Jurnal Tribologi 29 (2021) 72-83

Jurnal Tribologi
Website : jurnaltribologi.mytribos.org e-issn: 2289-7232

Adsorption phenomena of modified vegetable oils on steel contacting surfaces under boundary lubrication regime

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KEYWORDS

Adsorption
Additive
Friction
Boundary lubrication
Modified vegetable oil

ABSTRACT

This study investigated the adsorption phenomena of modified vegetable oils on steel contacting surfaces. Mineral additives are the major additives applied for the modification of vegetable oils. However, an organic additive that is biodegradable is necessary considering the environmental challenges posed by mineral additives. Hence, the study investigated the adsorption of organically modified avocado seed oil and palm *(ElaeisGuineensis L.)* oil on steel surfaces. The organic additive used was ginger and mustard seed extracts. The Gibbs adsorption value of -18.41 kJ/mol for avocado oil at 350^0 C was greater than that of palm oil (-17.26 kJ/mol) implying that the modified avocado seed oil was superior to that of palm oil at elevated conditions (120 – 300 N). The avocado seed oil performed better than the palm oil when modified and performed less than the palm oil in its unmodified condition; the decrease in the value of COF of avocado seed oil from 0.16 to 0.054 proved its serious lubricating ability over the palm oil which witnessed an increase from 0.113 to 0.114. The Gibbs, friction, wear, and physicochemical properties, all suggested better adsorption phenomena of modified avocado seed oil on the steel surfaces over modified palm oil.

Received 12 January 2021; received in revised form 19 February 2021; accepted 14 March 2021.
To cite this article: Chikezie et al., (2021). Adsorption phenomena of modified vegetable oils on steel contacting surfaces under boundary lubrication regime. Jurnal Tribologi 29, pp.72-83.

1.0 INTRODUCTION

Vegetable oils such as avocado (*Persea Americana*) seed oil and palm (*Elaeis Guineensis)* oil are viable and good alternative resources because of their environmental friendliness, non-toxic, and readily biodegradable nature. The triacylglycerol structure with long fatty acid chains and presence of polar groups in the vegetable oils make them amphiphilic; therefore, allowing them to be an excellent choice as lubricants and functional fluids (Amith et al., 2016). The triacylglycerol molecules in vegetable oils orient themselves with the polar end at the solid surface making a closed packed monomolecular or multimolecular layer, resulting in a surface film that provides desirable qualities in a lubricant (Rudnick, 2016). The free fatty acid in vegetable oil such as oleic and linoleic acids which are unsaturated are assumed to be the reason behind its high lubricity (Constantin et al., 2016).

Involvement of biodegradable liquids in lubricating oil preparation needs the understanding of its impact on adsorption and extreme pressure characteristics (Chen et al., 2015). Despite the adsorption capabilities of the biodegradable lubricant and its established excellent qualities like biodegradability, high index of viscosity (VI) and minimal value of coefficient of friction (COF), it still suffers oxidative instability in extreme conditions (Odi-Owei, 1989). The oxidative instability nature of the biodegradable lubricant which is as a result of the triacylglycerol molecules in the oils can be salvaged with the application of the additive.

An experimental study carried out on aluminium alloy lubricated with rapeseed oil using a pin-on-disk tribometer proved that the oil has very inferior friction and wear characteristics when compared with other vegetable oil samples. It was concluded that rapeseed oil requires enhancements before it can be used as a lubricant (Muraki et al., 2016). Studies carried out to investigate the tribological performances of Fatty Acid Methyl Ester (FAME), Hydrotreated Vegetable oil (HVO) from raw rapeseed oil and raw Jatropha Curcas L. oil (JCL) in the lubricating contact pair of AISI E52100 steel sliding against X210Cr12 steel using a ball-on-flat reciprocatory tribometer proved that the friction coefficient decreases with the increase in the frequency, with JCL showing the lowest COF amongst the three oils examined, followed by RME and HVO. The decision was made that the oil exhibited superior tribological properties; hence, could be favourably used as a lubricant in the industry (Ruggiero et al., 2017). Castor oil has been confirmed to have superior tribological properties when compared with mineral gear oil. This was observed in a study carried out to examine the tribological properties of the castor oil using a pin-on-disc tribometer. A decision arrived that castor oil can be used to lubricate contacting surfaces (Bhaumik et al., 2017).

