



Wear Behaviours of Sustainable Biolubricants: Influence of Fatty Acid Compositions and Surface Roughness in Mixed Lubrication Regimes

Dedison Gasni*, Haznam Putra, Rio Muhammad Nur

Department of Mechanical Engineering, Faculty of Engineering, Universitas Andalas,
Kampus Limau Manis, Padang, Sumatera Barat, 25163, Indonesia

***dgasni@eng.unand.ac.id**

Abstract. The advantage of biolubricant compared to mineral oil is that biolubricant can ensure environmental sustainability. This research aims to investigate sustainable biolubricants of virgin coconut oil (VCO) and olive oil on wear behaviours with different fatty acid compositions and surface roughnesses in the mixed lubrication regimes. Tests were carried out on pin-on-disc equipment at a speed of 500 rpm with loads of 50 and 100 N. We used two types of biolubricants and surface roughness of the disks (0.8 and 6.3 μm). The research results show that the lauric acid content in VCO could reduce the wear rate of the disk. The surface roughness of the disc had a significant influence on the wear rate for both biolubricants; the smoother the surface of the disc, the more wear rate will decrease. The effect of surface roughness of the disc with both biolubricants could reduce the scar width of the disc and the scar diameter of the pin. The scar width of the disk was higher when compared to the scar diameter of the pin; by using VCO, there was a decrease of percentage in scar width of 32% by using smooth surface. VCO could be a promising sustainable bio-based lubricant in the future, especially in the mixed lubrication regimes.

Keywords: vegetable oil-based lubricant, sustainable biolubricant, wear analysis, sustainable tribology, surface roughness, mixed lubrication regime

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1. Introduction

Mineral oil-based lubricants have several disadvantages, including being non-renewable and toxic to the environment [1-3]. Sustainable biolubricants are an alternative to mineral-based lubricants in the future because they have several advantages, namely being biodegradable, non-toxic, and environmentally friendly [1]. In addition, biolubricants have good tribological properties, especially against friction and wear, because they contain fatty acids that are not found in mineral oil [4]. In the mixed lubrication regimes, a contact occurs between two surfaces due to shared contact between

asperities and asperities and contact separated by a lubricant layer. In this regime, the surface roughness and chemical composition of the lubricant acting as a layer separating two contact surfaces play a very important role in improving the tribological properties of the contact.

The surface roughness resulting from a machining process greatly influences engine components that come into contact with each other, such as the piston surface and the cylinder wall of an engine. This surface roughness is very important in the characteristics of product quality and technical requirements for most machined products [5]. Achieving the surface quality requirements for machine components is very important so that the machine components can function properly. Especially for engine components that contact each other and move relatively, such as the cylinder and cylinder walls of an engine, which are lubricated with all lubrication regimes [6], namely boundary lubrication, mixed lubrication, and elastohydrodynamic lubrication.

The mixed lubrication regime is a lubrication regime that lies between hydrodynamic or elastohydrodynamic (EHD) lubrication and boundary lubrication. In this regime, part of the contact surface is separated by lubricant, and part of the contact occurs between asperity and asperity. To understand this mixed lubricant regime, we must not only understand boundary lubrication and elastohydrodynamic lubrication, but we must also understand how surface topography influences fluid behavior and the influence of contact behavior and fluid pressure on surface topography [7]. Figure 1 illustrates the lubrication regime of the mixed lubrication regime via the Stribeck curve. The curve depicts the relationship between the friction coefficient on the y-axis and the logarithmic viscosity, speed, and load known as the Hersey number on the x-axis. In the elastohydrodynamic lubrication regime, the film layer thickness is not influenced by the surface roughness of the contact surface, whereas in the mixed lubrication regime, the film layer thickness limit is influenced by the roughness of the contact surface. So the x-axis can be expressed by the lambda ratio, which is a measure of the thickness of the lubricant layer relative to the composite roughness of the two surfaces in contact with each other. If the lambda ratio is greater than 5 ($\lambda > 5$), the lubrication regime is elastohydrodynamic. Meanwhile, when the lambda ratio is smaller than 3 ($\lambda < 3$), the lubrication regime is boundary lubrication [8-9].

