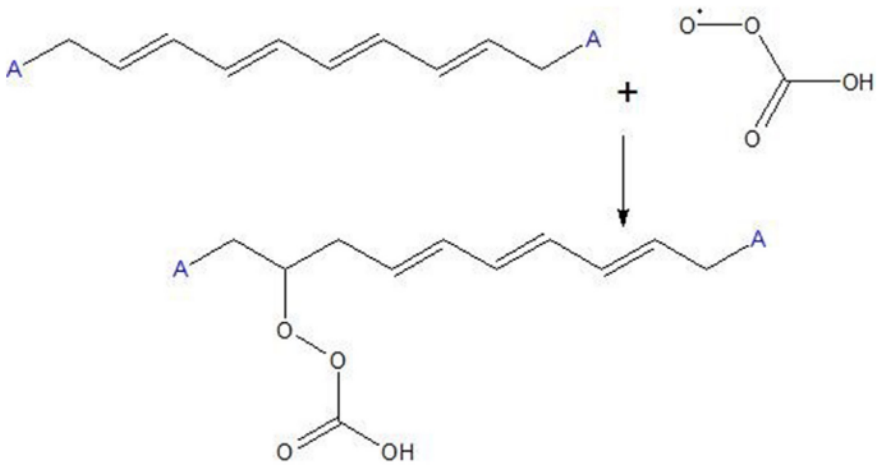



Figure 5. The radical reaction of unsaturated compounds (β-carotene).

**Table 8.** Comparison of acidity, peroxide and iodin number of tengkawang butter with SNI standard.

| Parameter | Unit | Standard Quality (SNI 2903:2016) | Initial Tengkawang Butter | | Thermal-Activated Bleaching | | Acid-Activated Bleaching | |
|---|---|---|---|---|---|---|---|---|
| | | | Nanga Yen TB | Sintang TB | Nanga Yen TB | Sintang TB | Nanga Yen TB | Sintang TB |
| Acidity | mg NaOH/g | Max 3.5 | 11 | 2.54 | 3.36 | 1.68 | 3.61 | 2.03 |
| Peroxide Number | meq $O_2$/kg | Max 10 | 9.45 | 14.03 | 4.84 | 7.26 | 4.82 | 5.735 |
| Iodine Number | g $I_2$/100 g | 25–38 | 31.5 | 32.02 | 30.75 | 30.01 | 30.99 | 30.69 |

threshold of the tengkawang fat standard, so that it is not a problem in this case.

The peroxide value after acid-activated bleaching treatment is smaller than thermal-activated bleaching. Acid-activated bentonite has a higher surface area than thermal-activated bentonite, causing it to absorb impurities. The peroxide value in oil must be below 10, but it will increase due to the thermal decomposition process [63]. The content of compounds with many double bonds, such as β-carotene, influences the number of peroxides. The double bond in β-carotene can undergo an oxidation reaction, which increases radical species in the system [64]. Radical species will add organic peroxide compounds, which increase the value of the peroxide and make fats quickly turn rancid. In previous studies, we have evaluated the use of bentonite to reduce β-carotene levels in tengkawang fat [35]. The β-carotene content was reduced from 125.42 μg/g to 25.51 using a 10% thermally activated bentonite.

Tengkawang butter with bleaching process using 10, 20, and 30% thermal-activated bentonite had levels of β-carotene (μg/g) respectively 25.51, 19.24 and 17.79. The β-carotene content is less with a higher content of bentonite in the bleaching process. Also, the peroxide level decreases with the reduction of β-carotene content. The peroxide value (meq $O_2$/kg) in the bleaching process using 10, 20 and 30% bentonite had values of 4.95, 4.87 and 4.78, respectively. The decrease in the β-carotene content influence the reduction of peroxide value. Condition factors such as temperature, pressure, oxygen concentration, and the reaction medium influence the adsorption of β-carotene in the bleaching process [35].

Figure 5 shows the reaction of peroxide radicals with unsaturated components such as β-carotene and fatty acids. The medium's polarity influences the pathways of oxidation of β-carotene. Only radical addition reaction is formed in non-polar solvent pathways, whereas carotenoid radical cations are formed in polar ones [65, 66]. Under non-polar conditions, the radical decomposition of β-carotene into non-radical products such as epoxides and cyclic ethers [65]. The products of radical decomposition can also produce aldehyde group products such as apocarotenals [67]. Under polar conditions, β-carotene changes to a peroxyl anion ion ($ROO^-$) and a carotenoid cation ($Car^+$) [65]. Although vegetable fats such as tengkawang fat are non-polar, the negative charge on bentonite, which tends to be polar, will support the reaction process for non-polar components (fatty acids and β-carotene) to interact with bentonite [66]. Based on Silva et al. (2013) research, the β-carotene adsorption process using bentonite is a chemisorption process [68].

## 4. Conclusions

The processes of thermal-activated bleaching and acid-activated bleaching can significantly reduce the acidity and peroxide number values for tengkawang fat. The acidity (mg NaOH/g fat) can be reduced from 11.00 to 3.36 by thermal-activated bleaching and 3.61 by acid-activated bleaching. Peroxide numbers (meq $O_2$/kg) can be reduced from 9.45 to 4.84 by thermal-activated bleaching and 3.47 by acid-activated bleaching. Peroxide value correlates with β-carotene content. The smaller the β-carotene content, the smaller the peroxide value. The acidity, peroxide number, and iodine number values from tengkawang fat after treatment adhere to the SNI 2903: 2016 standard. The main content of fatty acids in tengkawang fat is palmitic acid, stearic acid, and oleic acid. The fatty acid content of tengkawang is similar to that of chocolate, meaning that tengkawang fat can become a cocoa butter substitute. Specific surface area values ($m^2$/g) are 82.27 for natural bentonite, 131.08 for thermal-activated bentonite, and 230.82 for acid-activated bentonite. The results obtained from this study indicate that the quality of the tengkawang process results is in accordance with Indonesia national industry standards (SNI) for food. The processing method in this research can be a reference for the tengkawang fat process for producers in Kalimantan from traditional to an industrial method.

## Declarations

### Author contribution statement

Muhammad Arif Darmawan: Conceived and designed the experiments; Performed the experiments; Wrote the paper.

Bagas Zaki Muhammad: Performed the experiments.

Andre Fahriz Perdana Harahap, Muhammad Yusuf Arya Ramadhan & Muhammad Sahlan: Contributed reagents, materials, analysis tools or data.

Haryuni & Teguh Supriyadi: Analyzed and interpreted the data.

Suraini Abd-Aziz: Conceived and designed the experiments.

Misri Gozan: Conceived and designed the experiments; Wrote the paper.

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### Data availability statement

Data included in article/supp. material/referenced in article.

### Declaration of interests statement

The authors declare no conflict of interest.

### Additional information

No additional information is available for this paper.

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