EFFECT OF EXTRACTING METHOD OF COCONUT OILS ON TRIBOLOGICAL PROPERTIES AS BIO-BASED LUBRICANT

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The use of mineral and synthetic lubricants will have long-term impact on the environment. Vegetable oil can be an alternative for substitution due to having significant environmental benefits. In addition, vegetable oils also offer renewable resources and have proven to have excellent lubrication performance for automotive and industrial application. Coconut oil is one of vegetable-based oil that would have prospective characteristics to be exploited as bio-lubricant oil. The origin of coconut oil would distinguish its characteristic, hence differentiating its performance as a lubricant. Indonesia is well known as having abundant source of coconut oil which is made with different extraction methods. Extraction method can be envisaged for improving the performance of coconut oil as lubricant oil. Indonesian coconut oils that had been extracted through dry and wet methods would be a primary concern in this study. The prospective of extracting method of coconut oil as lubricant in term of physicochemical and tribological properties will be investigated. hydrogenated coconut oil (HCO), virgin coconut oil (VCO), and refined coconut oil (RCO) are product of coconut oil extracted from wet and dry, respectively. Results indicated that RCO and HCO posed high viscosity index, high ratio unsaturated to saturated fatty acids, and low wear and friction coefficient which are prospective as base fluid in lubricant industries.

Key words: coconut oils, wear, coefficient of friction, vegetable oil.

INTRODUCTION

The primary functions of lubricating oil are for reducing friction, minimizing wear, distributing heat, and removing contaminants from two moving parts as well as improving efficiency of a mechanical system. Hence, lubricating oil has been extensively utilized in many sectors such as automotive, industries, marine and processes. As such, the demand for lubricant oil would encounter a significant increase. Based on research report released by Transparency Market Research in January 2015, global lubricant oil market was undergoing a significant increase by the end of 2024 [1]. This report divided the global lubricant market into several criteria. One of the criterion was by product that classified lubricant oil into three products such as mineral oil based, synthetic based and bio-based lubricant. Among those products, mineral based lubricant oil took by 80% of market share. This, therefore indicates that mineral based lubricant oil is still dominant in use. Meanwhile, availability of crude oil as the primary source of lubricant based mineral oil is experiencing a decline trend. Moreover, the impact of utilizing lubricant based mineral to environment is also chemically detrimental due to its disposal. Thus, stringent regulation on disposal of waste lubricant oil especially that of derivation from mineral based oil has been introduced by many countries and organizations. Based on environmental facts, public awareness, markets globalization as well as economic incentive attempts on reducing environmental impairment caused by application of lubricated based mineral oil, those must be taken into account [2]. One of the effort that is considered to be a prospective solution is by utilizing bio-based lubricant types [3,4]. Besides chemically safe for human, bio-based lubricant also poses high biodegradable rating. Therefore, it would also save to the environment especially in regards to its disposal. In addition, it is also commercially cheaper than synthetic based oil that exaggerated to be a decent substitution of mineral based lubricant oil [5]. Coconut oil has long been recognized as one of sources of vegetable based oil stock that can be classified as an edible oil due to categorizing as food grade source. Coconut trees are commonly found in tropical countries, including Indonesia, especially in coastal areas. So, abundant volume of coconut oil can be produced from coconut trees grown in Indonesia region. However, as grouped into edible oil, coconut oil is so often to use as a food ingredient. Though, sometimes it has been utilized as massage oils. In fact, the utilizing of coconut oil can still be expanded to such as a source of bio-based lubricant oil so that its added value could be extended. This is attributable to triglyceride contained by coconut oil that has good lubricity properties [6]. Besides, many advantages of coconut oil such as a good solvents [5], producing less emission and hydrocarbon [7], load carrying capacity [8], good thermal properties [9], low volatility [10], high viscosity index [11], and non flammable substances would promote coconut oil to developed as commercially feasible bio-based lubricant as resumed by Chatra et al. [5]. Extensive research to improve the capacity of coconut oil ready as lubricant oil has been underway. Jayadas and Nair [12] introduced a procedure to improve oxidation stability and pour point of some vegetable-based oil including coconut oil. The sample was held on isothermal condition for 10 minutes after rapidly heating up to 500C and liquid nitrogen was utilized for cooling down the sample to -500C at a steady rate of 100C per minute. By adding optimum concentration of Copper oxide nano particles to coconut oil, Thottackkad et al. [13], Khosy et al. [14] was trying too add...
the MoS2 nano particles to a mixture of coconut oil and mineral oil (500 N base-oil) to minimizing the effect of friction and reducing the wear rate. Through ultrasonication, Rashin and Hemalatha [15] synthesized stable fluids by dispersing nano particles of ZNO in coconut oil. This procedure found to increase viscosity of newly generated nano coconut fluid. Most of researches described previously were mainly focused on how to improve the performance of coconut oil as lubricant oil by addition of additives. However, tribological evaluation of coconut oil extracted through different methods to prepare it as lubricant oil is seemingly omitted from the discussions. Meanwhile, the performance of coconut oil as bio-based lubricant might be enhanced due to the higher substance of phenolic compounds. Oxidative stability has been recognized as one of poor characteristics of vegetable oil as industrial lubricant that had to be overcome [16]. Hence, the interesting fact found by Marina et al [17]. It can be an attractive idea to be explore more in finding other benefit of coconut oils that are extracted through different processing methods. In this study, investigation were being focused on physicochemical and tribological properties of virgin coconut oil (VCO), hydrogenated coconut oil (HCO), and refined coconut oil (RCO) that were extracted through wet and dry processing methods. In addition, coconut oil samples were extracted from coconut palm that can be found in coastal area of West Sumatra, Indonesia. This was to confirm Marina et al. [17] and Kumar [18] finding that pinpoint the variation in fatty acid composition from different sources of coconut oil.