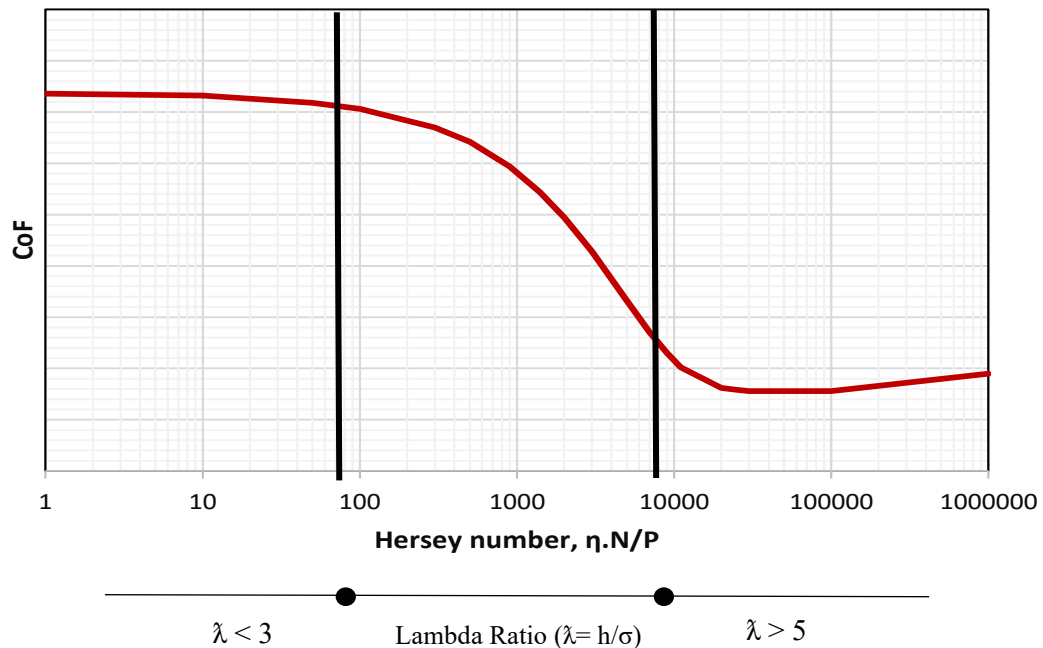


Figure 1. Stribeck curve [10].

The contact surfaces are completely separated by the lubricant when the geometry of the contact surfaces and the operating conditions are such that the applied load is completely supported by the fluid film. This condition is known as elastohydrodynamic lubrication (EHL), where the working load is supported by a thick film layer that separates the two surfaces, the fluid pressure of the film, and the asperity, which undergoes plastic deformation. So that wear and third-body particles are not formed, and in this regime the chemical properties of the lubricant do not play a significant role. If the working load is very large and the rotation speed is low, the hydrodynamic or hydrostatic pressure of the film layer is unable to withstand the load, resulting in asperity-to-asperity contact. This asperity contact will be influenced by many factors, including surface roughness, film fluid pressure, normal load, hardness, and elasticity of the asperities [11-12]. With increasing load, plastic deformation occurs in the asperity and the film layer will decrease so that the load is fully supported by the asperity contact. This will result in mechanical contact on the surface, which will cause wear, deformation, abrasion, adhesion, and fatigue in dry sliding contact [13]. As a result of relative motion on the contact surface, heat will occur, resulting in a chemical reaction between the lubricant molecules and the surface asperities, thus forming a chemical boundary layer in the form of an organic and inorganic surface film. According to Hsu and Klaus, some surface films can function as anti-wear, some surface films are soft, and some films can be detrimental (pro-wear) [14].

Engineering surfaces have microscopic roughness; when two contact surfaces touch in concentrated contact, this roughness deforms both elastically and plastically to form an interface. The interface depends on the relative surface hardness and relative surface roughness of the two surfaces in contact. The apparent contact area for sliding contact may be only 15 to 20% of the apparent contact area [15]. In nature, to describe surface roughness depends on the distribution of roughness heights. It can be Gaussian or non-Gaussian; if Gaussian, average roughness (Ra), root mean square (RMS) roughness, skewness, and kurtosis are used in describing surface roughness.

The chemical content of the lubricant has a significant influence, especially in the boundary lubrication and mixed lubrication regimes, where the film coating the two contact surfaces is very thin. In lubricants made from vegetable oil (bio-based lubricant), the chemical composition of the lubricant is influenced by the fatty acid content, which is not found in lubricants made from mineral oil [16]. Research conducted by Siniawski et al. [16] compared the fatty acid content of two types of vegetable oil, namely soybean and sunflower. The results of their research show that the fatty acid content of these vegetable oils affects the wear that occurs on the contact surface. The use of soybean vegetable oil shows lower wear compared to sunflower vegetable oil because soybean oil is low in linoleic and oleic acid content [17]. Rajasozhaperumal and Kannan researched the effects of fatty acid content in the form of methyl esters in three different types of vegetable oil, namely jatropha, karanja, and cottonseed, and the research results showed that jatropha has good tribological properties when compared with cottonseed and karanja [18].