Vegetable oils (avocado seed and palm oils) were found to have reduced wear rate as well as friction; this observation was made with the aid of a tribo-system in an investigation of wear characteristics of aluminium-silicon piston alloy. It was concluded that vegetable oil if properly formulated, can lubricate contacting surfaces better than the conventional mineral oil (Haque and Sharif, 2018).

Mineral additives in the form of nanoparticle of oxides or liquid additives are the major additives applied for the modification of vegetable oils. However, an organic additive that is biodegradable is necessary considering the environmental challenges posed by mineral additives. Despite the adsorption capabilities of the vegetable oil; the vegetable oil still suffers oxidative instability in extreme conditions (Muraki et al., 2016). Some authors (Balan et al., 2020) developed the epoxy-based composite reinforced with waste plastic and seashell powder; the water absorption and wear behaviour of the produced composite were analyzed, and it was concluded that the properties like wear and moisture absorption resistance got improved.

Another study carried out to establish the tribological properties of some vegetable oils using a four-ball set-up proved that the vegetable oils have superior tribological properties than the reference mineral oil on moderate conditions. However, under extreme conditions, vegetable oils were observed to have very poor behaviour. Hence, it was concluded that the vegetable oils if compounded with anti-oxidants will have better extreme pressure properties (Chikezie and Ossia, 2018). Having extensively carried out the literature survey, it was discovered that the vegetable oils have promising potentials to serve as lubricants; but they are hampered by their poor extreme pressure properties which scholars have advised can be improved by the application of additives. In response to this, many authors have used mineral additives to improve the tribological properties of the vegetable oils; some used nano-particles. But in this study, the adsorption of the organically modified indigenous vegetable oils on steel contacting surfaces was investigated to verify their efficacy for boundary lubrication.

There are a couple of ways to determine the potentials of vegetable oil as a lubricant, but a more robust approach that will ascertain the adsorption of the modified vegetable oils on the contacting surfaces is required. Hence, this study presents a comprehensive way of not only determining the tribological responses of the vegetable oils, but also their adsorption on steel contacting surfaces. It is hoped that the study will solve a critical industrial problem.

This study covered the physicochemical characteristics of avocado seed (*Persea Americana*) and palm (*Elaeis guineensis)* oil like viscosity, viscosity index (VI), acid number, free fatty acid (FFA), and iodine value. The friction and wear behaviours of the vegetable oils on steel contacting surfaces were carried out using a tribosystem with a ball-on-disc configuration (6.1.17 version, Austria). The steels used for this research included stainless steel grade AISI 52100 as a ball and a mild steel grade AISI 8620 as the disc. The organic additive used was a mixture of ginger (*Zingiber officinale*) and mustard seed oil (*Argemone Mexicana*).

Enhancement of vegetable oils with an additive as a tribological process is a necessary measure in the modification of their tribological properties. Liquid lubricants that may be applied include the following: mineral oils and biodegradable oils like avocado seed oil and palm oil. The avocado seed littered everywhere in Nigeria as waste is biodegradable; and as such, oil derived from it will be eco-friendly. Hence, can be used to replace conventional mineral oil and edible biodegradable oils such as the one from palm oil. The avocado seed oil was used in this study in comparison with the palm oil, to see if it can have comparable or better tribological characteristics to avoid unhealthy competition with the edible vegetable oils like the palm oil.

2.0 MATERIALS AND METHODS

2.1 Preparation and Extraction of the Vegetable Oils

All the specimens used for the study were procured in mile 3 market, Port Harcourt, Rivers State Nigeria. The chemical method of oil extraction was used with soxhlet extractor with normal hexane as the solvent. 100g of the samples were introduced into various 500ml beaker and a 100ml of the normal hexane gently introduced respectively. After which, the mixture was charged into soxhlet, every connection was perfected to ensure air tight (Egbuna et al., 2013). The entire set-up gripped up with a clamp and is kept on the mantle. Heating was kept on till complete evaporation of normal hexane, which was about 70^0C. The oil was given the necessary time to cool; after which, the oil was removed from the receiver and introduced into the flask for measurement. The ratio of the additive to base oil mixture was 1:10.