THEORY AND EXPERIMENTATION

Bio-lubricant is a term that applies to all lubricants that are biodegradable and non-toxic to environment and humans. The sources of bio-lubricants are numerous and encompass vegetable, animal, and marine sources [4]. As base oils for bio-lubricants, vegetable oils are attractive because they are mostly biodegradable and are made from edible feed stocks. Even though, vegetable oils are superior compare to mineral oils in term of biodegradability, excellent lubricity, renewable, and non-toxic but they are not capable of completely replacing the standard petroleum-based standard lubricants. Because vegetable oils have lack of physical properties, such as: oxidative stability, thermal stability, and viscosity range, the petroleum-based lubricants give high performance physical properties. However bio lubricants formulated from vegetable oils have the following advantages derived from the chemistry of the base stock [4]: (a) Higher lubricity, (b) Lower volatility, (c) Higher viscosity indices, (d) Higher shear stability, (e) Higher detergent, (f) Higher dispersant, and (f) Rapid bio degradation. To increase the physical properties of vegetable oils, the addition of additives, blending or modification of chemical means can increase the total cost and toxicity, and decrease biodegradability [19].
Table 2: Physicochemical properties of coconut oils

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Wet</th>
<th>VCO</th>
<th>Dry</th>
<th>Koshy [15]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Viscosity at 40°C, cSt</td>
<td>26.44</td>
<td>25.82</td>
<td>25.35</td>
<td>28.56</td>
</tr>
<tr>
<td>Viscosity at 100°C, cSt</td>
<td>5.391</td>
<td>5.664</td>
<td>5.754</td>
<td>6.76</td>
</tr>
<tr>
<td>Viscosity index</td>
<td>143.88</td>
<td>169.13</td>
<td>180.51</td>
<td>142</td>
</tr>
<tr>
<td>Density at 15°C, kg/L</td>
<td>0.9262</td>
<td>0.9257</td>
<td>0.9257</td>
<td>0.915*</td>
</tr>
<tr>
<td>Flash point, °C</td>
<td>307.5</td>
<td>309.5</td>
<td>309.5</td>
<td>278</td>
</tr>
<tr>
<td>Pour point, °C</td>
<td>21</td>
<td>21</td>
<td>21</td>
<td>20</td>
</tr>
<tr>
<td>Total acid number (TAN), mg KOH/g</td>
<td>0.73</td>
<td>1.62</td>
<td>1.64</td>
<td>-</td>
</tr>
<tr>
<td>Total based number (TBN), mg KOH/g</td>
<td>0.04</td>
<td>NP</td>
<td>0.02</td>
<td>-</td>
</tr>
<tr>
<td>Iodine value, g I₂ 100 g</td>
<td>8.65</td>
<td>7.99</td>
<td>9.16</td>
<td>6-8</td>
</tr>
</tbody>
</table>

*at temperature 25°C

C. Wear and friction tests

Wear and friction tests were conducted in accordance to ASTM standard G99 using a pin on disc test apparatus. The apparatus enables in determining the wear magnitude by calculating the volume of material lost as a result of rubbing in (probe) against the flat face of a rotating disc. Further, the coefficient of friction was determined from the ratio of frictional forces measured by using load cell attached to a flexible arm and the loading forces determined from the weight loaded on the pin. The friction of coefficient was measured at steady state condition.