In the mixed lubrication regime, the lubricant layer effect serves to protect the contact between the asperities and the asperities, which are influenced by the chemical composition of the lubricant. Chemical compositions, such as fatty acids, have a strong relationship to forming an inner layer protecting the contact surfaces from wear. In the same way, the roughness of the surface will influence wear on the contact surfaces. Indonesia, as a tropical country with the largest coconut plantation in the world [19], has not utilized coconut oil for the lubricant industry. Because coconut oil as a basic material for lubricants or as an additive for the lubricant industry has not been widely researched on its tribological properties, unlike olive oil, which grows in subtropical areas and has been investigated as a good bio-based lubricant and additive [1]. For this reason, research is needed on the use of coconut oil as a sustainable biolubricant in the future. In this study, coconut oil (VCO) and olive oil as sustainable biolubricants will be investigated for wear behaviour, including surface morphology of the sliding contact surface on the fatty acid compositions and disk surface roughness in the mixed lubrication regimes.

2. Methods

2.1. Types of Biolubricants

In this study, two types of biolubricant were used that are derived from vegetable oils, namely coconut oil type VCO and olive oil. Coconut oil is an oil that is widely found in tropical areas such as Indonesia. While olive oil is an oil from subtropical areas and has been widely studied for the lubricant industry, this olive oil will be used as a comparison for this study. Both of these oils have different physical and chemical properties. From the physical properties, olive oil has a high viscosity as well as a viscosity index and flash point when compared to coconut oil (VCO); the physical properties for both biolubricants can be seen in Table 1. VCO and olive oil have different fatty acid content, where VCO is rich in saturated fatty acids while olive oil is rich in unsaturated fatty acids. VCO has a high lauric acid content of around 49.98%, while olive oil has a high oleic acid content of around 76.9%, so the ratio between unsaturated fatty acids and saturated fatty acids of olive oil is higher than VCO, as shown in Table 2.

Table 1. Physical properties of biolubricants [4].

Physical Properties	Coconut oil (VCO)	Olive oil
Kinematic Viscosity @40°C, (cSt)	25.82	36.68
Kinematic Viscosity @100°C, (cSt)	5.664	8.348
Viscosity Index	169,13	214
Density @15°C, (kg/l)	0.9257	0.9154
Flash Point, (°C)	309.5	338
Pour Point, (°C)	21	6

Table 2. Fatty acid content of coconut oil (VCO) and olive oil.

Fatty Acid	Biolubricant	
	Cococonut oil (VCO) [4]	Olive oil [20]
Saturated fatty acid		
Caprylic Acid (C8:0)	8.39	-
Capric Acid (C10:0)	6.82	-
Lauric Acid (C12:0)	49.98	-
Myristic Acid (C14:0)	17.61	0.02
Palmitic Acid (C16:0)	8.45	10.5
Palmitoleic (C16:1)	-	0.6
Heptadecanoic (C17:0)	-	0.05
Heptadecenoic (C17:1)	-	0.09
Stearic Acid (C18:0)	6.02	2.6
Unsaturated fatty acid		
Oleic Acid (C18:1)	5.82	76.9
Linoleic Acid (C18:2)	1.37	7.5
Linolenic Acid (C18:3)	-	0.6
Arachidic Acid (C20:0)	-	0.4
Eicosenoic (C20:1)	-	0.3
Behenic (C22:0)	-	0.2
Lignoceric (C24:0)	-	0.1
Ratio between saturated fatty acid and unsaturated fatty acid	0.076	6.205

2.2. Wear Testing Using Pin-on-disc Test Equipment

Wear testing was conducted using a pin-on-disc apparatus according to ASTM 99, where a pin was loaded and in contact with the rotating disk surface as shown in Figure 2. The distance of the pin from the disk axis was 5 mm. The pin was loaded with 50 and 100 N with a disk rotation of 500 rpm. The disc is made of AISI 1015, which has a hardness of 135 BHN. This material is categorized as low-carbon

steel [21]. Whereas the pin is made of stainless steel 400 C with a hardness of 600 BHN. When the pin and disc are loaded, the two surfaces come into contact. The contact that occurs between the pin and disc was in a mixed lubrication regime. In a mixed lubrication regime, some of them contact between asperities and asperities, and some of them the surface is separated by lubricant. The lubricant thickness depends on load, speed, and viscosity of lubricant. According to Hamrock and Dowson [22], central film thickness in elastohydrodynamic lubrication can be calculated by using equation 1.

$$H = 2.69U^{0.67}G^{0.53}W^{-0.067}(1 - 0.6e^{-0.73k}) \quad (1)$$

Where k is an elliptical parameter. H , W , U , and G are nondimensional parameters of film thickness, load, velocity, and material, which are defined as follows:

$$H = \frac{h}{R'}, W = \frac{P}{2E^*R'^2}, U = \frac{\eta_0 u}{2E^*R'}, G = 2\alpha'E^* \quad (2)$$

