Asian Journal of Chemistry; Vol. 33, No. 6 (2021), 1363-1369

ASIAN JOURNAL OF CHEMISTRY

https://doi.org/10.14233/ajchem.2021.23209

Lubricity, Tribological and Rheological Properties of Green Ester Oil Prepared from Bio-Based Azelaic Acid

Waled Abdo Ahmed[1], Nadia Salih[2,*] and Jumat Salimon[2]

[1]Department of Chemistry, Faculty of Education, Thamar University, Thamar, Yemen
[2]Department of Chemical Sciences, Faculty of Science and Technology, Universiti Kebangsaan Malaysia, 43600 Bangi, Selangor, Malaysia

*Corresponding author: E-mail: nadiaalnami@hotmail.com

Received: 16 March 2021; *Accepted*: 16 April 2021; *Published online*: 5 June 2021; AJC-20369

Plant oil derived compounds have been used as the raw material for the synthesis of green biolubricant. Azelaic acid derived from oleic acid is one of the interesting compounds. The synthesis of azelate esters oil based synthetic green biolubricants was carried out. The esterification process of azelaic acid with variety type of alcohols was catalyzed by concentrated H_2SO_4. The yields percentages of azelate esters oil produced were varied depending their overall molecular structure. The azelate esters oil properties were evaluated for their suitability as a biolubricant. The results showed that the linear azelate esters oil showed high pour point values in a range of 15 to 5 ºC for di-decyl azelate, di-octyl azelate and di-hexyl azelate respectively. On the other hands, the branch azelate esters oil showed very low pour point of -70, -58 and -50 ºC for di-2-butyloctyl azelate, di-2-ethylhexanol azelate and di-2-ethylbutyl azelate, respectively. Moreover, the linear azelate esters oil gave high values of flash point, viscosity index and oxidative stability, and they were slightly affected by branching molecule structure. The tribological and rheological properties of high molecular azelate esters oil were also performed. Subsequently, most of them were classified as non-Newtonian fluids and having boundary lubrication properties with low friction coefficient. The branch azelate esters oil is plausible to be used as green biolubricants in many applications including automotive, marine engine oils, compressor oils, hydraulic fluids, gear oils and industrial biolubricants.

Keywords: Azelate esters oil, Green biolubricant, Pour point, Non-Newtonian fluids.

INTRODUCTION

Biolubricants are an importance compounds which are used to reduce the abrasiveness of moving metal surface in internal or external parts of the metal engine. By performing the protective layers on the metal surface, it can improve the smoothness and the motions of two moving metal surfaces without destruction. Furthermore, the smoothness of moving surfaces can be improved by reducing friction or steadily friction [1]. In view of vehicle engines, lubricating fluid is the most important form of engine lubricants and generally contain about 70-99% of base oil and 1-30% oil additives. Up to date, more than 95% of biolubricants used are petroleum based [2]. This is due to the fact that the petroleum based biolubricants show an excellent properties in terms of its functions. However, most of the petroleum based biolubricants are toxic to the environment and hard to be removed naturally. An alternative biolubricants produced from plant oils based are very important to be highlighted and introduced to the public. Plant oils based biolubricants are renewable sources, biodegradable in nature and non-toxic to the environment. Their low volatility behaviour is another advantage of its good lubrication and good anti-corrosion characteristics. However, plant oils based biolubricants naturally have two major advantages from low oxidative stability and high pour point. These are due to the presence of unsaturated fatty acids containing one or more double bonds that are likely prone to be oxidized. Poor oxidative stability of plant oils based biolubricant is also due to the rearrangement of β-hydrogen atoms in glycerol molecules to establish more stable molecule in case of free radical or carbocation being formed [3]. In addition plant oils based biolubricant has poor low temperature properties or pour point at low temperature condition due to the naturally presence of saturated fatty acids. Therefore, the chemical modification of the plant oil esters or plant oil based synthetic

This is an open access journal, and articles are distributed under the terms of the Attribution 4.0 International (CC BY 4.0) License. This license lets others distribute, remix, tweak, and build upon your work, even commercially, as long as they credit the author for the original creation. You must give appropriate credit, provide a link to the license, and indicate if changes were made.

esters production is another alternative research to be addressed in order to overcome this problem.

Plant oil based green synthetic esters have been synthesized to form polyol esters oil that contain more than one ester functional groups. It's can be di-, tri-, tetra-, penta-, hexa-, *etc*. esters group in polyol ester oil. Polyol ester oil has been widely used as the base stock of biolubricants. It can be produced through several processes such as the process of esterification and trans-esterification. The process of esterification is carried out between fatty acids and polyhydric alcohols, while the process of trans-esterification is carried out between methyl ester fatty acids (FAME) and polyhydric alcohols. There are several polyhydric alcohols have been used in the production of polyol esters oil for biolubricant. Among them are pentaeritritol, neopentylglycol, trimethylolpropane, di-trimethylolpropane and di-pentaeritritol [4]. Polyol esters oil are preferred for biolubricants used due to its capability to overcome the two main drawbacks of plant oil biolubricant. The main drawback is dealing with the oxidative stability which could be improved by reducing the double bonds in the molecule and the absence of a glycerol molecule. It's also improve the low temperatures performance by reducing the uniformity structural of the biolubricant oil through the production of branched esters. In general, synthetic esters are favourable to be used due to the advantages of low volatility due to the strong ester dipole moment to lubrication. The polarity of the ester group improve the attachment of biolubricant to the metal surface and high stability against temperature.