2.2 Physicochemical Property Test

Some physicochemical properties tested included viscosity, VI, flash point, pour point, relative atomic number, acid number, FFA, and density. Test materials such as glass capillary viscometer, flash point apparatus, pour point apparatus, and hydrometer were used for the measurement of the properties that are measurable (American Society for Testing and Materials [ASTM], 2017). While properties such as the relative atomic number, VI, iodine number, FFA, and acid number were calculated.

$$VI = \frac{L-U}{L-H}100 \tag{1}$$

U is the oil viscosity @ 40^0C, L and H are the corresponding values at 100^0C.

$$\text{Acid Value (mgKOH/g)} = \frac{Titre\ value\ x\ 5.61}{Weight\ of\ Sample\ used} \tag{2}$$

where titre value is 0.1M of KOH

$$\text{Free Fatty Acid (mgKOH/g)} = \frac{\text{Acid Value}}{2} \tag{3}$$

$$\text{Iodine Number} = \frac{(b-a)\ X1.269}{Weight\ of\ Sample} \tag{4}$$

2.3 Wear and Friction Materials

The tribometer 6.1.17 version, Austria with a ball-on-disc configuration was applied to investigate the friction and wear performances of the friction pairs on lubricated conditions. A hard steel ball of 6.0mm diameter reciprocates the normal load and a 1 mm stroke length at a frequency of 10 Hz for 506s and 55 [%] humidity. The lubricant temperature was kept at 29^0 C; the wear and friction coefficient was measured by a piezo-electric force transducer. Both ball and disc were cleaned by the ultrasonically agitated bath of acetone and toluene before and after the test. Each test was carried out three times to provide mean values of friction coefficient and wear rate. The tribometer estimated the wear rate using the worn track section measured during the pin-on-disk. Measurement of the lubrication regime was by the stribeck curve.

The disc materials used for this study were steel of grades: AISI 8620 with tensile strength 530 MPa, Rockwell B hardness 80 GPa, Poisson ratio 0.27, shear modulus 80 GPa and ball material of grade AISI 52100 with Poisson ratio 0.29, shear modulus 80 GPa.

$$\mu = \frac{F}{W} \tag{5}$$

where μ is COF, F is frictional force and W is normal load.

3.4 Determination of Adsorption of the Oils on Steel Contacting Surfaces

The analysis of the adsorption of modified vegetable oils on the steel contacting surface was carried out using the friction-derived adsorption isotherm based on Langmuir adsorption model (Biresawa *et al.,* 2017).

$$\Delta G_{ads} = \Delta G^0 + \alpha\theta \tag{6}$$

where ΔG_{ads} is the Langmuir adsorption, ΔG^0 is Gibbs adsorption, α is the free energy of lateral interaction of additives, and θ the fractional surface coverage.

Langmuir assumes the net free energy of lateral interaction of the additive is inconsequential ($\alpha \cong 0$); then the Langmuir adsorption equation reduces to the Gibbs adsorption equation:

$$\Delta G_{ads} = \Delta G^0 \tag{7}$$

Also, the Gibbs adsorption equation:

$$\Delta G^0 = -RTlnKo \tag{8}$$

where R is the ideal gas constant (8.314J.mol/K), T is adsorption temperature (K), and Ko the gradient of θ^{-1} against C^{-1}.

$$K_o = \frac{\theta^{-1}{}_{max} - \theta^{-1}{}_{min}}{C^{-1}{}_{max} - C^{-1}{}_{min}} \tag{9}$$

where $\theta^{-1}{}_{max}$ is the maximum value of the reciprocal of fractional surface coverage, $\theta^{-1}{}_{min}$ is the minimum value of the reciprocal of fractional surface coverage, $c^{-1}{}_{max}$ is the maximum value of the reciprocal of the concentration of the modified vegetable oil, and $c^{-1}{}_{min}$ is the minimum value of the reciprocal of the concentration of the modified vegetable oil.

$$\theta = (COF_{unm} - COF_{diff.\,conc.})/(COF_{Unm} - COF_{Mod.}) \tag{10}$$

where COF_{unm} is the coefficient of friction of the unmodified vegetable oil, $COF_{diff.conc}$ is the coefficient of friction of the modified vegetable oils, $COF_{Mod.}$ is the coefficient of friction of the modified vegetable oil, and θ is fractional surface coverage.