Where η_0 is the viscosity, α' is the pressure viscosity index obtained by plotting the natural logarithm of the dynamic viscosity against pressure; the slope of the graph is the pressure viscosity index. R' is the reduced radius expressed in the equation below.

$$\frac{1}{R'} = \frac{1}{R_x} + \frac{1}{R_y} \quad (3)$$

Where R_x and R_y are the radii in the x and y directions. Central film thickness can be calculated by using equation 1 with parameters for lubricant using Table 1, and the parameters of the pin and disc used can be seen in Tables 3 and 4, respectively. Surface roughness was measured by a surface roughness tester (Mitutoyo Surftest SJ-310) with two types of surface roughness, namely 0.8 and 6.3 microns, respectively. The lambda ratio can be calculated by using equation 4.

$$\lambda = \frac{h}{\sigma} \quad (4)$$

Where h is central film thickness and σ is a composite of the surface roughness of the disk and the pin [23]. From equation 4, the lambda ratio can be calculated, and the lubrication regime is mixed lubrication according to [24-25], [9].

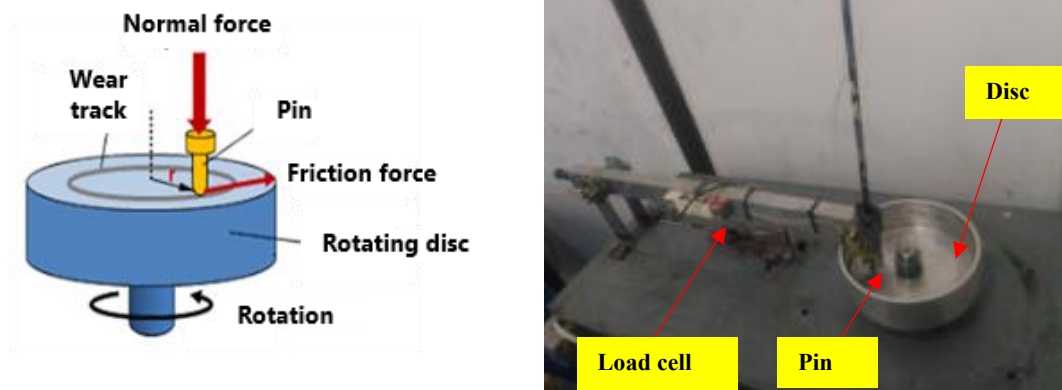


Figure 2. Schematic diagram and apparatus of the pin-on-disc.

Table 3. Pin material and its characteristics.

Type	Pin
Material	Stainless steel 400 C
Hardness	610 BHN
Diameter	7.9 mm

Table 4. Disc material and its characteristics.

Type	Disc
Material	AISI 1015
Kekerasan	135 BHN
Diameter and thickness	20 and 2 mm
Surface roughness	0.8 and 6.3 microns

The lubricant was fed continuously to the contact surface at a constant rate. Wear testing was repeated 3 times for each load, surface roughness, and rotation. Due to the relative motion between the disk and the pin, wear occurred on the surface of the disc and pin. The worn disc surface was measured by weighing every 10 minutes with a total wear test of 50 minutes. The wear rate of the disc surface was calculated based on Archard's wear law. The wear rate is expressed and calculated using equation 5.

$$W = \frac{V}{F.s} \quad (5)$$

where V refers to the wear volume and F.s is the normal load multiplied by the sliding distance. The value of this equation does not depend on the hardness. The sliding distance can be calculated as the linear velocity multiplied by the test duration.

2.3. Investigation of Wear Morphology of Pins and Discs

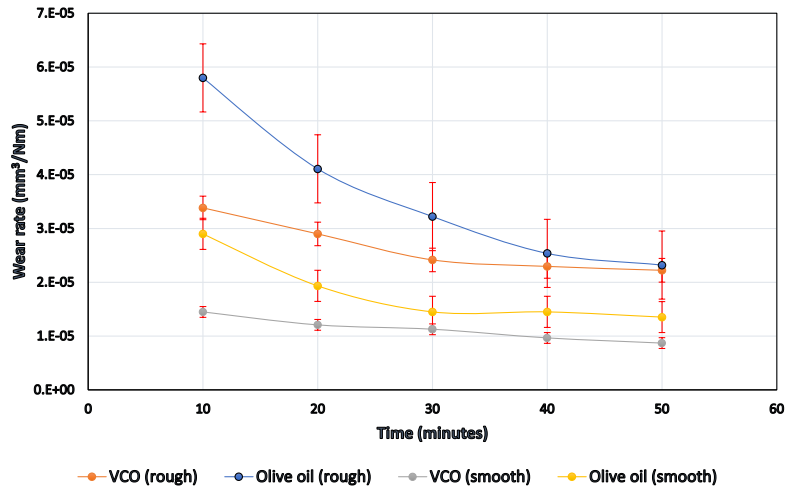
The wear morphology of the pin and disc surfaces was examined using an optical microscope. The optical microscope used was a stereo microscope (Olympus SZX 10) with a magnification of 1000 μm . The scar diameter of the pins and the scar width of the discs were measured. Measurements of scar diameter and scar disc were carried out on 3 samples, and for each sample, 4 measurement points were taken, then the values were averaged. Observations of the morphology of the pin and disc surfaces were carried out after the disc was rotated for 50 minutes with a load of 50 and 100 N and a rotational speed of 500 rpm.