Green polyol diester oil based on the plant oil dicarboxylic acid is another class of green polyol esters oil and has its own advantages in the lubrication. The most common plant oil based dicarboxcylic acid is azelaic acid which is produced from the oxidation of plant based oleic acid in the industry. It has the advantage in some lubrication properties due to its dumb-bells shape in their molecule configuration. Currently, the oil of green diesters are used in numerous applications, such as marine and automotive engine oils, hydraulic fluids, compressor oils, grease formulations and gear oils [5,6]. The excellent biodegradability and low toxicity of diesters provides additional benefits to their performance [6]. Pour point can be reduced by increasing the molecular branches with shorter chain fatty or the branched alcohol [7] while viscosity index (VI) can be increased by lengthening the chain acids/linear portion of diacid or alcohols [8]. In this study, green polyol diester oil of dicarboxcylic acid was synthesized and evaluated. To achieve a balance between those two main important lubrication properties in lubricating, azelaic acid (diacid with nine carbons) was chosen as the starting material in the esterification process with variety types of selected alcohols [9-11]. The synthesize green azelate esters oil *via* esterification reactions between azelaic acid with fifteenth different types of long chain and branched alcohols such as *n*-butanol, 2-butanol, 2-octanol, 2-hexanol, 2-decanol, 2-methyl-1-butanol, 2-ethyl-1-butanol and 2-ethyl-1-hexanol and 2-butyl-1-octanol were carried out. Lubrication characterizations *e.g*. oxidative stability (OS), viscosity index (VI), flash point (FP), pour point (PP), tribology and rheological properties of green azelate esters oil based biolubricant were tested and evaluated.

EXPERIMENTAL

Azelaic acid (99%) was obtained from Fisher Scientific. All other chemicals and reagents such as oleyl alcohol, 2-ethyl-1-hexanol, 2-ethyl-1-butanol, 2-butyl-1-octanol, 2-decanol, decanol, *n*-butanol, 2-octanol and *sec*-butanol were obtained from Sigma-Aldrich, USA. They were used directly without further purification and the organic extracts were dried using anhydrous sodium sulphate (Aldrich Chemicals).

Synthesis of diester: The oil of green diesters was produced through acid catalytic esterification by employing the slightly modified setup of Dean Stark distillation reported by Bahadi *et al*. [3]. Initially, a mixture of alcohol and azelaic acid (mole ratio 2:1) was placed in a three-necked round-bottom flask. As a reaction media, 40-50 mL of toluene was added. This reaction mixture was heated in an oil bath to a desired reaction temperature of 120-130 ºC with continuous stirring. As a catalyst, a certain quantity of 2% H_2SO_4 (based on the weight of azelaic acid) was added slowly to the reaction container. The reaction was conducted for 4 h. During the reaction, water produced as a byproduct was continuously separated from the reaction medium through distillation. Reaction progress was studied by determining the amount of water separated. In a dropping funnel, the removed water was collected. When no water could be further removed from the reaction container, the reaction terminated. The obtained crude diester oil was cooled down in air.

Purification of diester: After the termination of reaction, the crude product was placed in a round-bottom flask. First, from the reaction product, toluene was removed at 100 ºC using a rotary evaporator. The product was then cooled down at room temperature. Subsequently, the product was transferred in a separating funnel. Diethyl ether (50 mL) was added to this funnel and saturated $NaHCO_3$ (10 mL) was added to the resulting solution to remove and neutralize the unreacted azelaic acid. The addition step was iterated thrice. Subsequently, 10 mL of saturated NaCl solution was added to prevent the formation of any emulsion. In each step, aqueous layer acquired was decanted and the layer of diethyl ether was dried on adequate anhydrous sodium sulphate. The hydrated Na_2SO_4 subsequently was filtered. To eliminate decomposed materials generated during the reaction, this dried product was passed through the column packed having silica gel 60. For solvent removal, this product was subjected to rotary evaporation. Furthermore, the trace solvent was eliminated in the presence of nitrogen gas. Through distillation, the excess alcohol present in the mixture was separated. The percentage yield of green diesters oil was estimated.

Characterization: To identify the functional groups present in green diesters oil, Fourier transforms infrared spectroscopy (FTIR) analysis was carried out as reported by Aigbodion & Bakare [12]. The FTIR of the products was recorded on Perkin-Elmer Spectrum GX spectrophotometer in the range 4000-400 cm^{-1}. A very thin film of product was covered on NaCl cells (25 mm i.d × 4 mm thickness) and used for the analysis. ^1H & ^{13}C NMR analysis was performed with model JEOL FCP 400 MHz with solvent $CDCl_3$. ^1H & ^{13}C NMR of diesters oil were recorded on Bruker 300 NMR spectrophotometer 20 mg of sample was dissolved in 1 mL of $CDCl_3$ and introduced into the NMR tube.