D. The test specimens

The test specimen comprised of a 7.938 mm diameter of 440C stainless steel pin and a 100 mm diameter of AISI 1015 disc. The flat faces of AISI 1015 disc were ground to have a surface finish of 0.8 μm Ra and to ensure its parallelism of the surface. The measured surface hardness of the pin and the disc was 610 and 135 BHN, respectively. The chemical composition of these materials were listed in Table 1.

E. Test procedures

All tests were set at room temperature. The wear test was performed using a pin on disc apparatus under lubricated condition. The 440C stainless steel pin was mounted vertically in a steel vice such that its face would be pressed against a rotating AISI 1015 disc. The holder along with the steel 440C stainless steel pin was positioned at a particular track diameter. A track radius of 30 mm was selected for this experiment and was kept constant for the entire observation. For each test, the pin, lubricant sample, new AISI 1015 disc were used. The test was conducted by dripping down lubricant sample to rotate the top surface of the disc and the pin was pressed with the constant pressure against the rotated surface by using the flexible arms. After completion of the test, the pin was taken out from observation area to get cleaned with alcohol and dried. While the disc was removed and replaced with a new disc. The removed disc was then cleaned with alcohol and dried to further being weighed by a balance having tolerance of 0.01 g to determine the mass loss due to wear. The difference in the mass measured before and after the test would indicate the wear of the AISI 1015 disc. The ratio of mass loss of sliding distance was defined as wear rate. The wear test was carried on by keeping the load, speed and time at a constant value. The rotational speeds were 500 and 1400 rpm while the time was set to increase every 10 minutes of total 50 minutes period.

RESULTS

A. Physicochemical properties of coconut oils

The results of physicochemical analysis such kinematic viscosity values, density, flash point, pour point, total acid number (TAN), total based number (TBN), and iodine value are shown in Table 2. The analysis were performed in order to characterize the most relevant properties of the extracted coconut oils from dry and wet processing. From Table 2, it shows that the physicochemical properties varied among coconut oils (HCO, VCO, and RCO) unless the pour point values.

B. Fatty acid composition

The properties such as viscosity, viscosity index (VI) are of great importance for lubricant oil. Those properties can be influenced by the structure of the ester molecules that are characterized by the composition of fatty acid. Fatty acid contained in vegetable oil is attributable to superior lubricity capacity. In addition, it could act as oiliness agents to assist the formation of the protective films that are influenced by physical adsorption and chemisorptions between the substrate and inorganic elements. Composition of fatty acid of coconut oils extracted from dry and wet processing can be classified into two groups; saturated fatty acids and unsaturated fatty acids. The results from this present
study are presented in Figs. 1, 2, 3, and 4 for composition of fatty acid, ratio between unsaturated and saturated fatty acids, percentage of saturated fatty acid, and percentage of unsaturated fatty acid, respectively.

Figure 1: Composition of saturated fatty acid and unsaturated fatty acid in extracted fatty acid in extracted coconut oils

Figure 2: Ratio between unsaturated and saturated fatty acids in extracted coconut oils

Figure 3: Percentage of saturated fatty acids in extracted coconut oils

Figure 4: Percentage of unsaturated fatty acids in extracted coconut oils

C. Wear and friction properties

The investigation of tribological behavior of coconut oils was completely obtained using a pin on disc tester. The abrasive wear occurred on discs has been investigated at different extracted coconut oils. One test of wear from each lubricant is presented in the same graph to get a comparison of the time behavior in the first 300 seconds during running-in period. Wear and wear rate of coconut oils extracted through different processing method are shown in Figs. 5 and 6. Fig. 5 exhibits wear progression of the disc used in this study over a sliding time. Wear volume increases as sliding time progresses. From Fig. 5, it is revealed that the volume of material removed from the disc at 500 rpm was higher than 1400 rpm. HCO and RCO oils were lower wear mass at 500 and 1400 rpm, respectively. This occurred at 100 N of load. In general, at the first 300 seconds of running-in period, wear rate value for all extracted coconut oils was maximum in the first 10 seconds where HCO and RCO were lower wear rate at 500 and 1400 rpm, respectively as shown in Fig. 6.

Figure 5: Wear progression of discs under different extracted methods of coconut oil at normal load of 100 N and rotational speed of 500 and 1400 rpm
Fig. 6 shows wear rate of discs under different extracted methods of coconut oils at normal load of 100 N and rotational speed of (a) 500 and (b) 1400 rpm.