3. Results and Discussion

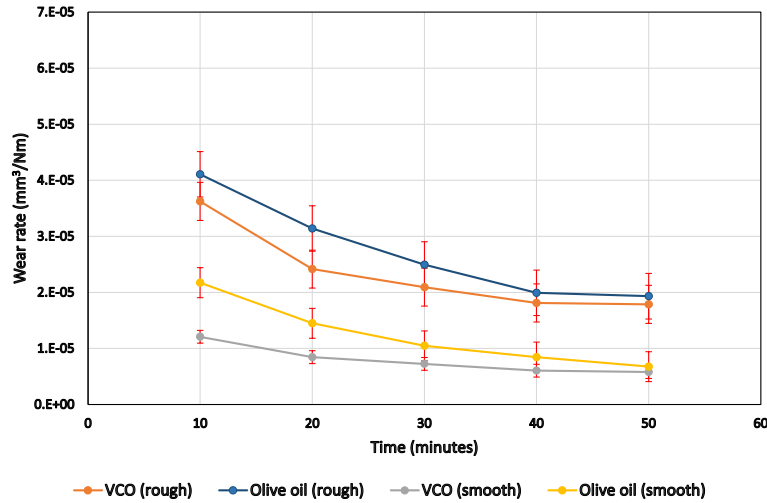
3.1. Wear rate and Scars

Wear rate is the rate at which material is worn away from a surface contact, measured in weight or volume. The wear rate of discs that occur in sliding contact with different biolubricants, forces, and surface roughnesses is shown in Figures 3a and 3b. Figures 3a and 3b are the wear rate of the disc at a load of 50 and 100 N, respectively with a speed of 500 rpm. From the figures, it can be seen that the wear rate was influenced by the chemical composition of the biolubricant, load, and surface roughness. The higher the surface roughness, the higher the wear rate. Likewise, the content of unsaturated fatty acids in olive oil, which is rich in oleic acid, caused the wear rate to increase. During running-in conditions, the wear rate was very high, especially in olive oil lubricants at a load of 50 N, resulting in high wear (severe wear). Along with increasing time, the wear rate would decrease for all lubricants until it reached a steady state where the wear rate approached a constant value. The content of saturated fatty acids in VCO in the form of lauric acid could reduce the wear rate that occurred on the disk. Likewise with the surface roughness of the disk, the smoother the surface of the disk, the lower the wear rate. The effect of load was very significant on the wear rate for both biolubricants; the higher the load, the lower the wear rate of the disc.

From the wear test results in Figure 3, it shows that the wear rate of the disc was lowest when lubricated with VCO. This is because VCO is rich in saturated fatty acids in the form of lauric acid, although in terms of its physical properties, it has a lower kinematic viscosity when compared to olive oil. The effect of viscosity on this wear test does not have much effect at a speed of 500 rpm. From the results of tests conducted by several researchers [17] and [18], it shows that the content of saturated fatty acids can reduce the wear rate because it has a low oleic acid content. According to [26], the effect of load on the wear rate of 35NCD16 steel is that if the load is low, it will result in a high wear rate.