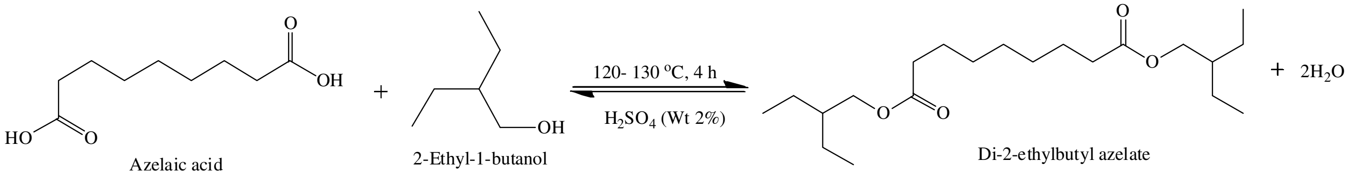

Fig. 1. Esterification reaction between azelaic acid and 2-ethyl-1-butanol to form di-2-ethylbutyl azelate

Lubricity analysis: The lubrication properties of the green diesters oil were evaluated by measuring the pour point, flash point, kinematic viscosity, viscosity index and oxidative stability values. All these measurements were performed according to the ASTM method [13]. Triplicate measurements were made and reported the average values with the standard deviation (±SD).

Analysis of tribology and rheology: By following the ASTM D4172-94 method [14], the tribological properties of the oil of green diesters were analyzed. These properties were discussed on the basis of Stribeck curves obtained at 40 and 100 °C [15]. At 40 and 100 °C in 10^{-1}–10^{3} mm/s, the sliding speed was varied. The rheological properties of diester oil were investigated using a rheometer having a plate and cone geometry [16]. CP 25-2 was the cone spindle employed and its diameter was 0.051 mm. At 25 °C, the shear rate was 0-100 s^{-1}.

RESULTS AND DISCUSSION

Chemical reaction and yield %: The esterification chemical reaction between azelaic acid and 2-ethyl-1-butanol to form di-2-ethylbutyl azelate (D2EBAz), as an example is shown in Fig. 1. In general, the yield percentages of azelate esters oil produced were different depending on type of alcohol used both short or long carbon chain alcohols and branch carbon chain alcohols (Fig. 2). The results show that branch alcohol has a negative effect on the ester yield percentage and this is attributed to the hindrance steric effect [17]. The ester oil yield percentages also depends on the structure and the hydroxyl alcohol functional group (-OH) position of the alcohols used. The esterification yield was increased when the carbon number of primary alcohol increase. On the other hand, it was decreased when the secondary alcohols were used due to steric hindrance factor. This observation is proven by the ester oil of didecyl azelate (DDAz) with the highest yield percentage of 97% whereas ester oil of di-2-butyloctyl azelate (D2BOAz) gave the lowest yield percentage of 83% among the synthesized green azelate esters oil.

Characterization of ester functional group: The FTIR results showed that the absorption bands of aliphatic carboxylic and hydroxyl alcohol groups at 1697 and 3450 cm^{-1}, respectively, were not observed. This observation suggests that esterification reaction was successfully completed. The absorption bands of C=O (ester) was at the range of 1739-1730 cm^{-1}, C-O (ester) stretching vibrations was at 1244-1172 cm^{-1} and C-O-C (ester) bands at 1101-965 cm^{-1} for all synthesized diester (Fig. 3).

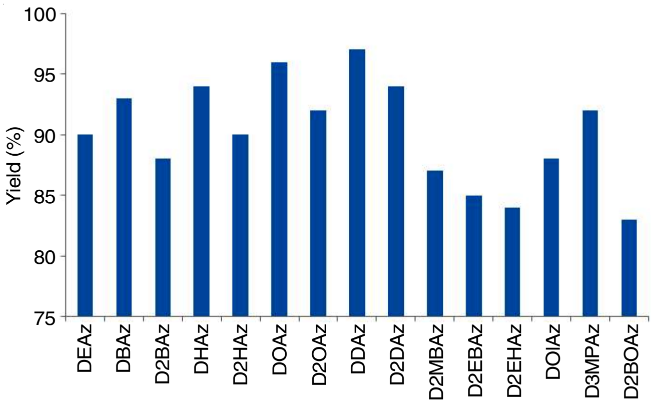

Fig. 2. Yield percentage of azelate esters; Note: DEAz: di-ethyl azelate, DBAz: di-butyl azelate, D2BAz: di-2-butyl azelate, DHAz: di-hexyl azelate, D2HAz: di-2-hexyl azelate DOAz: di-octyl azelate, D2OAz: di-2-octyl azelate, DDAz: di-decyl azelate, D2DAz: di-2-decyl azelate, D2MBAz: di-2-methylbutyl azelate, D2EBAz: di-2-ethylbutyl azelate, D2EHAz: di-2-ethylhexyl azelate, DOlAz: di-oleyl azelate, D3MPAz: di-3-methylpentyl azelate, D2BOAz: di-2-butyloctyl azelate

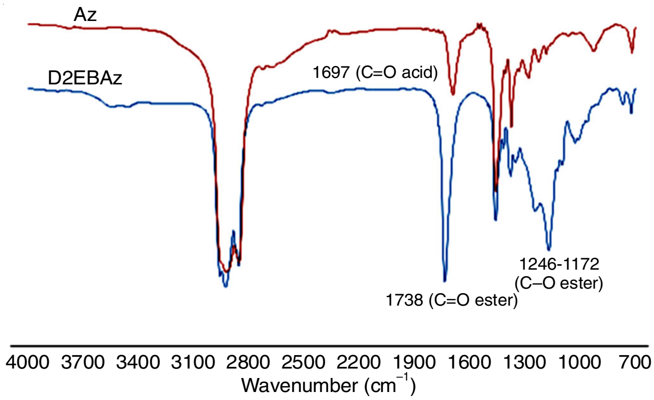

Fig. 3. FTIR spectra of azelaic acid (Az) and di-2-ethylbutyl azelate (D2EBAz)