3.0 RESULTS AND DISCUSSION

3.1 Physicochemical Characteristics of the Oil Samples

Behaviours of the unmodified and modified avocado seed oil and palm oil are illustrated in Table 1. The characteristics examined included VI, viscosity, density, pour and flash points, molecular weight, acid number, FFA, and Iodine number.

The decrease in pour point and increase in flash point of the vegetable oils after modification suggest the modified vegetable oils have been enhanced to have better oxidative characteristics when compared with their original form (Emmanuel and Mudiakeoghene, 2015). The decrease in pour point of the vegetable oils which was more prevalent in avocado seed oil shows the vegetable oil's pour point has enjoyed suppressants modification. The modification of these vegetable oils did not have any effect on the viscosity; this is an indication that the organic additive is an extreme pressure additive rather than viscosity improver. The decrease in both acid and iodine numbers are indications that the additive has broken some unsaturated bonds to saturated; making it possible for the oils to lubricate better in extreme conditions than at a low temperature (Alves et al., 2017). The excellent pour point and flash point performances of the avocado seed oil that is very much comparable with the mineral oil, suggests that it can perform better than the mineral oil under boundary lubrication regimes (Chikezie and Ossia, 2018).

Table 1: Physicochemical properties of unmodified and modified vegetable oils.

Properties	**Unmodified**		**Modified**	
	Avocado	**Palm**	**Avocado**	**Palm**
Viscosity at 40^{0}C (cSt)	32.60±0.1	36.60±0.1	37.40±0.1	36.60±0.1
Viscosity at 100^{0}C (cSt)	6.21±0.1	6.80±0.1	6.91±0.1	6.80±0.1
Viscosity Index (VI)	129	136	142	136
Pour point (^{0}C)	7±0.5	9±0.5	-3.20±0.5	6±0.5
Flash point (^{0}C)	195±0.5	188±0.5	207±0.5	190±0.5
Relative Atomic Number (g/mol)	200	256	200	256
Acid Number	0.35±0.1	2.41±0.1	0.20±0.1	2.30±0.1
Iodine Number	7±0.1	21.9±0.1	4.00±0.1	19.80±0.1
Free Fatty Acid (FFA)	0.18±0.1	1.21±0.1	0.10±0.1	1.15±0.1
Density (g/cm^3)	0.85±0.1	0.89±0.1	0.88±0.1	0.87±0.1

Avocado seed oil having the least acid value of 0.35 and small iodine number of 7 followed by palm oil with 2.41 and 21.9 as its acid and iodine numbers respectively, could be attributed to the saturated fatty acid's presence in both oils (Uchechi, 2015). The physicochemical characteristics of the modified vegetable oils is suspected to be as a result of the oleoresin and polyphenols in the ginger extract and the erucid acid in mustard oil that acts as inhibitor of FFA oxidation (Eleazu and Eleazu, 2017). The increase in the density of avocado seed oil after modification suggests that it may have more additive concentration than the modified palm oil. Hence, may adsorb more than it.

3.2 Tribological Responses of Unmodified Vegetable Oils under Moderate Loading Conditions

Figures 1 and 2 present the coefficient of friction (COF) of steel sample AISI 8620 under moderate (2-20N) lubricated conditions as simulated by a tribo-system, with a contact frequency of 10 Hz for 506 rubbing duration. Figure 3 summarizes the mean COF of moderate (2-20) N lubricated conditions of the steel sample. The base oils involved included avocado seed oil and palm oil, with the palm oil having the less COF in the range of 0.06 to 0.084 and as such, the most superior in terms of lubricity followed by avocado seed oil with COF of 0.09±0.01 to 0.11±0.01. The wear rate of the oil samples under moderate loading conditions (2-20) N as presented in Figure 4 responded in the same manner with the COF, with the palm oil's response in the loading conditions: 0.02012±0.01 mm^3/Nm to 0.07001 ±0.01 mm^3/Nm and avocado seed oil being the least in wear reduction with 0.02861±0.01 mm^3/Nm to 0.07341±0.01 mm^3/Nm.