Fig. 7 shows coefficient of friction of discs with different coconut oils, loads and disc rotations. From the figure, it shows that coefficient of friction was depended on speed and load. In general, HCO and RCO had the lowest coefficient of friction among the coconut oils at 500 and 1400 rpm, respectively.

D. Surface texture analysis

Fig. 8 shows the surface texture of pins and discs obtained from wear test that was captured by using an optical microscope. In Fig. 8, it exhibits the different surface finish the pins and discs at three different extracting methods of coconut oil: HCO, VCO, and RCO. From Fig. 8, it reveal that abrasive wear occurred in the middle of the disc. The most common form of that damage is the loss or displacement of material and volume of material removed or displaced. They can be used as measuring wears. As the two surfaces in contact (pin on disc) due to high-pressure load, it can induce plastic deformation on the surface of the disc. The plastic deformation occurred on disc was depended on the type of lubricant applied. This was shown in Fig. 8. The surface texture of the worn surface of the disc was changed from smoother to rougher when rotational speed increased from 500 to 1400 rpm. The transition of surface texture was marked by of decreasing of wear rate.

Fig. 8: Worn surfaces of the pins and discs captured by microscope optic for various coconut oils, normal load of 100 N and rotational speed of (a) 500 and (b) 1400 rpm for 50 minutes.
Thus, it is preferable in the lubricant industry due to the high rate of change in viscosity. This means that RCO undergoes a high viscosity index among extracted coconut oils. Therefore, it would affect the water content of the extracted oil. The differences among extracted coconut oils were reported by Koshy et al. [14], Mia and Ohno [20], Gopala et al. [21], and H. Noureddini et al. [22]. This might be influenced by the source of coconut milk. Coconut milk used in this study originated from West Sumatra, Indonesia where it has been recognized as having high rain intensity; thus, it would affect the water content of the extracted coconut oil. The differences among extracted coconut oil that can be identified are regarding to viscosity index (VI) and iodine value. In this study, RCO showed the highest viscosity index among extracted coconut oils. This means that RCO undergoes a high rate of change in viscosity. Thus, it is preferable in the lubricant industry due to ther-
fatty acid found in VCO was much higher compared to theoretical composition. This might be influenced by processing of VCO from coconut milk. The method of process VCO from coconut milk was fermenting naturally. The major saturated fatty acid obtained from the coconut oils were lauric and myristic acids. The results from this study, showed that the percentage of the lauric acid from 48.34 % for RCO to 48.61 % for HCO. These findings were similar with those of Kostik et al. [26] and Gopala et al. [21], where total content of these two saturated fatty acid was found to be 48 ± 4 and 45 - 50 %., respectively. Except for VCO, lauric acid (C12:0) was the major saturated fatty acid (68 %) for all coconut oils, followed by myristic acid (C14:0) only (15.56 %), as shown in Fig. 3. Interestingly, the most percentage of unsaturated acid for RCO and HCO is dominated by oleic acid and linoleic acid (Fig. 4). RCO had lower both oleic acid and linoleic acid compared to HCO.

C. Wear and friction properties

This is owing to at the initial running-in period, initial in conformity between pin and disc. As conformity was achieved, the wear rate value would be declined and conformity is achieved. Therefore, the running-in phenomena is responsible in increasing wear resistance and promoting quick conformity. In the case of the RCO and HCO, these types of extracting coconut oil conceives to have a good stabilization period. This was proved by the lower declination angle of wear rate as sliding time increases. Thus, it implies that RCO and HCO can maintain its performance for longer period of running time. Furthermore, coefficient of friction for various extracted coconut oils was measured after running-in period of 50 minutes after wear test. This measurement was conducted on dynamic condition. Fig. 7 shows the comparison of coefficient of friction of various coconut oils at dynamic condition with different loads. Yet, RCO exhibits better lubrication capability under different loads as shown in Fig. 7. This is consistent with the results shown in Fig. 5 and Fig. 6. When wear can be reduced, the coefficient of friction would be low. The higher contain of unsaturated fatty acid in RCO and HCO is also responsible in reducing coefficient of friction [28]. In addition, the regime of lubrication of coconut oils can influence coefficient of friction. To see the regime of lubrication, it can be explained by using Strubeck curve. The physical and tribological properties of coconut oils can also influence coefficient of friction in the lubrication regime. The Strubeck curve plays an important role in identifying boundary, mixed, elastohydrodynamic, and hydrodynamic lubrication regimes. Recent advances in elastohydrodynamic lubrication together with rough surface interaction have made it possible to develop a methodology for predicting the trend of the Strubeck curve. In prediction of Strubeck curve, the Greenwood and Williamson [29] contact model and the Hamrock and Dowson elastohydrodynamic [30] film thickness were used. The results of a theoretical prediction of the Strubeck-type behavior are shown in Fig. 11. In this figure shows that the area of investigation of wear properties was mixed lubrication regime. This is consistent with Fig. 6 where the order of wear rate was 10-4 - 10-6 mm3/Nm and it is also consistent with [31]. In mixed lubrication regime both viscosity and fatty acid would influence lubricity. Based on investigating of R Ishida et al. [32], it reveals that the concentration of unsaturated fatty acid has affect on change in friction behavior.