^{1}H NMR analysis: The frequency at which any given hydrogen atom in an organic compound resonates is strongly dependent on its precise molecular environment. The ^{1}H NMR results show the main signals of assignments in tested diesters. The distinguishable peaks were appeared at 3.89 to 4.01 ppm for RCOO-CH_2 ester, 2.22 ppm to 2.32 for H_2C-COOR and -CH (3° aliphatic), 1.57 to 1.62 ppm for -CH_2-CH_2-COOR, 1.24-1.30 ppm for -CH_2- (saturated alkyl chain) and 0.80 to 0.89 ppm for - CH_3 (terminal methyl in alkyl chain) (Fig. 4).

^{13}C NMR analysis: The ^{13}C NMR results show the main signals of assignment of tested diesters. The appearance of carbon atom of the carbonyl group of ester clearly appeared at the range of 172 to 174 ppm while the others ^{13}C chemical shifts appeared at 65.25 to 69.87 ppm for (O-C) ester, 31.55 to 31.98 ppm for (-CH_2-C=O) ester, 37.02 to 37.69 ppm for -CH_3 aliphatic, 22.69 to 29.68 ppm for -CH_2- (saturated alkyl chain) and 14.01 to 16.03 ppm for -CH_3 (terminal methyl in alkyl chain) as shown in Fig. 5. In general, the NMR results of tested diesters confirmed

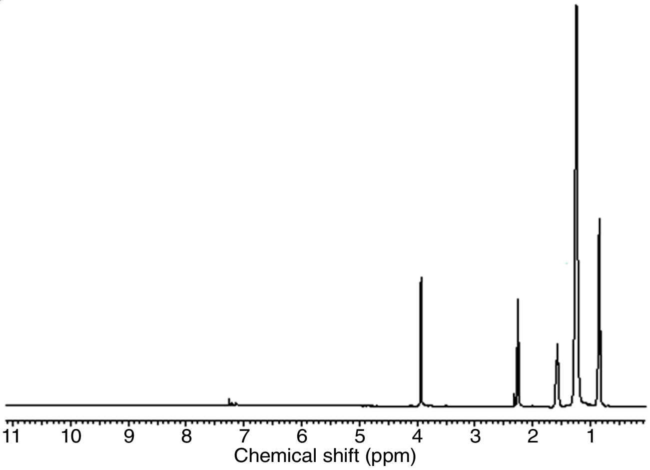

Fig. 4. ^{1}H NMR spectrum of di-2-butyloctyl azelate (D2EBAz)

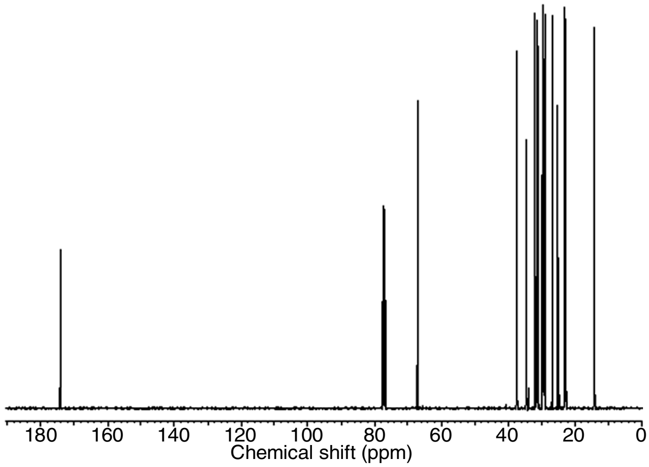

Fig. 5. ^{13}C NMR spectrum of di-2-butyloctyl azelate (D2EBAz)

the assignments chemical shifts of ester groups which are important signals. These signals verify that the esterfication reaction was successfully performed with good purity.

Lubricity characterization: The lubrication properties including viscosity index, viscosity, flash point, oxidative stability and pour point of the green oil of azelate esters are presented in Table-1. The kinematic viscosity largely influences the biolubricant efficiency for lowering wear and friction. At 40 ºC, most of the oils of green azelaic esters exhibit viscosity of 5-38.9 cSt, which renders them suitable hydraulic fluids [7]. For some applications, including gear biolubricants, this viscosity is low. Thus, viscosity modifiers must be added to expand diester′s applications as biolubricants [18,19]. At 40 and 100 ºC, the existence of branch groups affected kinematic viscosity. The kinematic viscosity is high and low for the linear and branch carbon groups, respectively, in diesters.