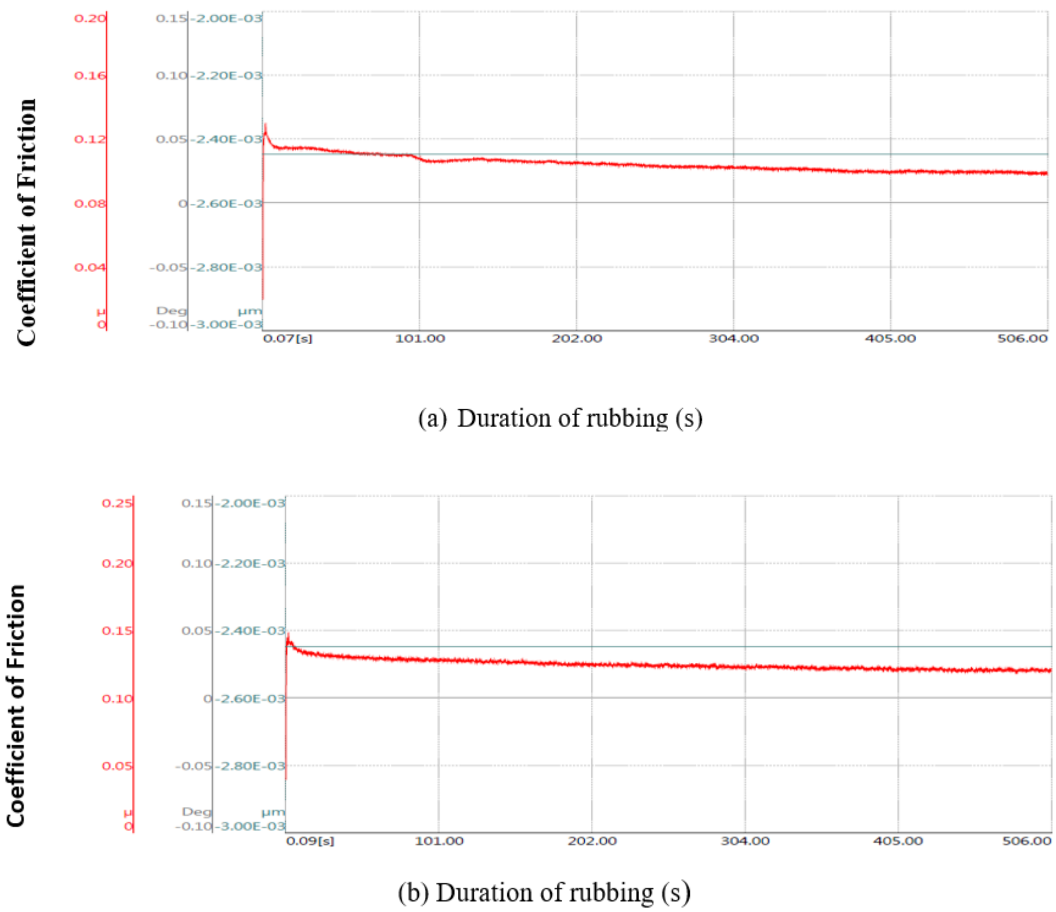


Figure 1: Friction coefficients of the lubricated conditions of the steel samples at 8N as a function of time: (a) Steel Sample with avocado seed oil and (b) steel sample with palm oil.