(a) $F = 50 \text{ N}$

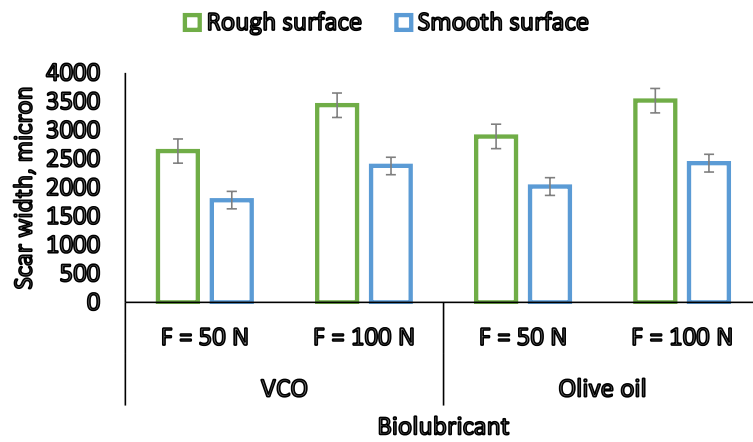


(b) $F = 100 \text{ N}$

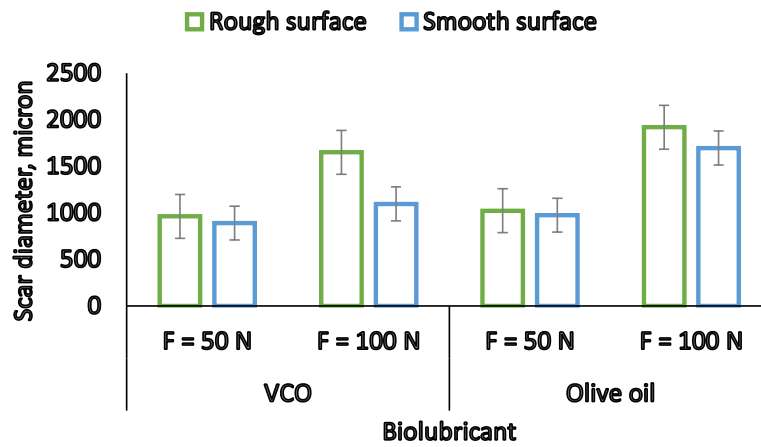
Figure 3. Wear rate of the disc for different surface roughnesses and biolubricants with loads (a) $F = 50 \text{ N}$ and (b) $F = 100 \text{ N}$ and disc rotation speed of 500 rpm.

The results of scar measurements from wear on pins and discs for both types of biolubricants and surface roughnesses are shown in Figures 4a and 4b. From the figure it can be seen that the scar width of the disc was larger than the scar diameter of the pin. Along with increasing load, the scar width of the disc and the scar diameter of the pin would increase. The surface roughness of the disc greatly affected the size of the scar width of the disc and the scar diameter of the pin; the smoother the surface, the size of the scar width and scar diameter would decrease. The saturated acid content in VCO, which is rich in lauric acid, could reduce the scar width of the disk and the scar diameter of the pin.

Surface roughness also greatly affects the wear rate, scar width, and scar diameter, as can be seen in Figure 4. The wear rate of the disc can be reduced by making the surface roughness of the disc smoother [27-28]. The same results were also found by Hanief and Wani [20] on the contact between En31 and steel, showing that surface roughness correlates very strongly with the wear rate; the rougher the surface, the higher the wear rate. Likewise with the scar width and scar diameter that occur on the disc and pin, where the smooth disc surface could suppress the size of the scar on the disc and pin.



(a) Scar width



(b) Scar diameter

Figure 4. Scar width of the disc (a) and scar diameter of the pin (b) with different biolubricants, loads, and surface roughness of the disc at a speed of 500 rpm.

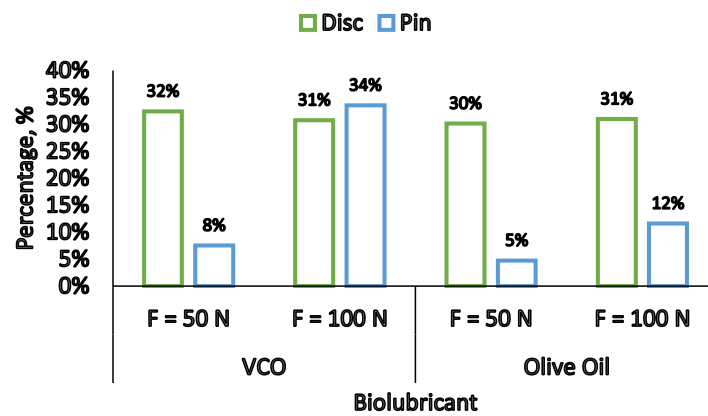


Figure 5. Percentage decrease of scar width of disc and diameter of pin between rough and smooth surfaces.

The percentage increase in wear from the scar width of the disc and the scar diameter of the pin can increase if the surface of the disc used is rougher, as can be seen in Figure 5. From the figure it can be seen that the percentage increase in the scar width of the disc using a rough surface is greater when compared to the percentage increase in the scar diameter of the pin. The higher the load, the percentage increase in scar size increases. A higher percentage increase in scar size occurred when lubricated with VCO oil, where the percentage was around 30 and 31% for the disc and 5 and 12% for the pin for loads of 50 and 100 N, respectively.