The viscosity index provides the influence of temperature on viscosity [20]. Low and high viscosity indexes indicate a large and small viscosity change, respectively, with temperature over a wide range. Viscosity index reveals how the viscosity of biolubricants varies with changes in operating machine temperatures. The applications of many biolubricant require performing under various conditions. The automotive biolubricant must minimize friction among engine components when the engine is running at up to 200 °C and started when it is relatively colder than operating temperatures [21]. When an engine is cold, biolubricants with high viscosity indexes prevent excessive thickening and promote prompt circulation and rapid starting. When a motor is hot, biolubricants prevent excessive thinning, thereby preventing excessive oil consumption and providing complete lubrication. The viscosity index of the oil of green azelate esters increased with an increase in molecular weights, the degree of branch in the alcohol and chain lengths (Table-1). The oils of liner green and branch azelate present viscosity index in 141-191 and 130-224, respectively. For instance, D2HAz and D2MBAz provided the maximum viscosity indexes among all the diesters at 224 and 222, respectively. Ester oils having a high viscosity indexes are obtained by properly selecting starting materials with linear structures [22]. High viscosity indexes are mainly advantageous because the base oil does not require viscosity enhancers at high temperatures to maintain its kinematic viscosity [23].

The most crucial low-temperature property of biolubricants is the pour point (PP), which represents temperature below the oil flow stops. The flow properties of oils of some green azelate esters having various alcohols with long chains are poor,

TABLE-1
LUBRICATION CHARACTERIZATION OF GREEN AZELATE ESTERS OIL

Diester	Kinematic viscosity at 40 °C (cSt)	Kinematic viscosity at 100 °C (cSt)	Viscosity index	Flash point (°C)	Pour point (°C)	Oxidative stability (°C)
DEAz	3.7 ± 0.1	1.4 ± 0.1	169 ± 5	160 ± 3	-17 ± 1	188 ± 2
DBAz	5.8 ± 0.1	2.0 ± 0.0	141 ± 3	170 ± 2	-12 ± 3	176 ± 3
D2BAz	5.5 ± 0.2	1.8 ± 0.1	181 ± 3	165 ± 5	-30 ± 3	205 ± 3
DHAz	11.7 ± 0.2	3.2 ± 0.1	148 ± 5	190 ± 5	-5 ± 2	193 ± 4
D2HAz	7.3 ± 0.1	2.8 ± 0.2	224 ± 1	175 ± 4	-25 ± 2	209 ± 3
DOAz	15.1 ± 0.1	4.1 ± 0.2	177 ± 7	215 ± 3	10 ± 3	201 ± 3
D2OAz	14.6 ± 0.3	3.8 ± 0.1	165 ± 1	195 ± 5	-20 ± 1	195 ± 4
DDAz	18.9 ± 0.1	5.4 ± 0.1	191 ± 1	220 ± 5	15 ± 3	219 ± 2
D2DAz	14.9 ± 0.2	4.4 ± 0.1	207 ± 7	205 ± 3	-15 ± 2	206 ± 2
D2MBAz	7.0 ± 0.1	2.7 ± 0.2	222 ± 2	150 ± 5	-60 ± 5	213 ± 3
D2EBAz	8.5 ± 0.1	2.7 ± 0.1	164 ± 1	175 ± 6	-50 ± 5	205 ± 2
D2EHAz	13.0 ± 0.2	3.3 ± 0.1	130 ± 1	205 ± 5	-58 ± 2	200 ± 2
DOlAz	38.9 ± 0.3	9.9 ± 0.1	167 ± 3	275 ± 3	17 ± 2	173 ± 4
D3MPAz	8.8 ± 0.1	3.0 ± 0.1	201 ± 3	225 ± 3	-35 ± 2	200 ± 4
D2BOAz	22.4 ± 0.2	6.0 ± 0.1	175 ± 3	270 ± 4	-65 ± 3	195 ± 2

which limits their application as the oils of automotive engines at low operating temperature. However, branched diesters including D2EBAz, D2MBAz, D2BOAz and D2EHAz are the most effective for lowering the pour point at -50, -60, -65 and -58 ºC, respectively. Nagendramma & Kaul [24] found that branching in alcohol fragments results in considerably low pour point. This change in pour point revealed that the alkyl chain branching of an ester plays an important role in the pour point decrease. Cermak *et al*. [25] showed that the branched esters exhibit lower pour points than linear chain analogous esters because the existence of a large point of branching on the oil of green azelate diesters causes crystallization inhibition and the formations of steric barriers near individual molecules, which leads to lower pour points [26]. Rizvi [27] found that the ester pour points rely on the employed alcohols' weight and structure. At room temperature, the oils of all green azelate esters exhibit the liquid state. In dioleyl sebacate (DOlAz), the effect of used unsaturated alcohols was obvious at 17 ºC. In spite of long-chain carbons and considerable molecular weight, DOlAz exhibited slightly low pour point due to DOlAz unsaturation. These results agree with the work of Rizvi [27], which reported that unsaturated diesters lead to a decrease in pour points. The fluidity of dioctyl azelate (DOAz) and di-2-octyl azelate (D2OAz) of 10 and -20 ºC, respectively, was obtained at lower temperature because of the presence of branching than that of di-2-ethyl hexyl azelate (D2EHAz) (-58 ºC).