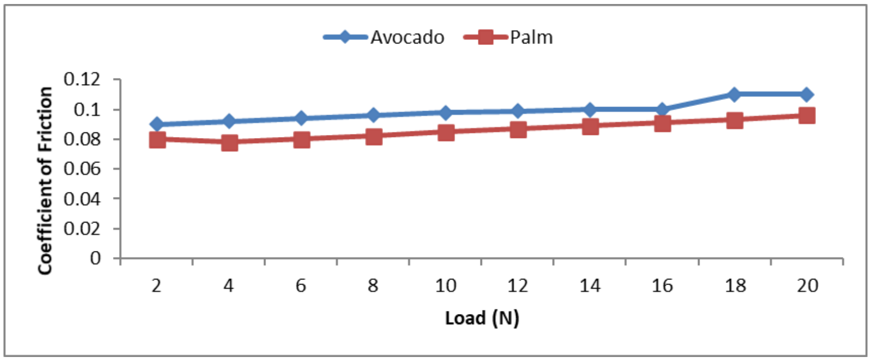


Figure 2: Mean coefficient of friction of the unmodified vegetable oils under moderate loading conditions.

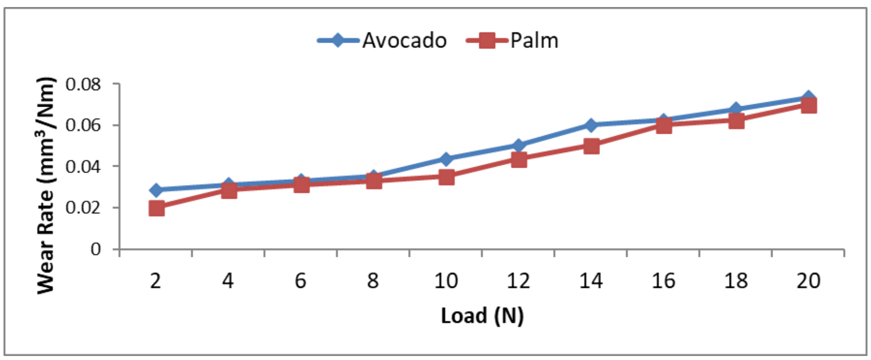


Figure 3: wear rate of the unmodified vegetable oils under moderate loading conditions.

As evidenced in Figures 1, 2, and 3, it is clear that all the oils possess antiwear properties. This result is in line with an earlier result obtained by Haque and Sharif (2018). It can as well be observed that this antiwear behaviour is more prevalent in palm oil than in avocado seed oil under unmodified conditions. This trend suggests that the antiwear property of the vegetable oils is a function of its chain length (Castle and Bovington, 2018). In all the results acquired from the tribometer, it is obvious that prolonged operation gave rise to a lower COF (see Figures 1a and 1b). All materials under study have shown a sharp decline in COF at the beginning, and after some time of operation, its slight increase taking place (see Figures 1a and 1b).

Figure 2 summarizes the mean friction coefficient of the lubricating oils at moderate loading conditions for the disc sample. Expectedly, all the lubricants showed slight positive gradient signifying a linear relationship between COF and load. Palm oil exhibited best lubricity throughout and avocado seed oil showed the worst tribological behaviour under this moderate loading condition.

3.3 Tribological Responses of Modified Vegetable Oils and Mineral Oil on Sample under Boundary Lubrication Condition

Figure 4 illustrates the response (COF) of the modified vegetable oils of avocado seed oil and palm oil on the steel sample which was investigated to ascertain which amongst the vegetable oil samples has superior antiwear property when subjected to extreme pressure condition. It is observed that addition of organic additive to the vegetable oils, only improved the lubrication behaviours of the avocado seed oil significantly, but could not alter the behaviour of palm oil at elevated conditions. The avocado seed oil lubricated better than the palm oil; the decrease in COF of avocado seed oil from 0.16 $\pm$0.01 to 0.054 $\pm$0.01 proved its serious lubricating ability over the palm oil which witnessed an increase from 0.113 $\pm$0.01 to 0.114$\pm$0.01.

Figure 5 presents wear rate of modified vegetable oils under boundary lubrication regime. The Figure shows avocado seed oil as having superior wear rate over the palm oil. Avocado seed oil decreased from (0.28642 $\pm$0.01 to 0.09012$\pm$0.01) mm^3/Nm, palm oil increased from (0.32248 $\pm$0.01 to 0.38897$\pm$0.01) mm^3/Nm.