3.2. Surface morphology

The surface morphology of the disc and pin lubricated with different biolubricants and surface roughness are shown in Figures 6 and 7. Figure 6 is the surface morphology of the pin with different biolubricants, loads, and surface roughnesses. The shape of the scar formed on the pin surface was in the form of scratches due to abrasive wear on the contact surface. The size and shape of the scar vary, whereas for both biolubricants with increasing roughness and load, the scar size increased. The effect of lubrication using VCO shows that the size and shape of the scar were smaller when compared to olive oil biolubricants. Figure 7 shows the surface morphology of the disc with different variations of load, biolubricant, and surface roughness. The effect of increasing load and surface roughness would cause the scar size to increase. The effect of the load greatly affected the scar formed on the disc surface, where at a force of $F = 100$ N plastic deformation occurs (Table 5). This can be seen from the results of optical microscopy photos, especially in the scar edge area. Lubricants with olive oil produced a wider scar shape than scars with VCO lubricants.

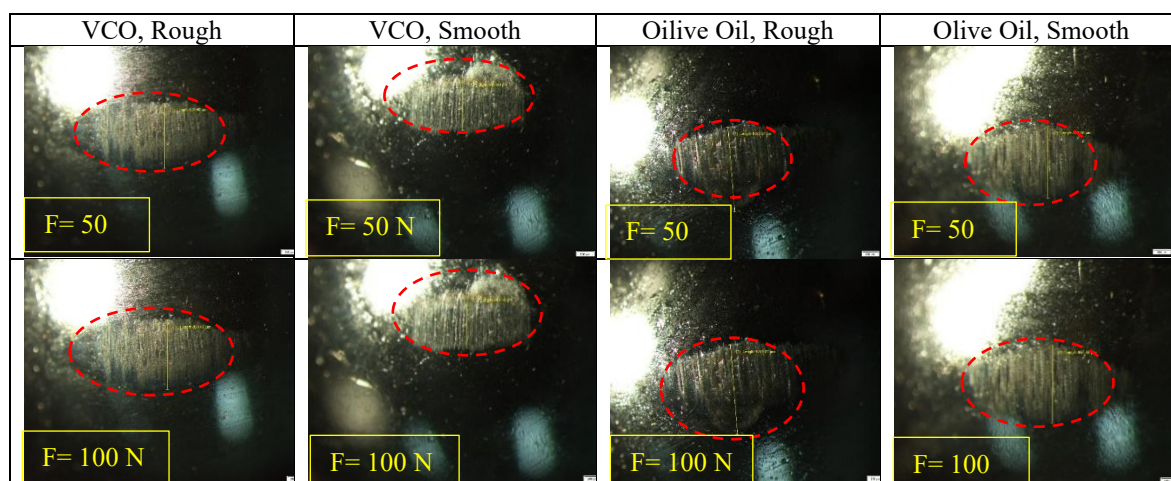


Figure 6. Surface morphology of pins with different biolubricants, loads, and surface roughness at a speed of 500 rpm.

The effect of rough and smooth surfaces of the disc on the contact area can be illustrated in Figure 8, where on the rough surface, the contact that occurs between asperities with asperities is less when compared to the contact on the smooth surface, where the contact between asperities with asperities occurs more in the contact area. So that the maximum pressure that occurs in the contact area on the smooth surface is smaller because it produces a larger contact area when compared to the smaller contact area on the rough surface so that it produces a greater maximum contact pressure. The large pressure in the contact area will cause a high wear rate, and the resulting scar will also be larger so that the width of the scar from the disc and the diameter of the scar from the pin will increase. Analytical calculations of the size of the contact radius and the maximum pressure that occurs in the contact area for loads of 50 and 100 N can be seen in Table 5. In the table it can be seen that the maximum pressure working in the contact area exceeds the yield strength of the disc, so that at loads of 50 and 100 N the disc surface has experienced plastic deformation; this is shown in Figure 7. In this study, the effect of disc hardness has not been studied, where hardness greatly affects its tribological behavior. For future

research, there needs to be a study of the effect of fatty acid content, load, surface roughness and hardness of the disc.