To determine the fire resistance and volatility of biolubricants, flash points are useful. Flash points are employed to estimate the storage and transportation temperature for biolubricants. To ensure minimum volatilization and safe operations, flash points must be high at highest operating temperatures. An effective flash point >300 ºC may be required in demanding applications, including aviation jet engines [28,29]. Flash point data indicated the possibility of presence of highly flammable and volatile materials in non-flammable and non-volatile materials. This mixture is ignited only when in the air, the fuel vapour concentration is > 1% or < 6% by volume [30]. Flash point is a crucial factor for determining the extent of efficiency of oils to serve as potential biolubricants. Flash point is employed as a distinct oil fuel characteristic and to describe the oils, which are normally not used as fuels. Flash points refer to the combustible and flammable oils. Various international standards are available for defining combustibility and flammability. However, most standards state that oils having a flash point of < 43 and > 43 ºC are flammable and volatile and combustible, respectively [31]. The finding showed that the flash point of the oil of azelate esters increased with an increase in the chain length of alcohols. The highest flash point was obtained for DOlAz at 275 ºC because of its high molecular weight. The lowest flash point was acquired for D2MBAz having a short-chain alcohol at 150 ºC. The isomer molecular structure can slightly affect flash point. Linear DHAz has the flash point of 190 ºC and both the small branch of D2HAz and large branch of D2EHAz have the flash point of 175 ºC.

The resistance of biolubricants towards molecular rearrangement or breakdown in the air environment at elevated temperatures is called oxidation stability. This stability is a crucial characteristic of oils, especially where extended life is required (*e.g.*, transformers, turbiness, heat transfer and hydraulic units). In this study, the fabricated oils of azelate esters were screened to estimate their oxidation stability through PDSC by determining temperatures at which the oxidation rate increased rapidly. The results show that most of the tested azelate esters oil having a good stability of oxidation. Those diesters oil differ in their oxidation stability depending on the chemical structure and branching of the hydrocarbon chain [32]. For example, D2EBAz with branch structure having oxidation stability at 205 ºC, while D2EHAz at 200 ºC and D2BOAz at 195 ºC, respectively. Kubouchi *et al*. [33] have also reported that the oxidation stability increased with increasing the chain length of the esterified acid and alcohol. In case of DOlAz, the oxidation stability was low (173 ºC) due to the presence of unsaturation in its molecule structure. In summary, the results showed that the branch and unsaturation functional groups presence in azelate esters oil has affected on lowering the pour point of the ester oil and had less effect on their flash point, viscosity indexes and oxidative stability.

Tribology and rheological properties: The polarity section at the biolubricant oil molecule increases its lubrication efficiency by reducing wear [31]. Due to this polarity properties, azelate esters oil base fluids have a high affinity for metal surfaces. This makes them act as a boundary biolubricants with low friction coefficient (μ) at low and high temperature. Tribological properties as friction coefficient value of selected azelate esters oil are shown in Table-2. Three azelate esters oil were tested for their friction coefficient. They had an impact on the values of the friction coefficient, where those values decreased with their high molecular weight and kinematic viscosity of azelate esters oil. The results showed that the friction coefficient was low at 40 and 100 ºC. The effect of molecular weight as well as the kinematic viscosity were clearly noted for DOlAz where its lowest friction coefficient value of 0.17 ± 0.02 and 0.19 ± 0.01 at 40 and 100 ºC, respectively. The tested azelate esters oil have recorded low friction coefficient in the range of below than 0.30 at 40 and 100 ºC. These results indicate of their good tribological properties quality even in the case of high temperature. In short, the results showed that the tested azelate esters oil were boundary biolubricants with low friction coefficient at 40 and 100 ºC. Fig. 6 shows graphical distribution of the friction coefficient values (μ) against sliding distance for D2EHAz, D2BOAz and DOlAz, respectively. Non-Newtonian and Newtonian properties used to understand handling, processing and storage for the hydraulic system design of biolubricants and liquid oils. The shear stress (τ) *versus* shear rate (γ) curves indicated that fluids can be classified into non-Newtonian and Newtonian fluids. The Newtonian fluids show constant viscosity after increasing the shear rate, and the non-

TABLE-2
FRICTION COEFFICIENT OF TESTED AZELATE ESTERS AT 40 AND 100 °C

Diester	Friction coefficient	
	40 °C	100 °C
D2EHAz	0.24 ± 0.02	0.30 ± 0.01
D2BOAz	0.21 ± 0.01	0.25 ± 0.01
DOlAz	0.17 ± 0.02	0.19 ± 0.01