D. Surface texture analysis

Based on IRG transition diagram, transition to severe-wear regions depends on the metalurgy of the surface and the tribochemistry of the lubricant [33]. From Fig. 6, the wear transition by applying of VCO shows severe wear damage compare to HCO and RCO. This fact is due to oil creates a lubricating layer between asperity to prevent from exaggerated wear, hence inducing sliding dominantly than sticking. This confirms from Fig. 9 that VCO could increase depth of wear and decrease scar width/scar diameter of discs and pins, respectively. Meanwhile, for the surface that was pre-lubricated with RCO dan HCO, result in a rough surface and creates a coherent profile. This is due to a weakened very thin layer formed in the asperity caused by diffusion of lubricant into the disc as the disc and lubricant interact. Diffusion of various coconut oils was different owing to the different structure of fatty acid composition of each coconut oil. Higher saturated fatty acid composition as found in VCO generated weaker protective layer [34] hence creating a more apparent surface texture as shown in Fig. 8. However, as a consequence of having an excess composition of certain saturated fatty acid leads to poor cold flowing characteristics of the lubricant [35] hence it generated high surface roughness as experienced by VCO. Moreover, triglyceride structure of vegetable oils also contributes to desirable qualities of a lubricant since the long, polar fatty acid chains could provide high strength lubricant films that interact strongly with metallic surfaces thus low friction and wear can be ascertained. From foregoing discussion it can be seen that RCO showed

Figure 11: Regime lubrication of Strubeck curve based on mixed lubrication model on pin on disc
the lowest saturated acid among coconut oils, which it had the lowest lauric acid. In addition, in unsaturated fatty acid constituents, RCO had oleic acid and linoleic acid which were low in oleic acid and linoleic acid. Since the major saturated fatty acid constituent of coconut oils is lauric acid it has slightly inferior tribological properties compared to high oleic vegetable oils [36]. According to Fox et al. [37], the level of unsaturated fatty acid has a noticeable effect on boundary lubrication. Especially for oleic acid consistently improved the wear performance, however, linoleic acid have no any significantly effect on wear.

CONCLUSIONS

Based on the results obtained from the various test carried out to investigate physicochemical and tribological properties of coconut oils planted in West Sumatra, Indonesia, the following conclusions can be drawn. The present study measured and examined the physicochemical and tribological properties of coconut oils extracted through dry processing (i.e. RCO) and wet processing (i.e. HCO and VCO). The extracted process of coconut oils could influence physicochemical and tribological properties. RCO has good physicochemical property in terms of viscosity index. Thus, higher viscosity index of RCO would make it possible working at higher temperatures without affecting its performance. The saturated nature of its fatty acids constituents of coconut oils is very higher than unsaturated fatty acid, within the range from 48.34 - 68.73 % for lauric acid to 15.56 - 18.99 % of myristic acid for saturated acid and the range from 0.97 - 6.3 % oleic acid to 0.24 - 1.59 % linoleic acid. RCO had the lowest saturated acid among coconut oils and lower unsaturated acid compare to HCO. These chemical properties were good for lubricity. Physical and tribological properties of RCO had resulted in good wear and friction when operational condition of RCO lubricant in the mixed lubrication. In this regime, viscosity and fatty acid contained of RCO have effect on friction of coefficient. On the other hand, RCO had good in high viscosity index and the lowest saturated acids. Coconut oils are naturally suitable to be used as base lubricant oils. The results show that among extracted coconut oils, the effect of processing could influence viscosity, viscosity index and percentage of fatty acids contained. However, the pour point of coconut oils is very high so the chemical modification is needed to reduce its pour point. Dry processing of coconut oil (RCO) could be as an alternative to improve physicochemical and tribological properties of bio-based lubricant. This was evident from low wear rate, low coefficient of friction and better surface finish of the disc dripped by RCO that was produced through dry extraction.

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