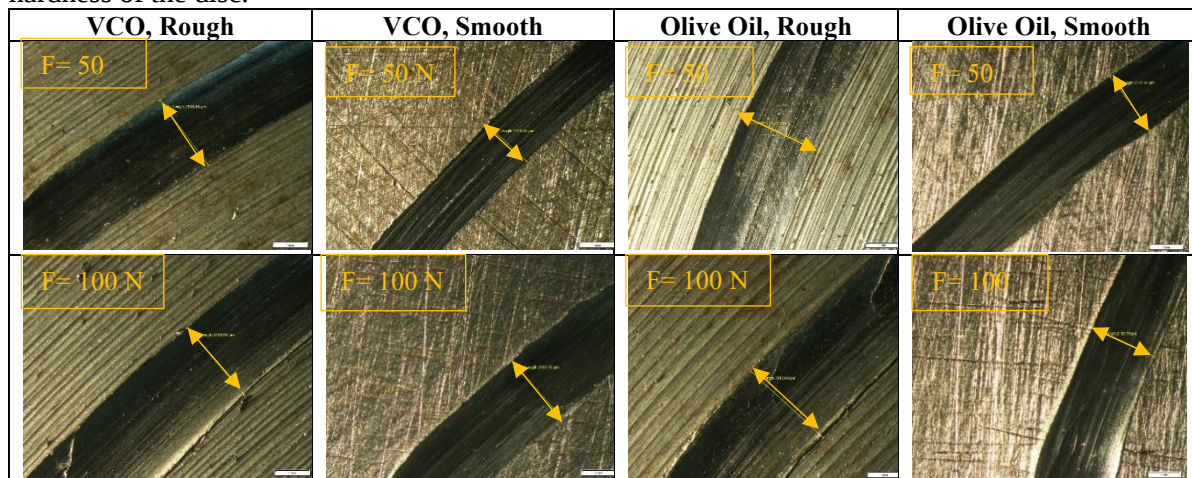


Figure 7. Surface morphology of discs with different biolubricants, loads, and surface roughnesses at a speed of 500 rpm.

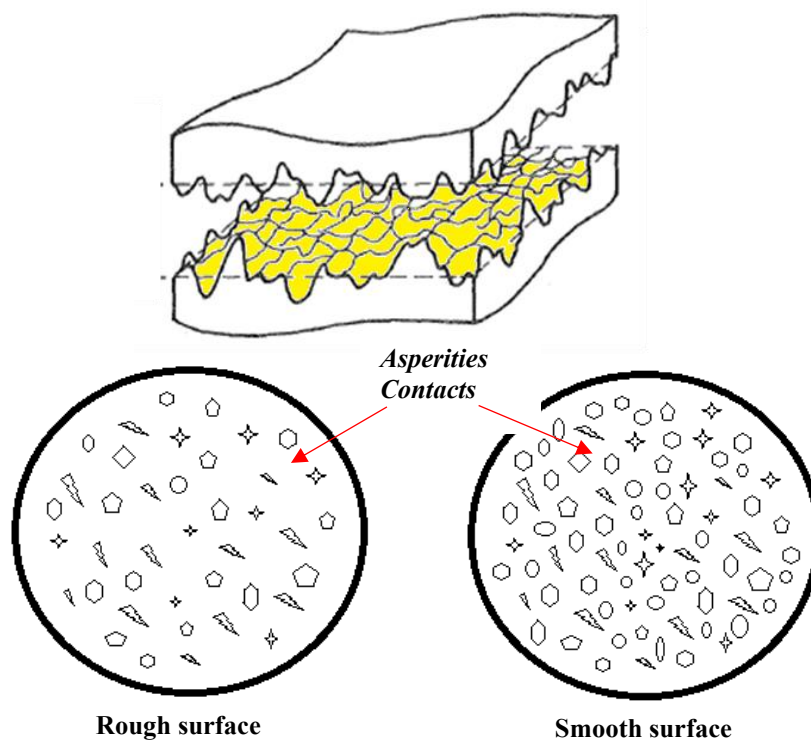


Figure 8. Comparison of contact asperities between rough and smooth surfaces.

Table 5. Differences in contact size and maximum pressure at loads of 50 and 100 N, as well as yield strength and maximum shear stress occurring on the surface of the disc and pin.

No.	Remarks	Load, F = 50 N	Load, F = 100 N
1.	Contact radius	0.00014 m	0.00018 m
2.	Maximum pressure	1.2 GPa	1.5 GPa
No.	Remarks	Disc	Pin
1.	Yield strength	0.46 GPa	2.1 GPa
2.	Maximum shear stress	0.35 GPa	1.58 GPa

4. Conclusion

From the results of the wear rate and surface morphology investigation of the disc and pin that has been carried out on the pin-on-disc testing with two types of biolubricating oils that have different fatty acid content (lauric acid and oleic acid) and different roughnesses of the disc surface (0.8 and 6.3 μm), it shows that: (1) In the mixed lubrication regime, the chemical properties of biolubricants, especially saturated fatty acids in coconut oil (VCO), which is rich in lauric acid, could reduce the wear rate on the disc even though coconut oil has physical properties in the form of a lower kinematic viscosity value when compared to olive oil. The physical properties of biolubricants do not affect this in mixed lubrication regime. (2) The surface roughness of the disc greatly affected the wear rate and scars formed on the surface of the disc and pin; the smoother the surface, the smaller the wear rate and scars formed because the maximum pressure that occurs is smaller when compared to the maximum pressure from the rough surface. (3) The scar width of the disc was larger than the scar diameter of the pin. (4) The percentage increase in scar width from the disc due to high surface roughness was higher when lubricated with VCO compared to olive oil biolubricant, where the percentage increase due to surface roughness reached 32%. (4) VCO as a biobased lubricant has the potential to become a sustainable biobased lubricant in the future, especially in mixed lubrication regimes.

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