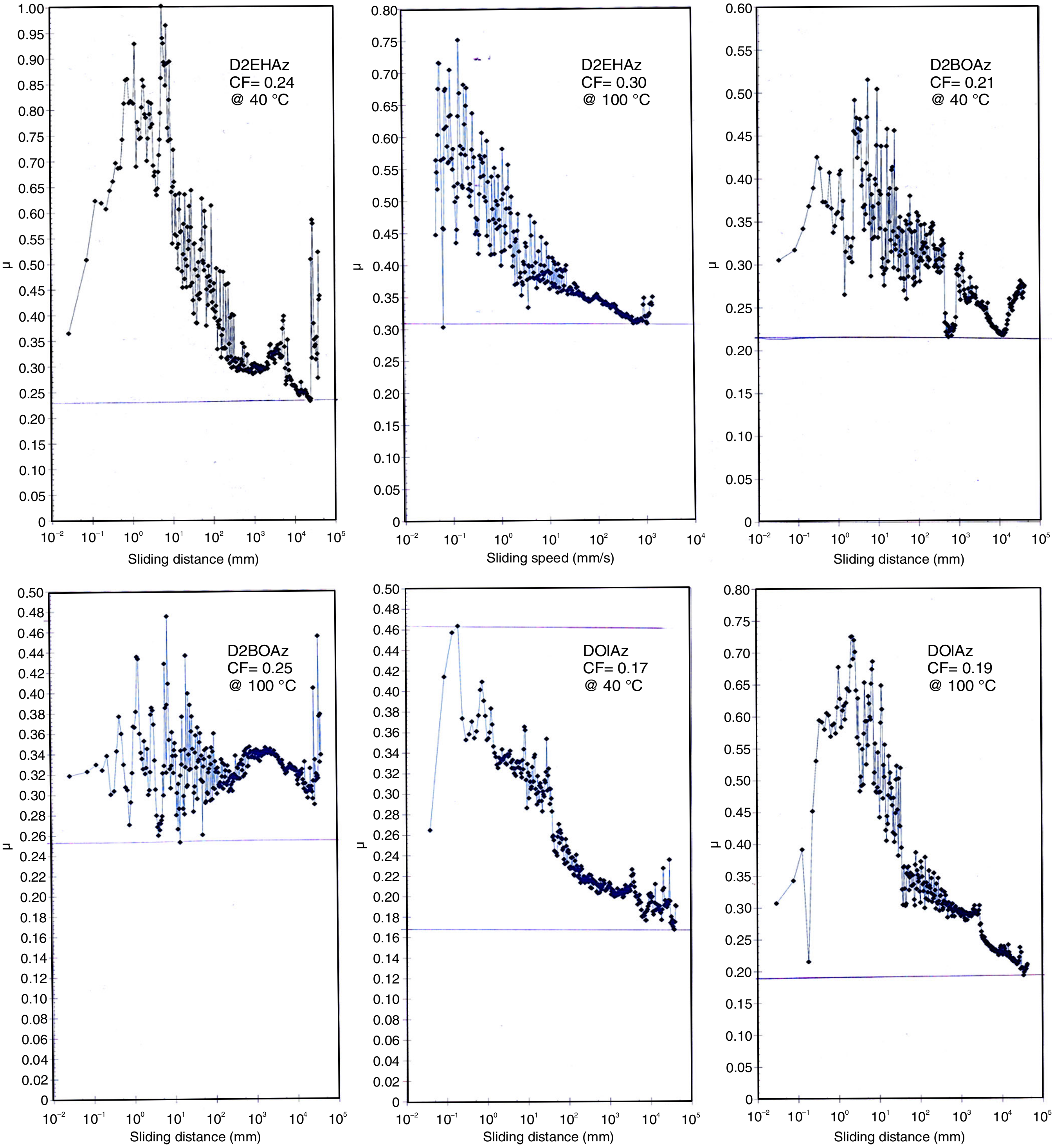

Fig. 6. Friction coefficient of D2EHAz, D2BOAz and DOlAz at 40 and 100 °C

Newtonian fluids do not exhibit constant viscosity even after increasing the shear rate [34]. According to the same plot curves presented Fig. 7, D2EHAz, D2BOAz and DOlAz (as a representation) are non-Newtonian fluids.

Conclusion

The azelate esters oil based synthetic green biolubricants was successfully carried out in high yields percentages especially for the linear long-chain azelate esters oil. The linear long chain of azelate esters oil enhance the kinematic viscosity, viscosity index, flash point and oxidative stability. On the other hand, azelate esters oil are significantly affected by branching molecule structure that leads to a good low pour point and slightly affected on other properties. It is recommended that the importance of selecting the raw materials in order to havea good lubricity and anti-wear properties of the azelate esters oil to be used as a synthetic green biolubricants. However, some of the azelate esters oil synthesized in this study have high potential and plausible to be used as a synthetic green biolubricants.

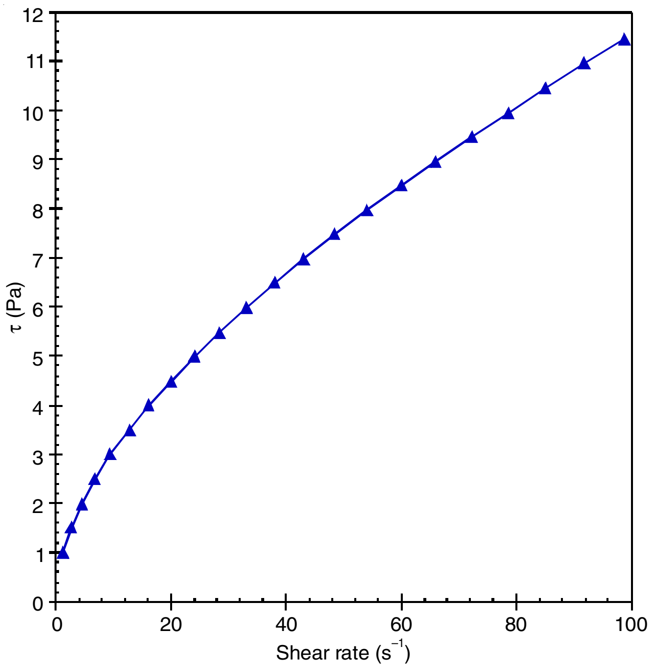

Fig. 7. Shear stress (τ) vs. shear rate (γ) plots of DOlAz

ACKNOWLEDGEMENTS

The authors thank Universiti Kebangsaan Malaysia for the project funding under university research grants no. DPP-2014-058, GGPM-2014-033 and the Department of Chemical Sciences, Faculty of Science and Technology, Universiti Kebangsaan Malaysia, Selangor, Malaysia for their support and encouragement.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