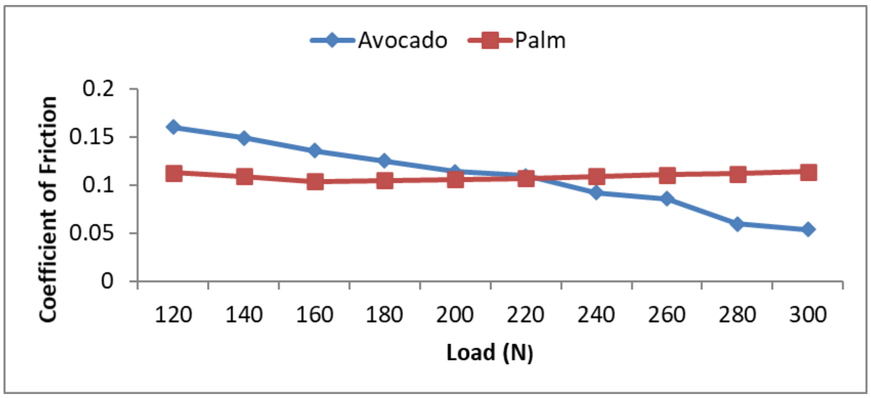


Figure 4: Coefficient of friction of the modified vegetable oils and the mineral oil on disc sample under boundary lubrication condition.

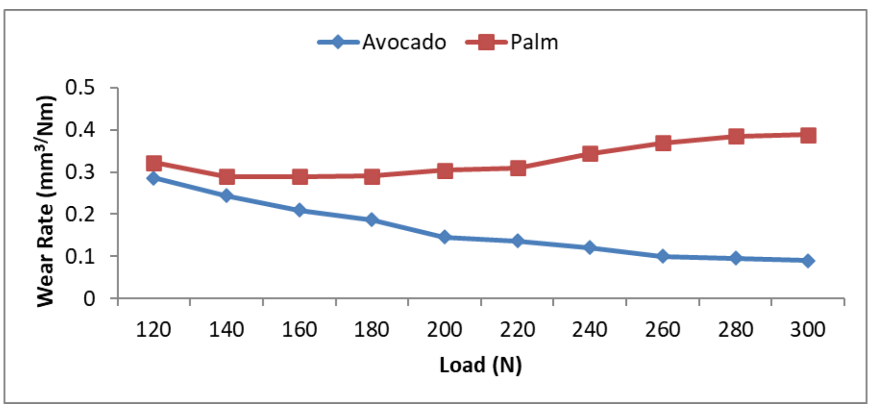


Figure 5: Wear rate of the modified vegetable oils and mineral oil under boundary lubrication condition.

It can be observed from Figures 4 and 5 that amongst the oils, the avocado seed oil proved the best tribological responses (friction and wear) as compared to the palm oil under extreme pressure conditions (120 – 300) N. The excellent adsorption capacity of the organically modified vegetable on disc specimen AISI 8620 is suspected to be associated with large microporous structure, large specific surface area, high hydrophobicity and its chemical compositions (Bahari et al., 2017a). The excellent tribological responses (friction and wear) of the surfaces lubricated with the avocado seed oil is in agreement with its superior thermal physicochemical properties of pour and flash points see Table 1, (physicochemical properties of the modified vegetable oils). Obviously, lubrication regime of the contacts is boundary; this is validated in Figure 4 where the COF value decreased from 0.18 $\pm$0.01 to 0.05 $\pm$0.01 for avocado seed oil which is within the boundary lubrication regime from the stribeck curve (Ajayi et al., 2015).

3.4 Adsorption of the Unmodified and Modified Vegetable Oils on Steel Contacting Surfaces

The results of the adsorption of unmodified biodegradable oils on steel contacting surfaces as calculated using Langmuir Adsorption model is illustrated in Table 2, with avocado seed oil having -0.45 KJ/mol and 0.49 KJ/mol for 300^0 C and 350^0 C respectively. Palm oil 0.50 KJ/mol and 0.55 KJ/mol. For the modified vegetable oils the result is presented in Table 3. Evidently, avocado seed oil has the most negative adsorption of -16.94 KJ/mol at 300^0 C and -18.41 KJ/mol at 350^0 C, followed by palm with -15.90 KJ/mol at 300^0 C and -17.26 KJ/mol at 350^0 C respectively.

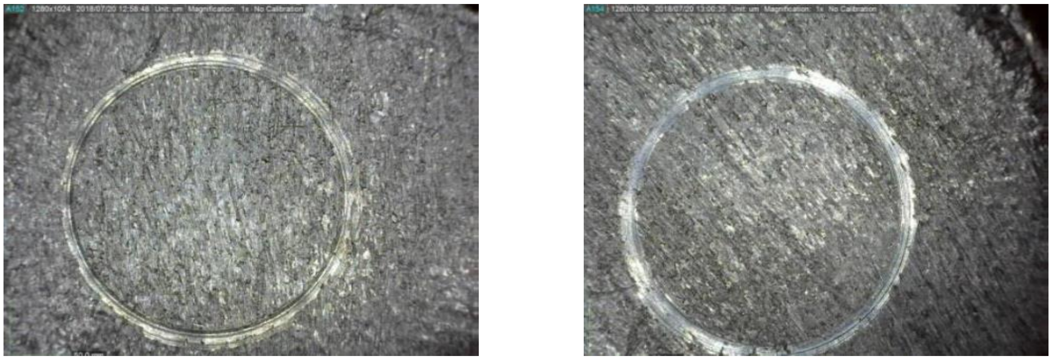
Table 2: Adsorption of the unmodified vegetable oils on steel contacting surfaces under boundary lubrication condition.

S/N	Base Oil	COF_{Unm}	K_0	$\Delta G^0{}_{300^0C}$ KJ/mol	$\Delta G^0{}_{350^0C}$ KJ/mol
1	Avocado	0.090	1.1	-0.45	-0.49
2	Palm	0.114	0.9	+0.50	+0.55

Table 3: Adsorption of the modified vegetable oils on steel contacting surfaces under boundary lubrication condition.

S/N	Base Oil	K_0	$\Delta G^0{}_{300^0C}$ KJ/mol	$\Delta G^0{}_{350^0C}$ KJ/mol
1	Avocado	35	-16.94	-18.41
2	Palm	28	-15.90	-17.26

Avocado seed oils highest negative Gibbs adsorption energy ΔG^0 value is an indication that the oil adsorbed better than the palm oil. This is a strong indication that the additive enhanced the anti-oxidation properties of the oil. The increase in adsorption with temperature is an indication that the adsorption type of oils on the steel surfaces is chemical adsorption (chemisorption). Also, the iron-oxygen large negative bond of the heat of formation indicates that the iron is easily oxidized. This suggests that oxygen played an important role in this reaction system. Oxygen provides low energy paths for the reaction to proceed. This also implies fatty acid oxidation products such as acids, peroxides, and oxygenated polar impurities will react with iron relatively easy. The formation of such organometallic compounds in boundary lubrication probably leads to “boundary film” reducing wear. The positive adsorption value of palm oil in its unmodified form, is a proof that the oil did not adsorb, while that of avocado showing slight negative value had little adsorption.



Avocado seed oil Palm oil

Figure 6: Worn surface topography (Scale: 1:2.5).

4.0 CONCLUSIONS

Studies were performed to investigate the adsorption of modified biodegradable oils on steel contacting surfaces under boundary lubrication regime. Conclusions can be drawn:

1. Palm oil has superior lubricating properties over the avocado seed oil under moderate loading conditions of (2 – 20) N, but at extreme conditions (120 – 300) N, the avocado seed oil proved superiority. This is a strong indication that the anti-oxidation effect of the organic additive is more pronounced in avocado seed oil under (200 – 300) N loading conditions.
2. The formation of a protective lubricating tribochemical film on the steel contacting surfaces that are required to reduce friction requires elevated temperature conditions.
3. The decrease in Langmuir value of the modified vegetable oils from -16.94 kJ/mol to -18.41 kJ/mol at elevated temperatures of (300 – 350)^{0}C with COF values of 0.06 and above which from the stribeck curve falls under boundary lubrication regime, suggests the lubricant did not fail even when the friction was on the extreme (boundary lubrication condition). So, the stribeck curve was used to determine the lubrication regime.
4. This study could not investigate the tribochemistry of the wear debris; thus, one of its limitations.

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