REFERENCES

1. D. Gropper, L. Wang and T.J. Harvey, *Tribol. Int.*, **94**, 509 (2016); https://doi.org/10.1016/j.triboint.2015.10.009
2. N. Salih and J. Salimon, *Biointerface Res. Appl. Chem.*, **11**, 13303 (2021); https://doi.org/10.33263/BRIAC115.1330313327
3. M. Bahadi, M.F. Yusoff, J. Salimon and D. Derawi, *Ind. Crops Prod.*, **139**, 11452 (2019); https://doi.org/10.1016/j.indcrop.2019.06.015
4. T.M. Panchal, A. Patel, D.D. Chauhan, M. Thomas and J.V. Patel, *Renew. Sustain. Energy Rev*., **70**, 65 (2017); https://doi.org/10.1016/j.rser.2016.11.105
5. S. Samidin, N. Salih and J. Salimon, *Biointerface Res. Appl. Chem.*, **11**, 13638 (2021); https://doi.org/10.33263/BRIAC115.1363813651
6. N. Salih and J. Salimon, *Biointerface Res. Appl. Chem.*, **12**, 1185 (2022); https://doi.org/10.33263/BRIAC121.11851207
7. A. Sagan, A. Blicharz-Kania, M. Szmigielski, D. Andrejko, P. Sobczak, K. Zawiœlak and A. Starek, *Sustainability*, **11**, 5638 (2019); https://doi.org/10.3390/su11205638
8. L.K. dos Santos, R.R. Hatanaka, J.E. de Oliveira and D.L. Flumignan, *Renew. Energy*, **130**, 633 (2019); https://doi.org/10.1016/j.renene.2018.06.102
9. N.M. Nor, N. Salih and J. Salimon, *Hem. Ind.* **75**, 117 (2021); https://doi.org/10.2298/HEMIND200809009N
10. M. Shahabuddin, M. Mofijur, M.A. Kalam and H.H. Masjuki, *Tribol. Ind.*, **42**, 41 (2020); https://doi.org/10.24874/ti.2020.42.01.04
11. J.A. Cecilia, D. Ballesteros Plata, R.M. Alves Saboya, F.M. Tavares de Luna, C.L. Cavalcante Jr. and E. Rodríguez-Castellón, *Processes*, **8**, 257 (2020); https://doi.org/10.3390/pr8030257
12. A.I. Aigbodion and I.O. Bakare, *J. Am. Oil Chem. Soc.*, **82**, 465 (2005); https://doi.org/10.1007/s11746-005-1095-0
13. R.A. Nadkarni, Guide to ASTM Test Methods for the Analysis of Petroleum Products and Lubricants, ASTM International, USA, Ed.: 2 (2004).
14. ASTM, Standard Test Method for Wear Preventive Characteristics of Lubricating Fluid, (Four-Ball Method), West Conshohocken, PA, USA (2011).
15. F.J. Owuna, *Egyptian J. Pet.*, **29**, 251 (2020); https://doi.org/10.1016/j.ejpe.2020.09.003
16. D.R. Moreira, P.O.B. Chaves, E.N. Ferreira, T.B.M.G. Arruda, F.E.A. Rodrigues, J.F.C. Neto, C.L. Petzhold, M.E. Maier and N.M.P.S. Ricardo, *Ind. Crops Prod.*, **158**, 112937 (2020); https://doi.org/10.1016/j.indcrop.2020.112937
17. A. Kushairi, M. Ong-Abdullah, B. Nambiappan, E. Hishamuddin, M.N.I.Z. Bidin, R. Ghazali, V. Subramaniam, S. Sundram and G.K.A. Parveez, *J. Oil Palm Res.*, **32**, 159 (2020); https://doi.org/10.21894/jopr.2019.0026
18. J. Nie, J. Shen, Y.Y. Shim, T.J. Tse and M.J.T. Reaney, *Eur. J. Lipid Sci. Technol.*, **122**, 1900207 (2020); https://doi.org/10.1002/ejlt.201900207
19. Djomdi, M.T. Leku, D. Djoulde, C. Delattre and P. Michaud, *Bioengineering*, **7**, 41 (2020); https://doi.org/10.3390/bioengineering7020041
20. G. Karmakar, P. Ghosh and B.K. Sharma, *Lubricants*, **5**, 44 (2017); https://doi.org/10.3390/lubricants5040044.
21. A.N. Afifah, S. Syahrullail, N.I. Wan Azlee, N.A. Che Sidik, W.J. Yahya and E. Abd Rahim, *Biochem. Eng. J.*, **148**, 178 (2019); https://doi.org/10.1016/j.bej.2019.05.009
22. Z. Xiong, F. Fu, X. Li and Y. Li, *Petroleum*, **5**, 77 (2019).
23. N.M. Nor, D. Derawi and J. Salimon, *Malaysian J. Chem.*, **21**, 28 (2019).
24. P. Nagendramma and S. Kaul, *Renew. Sustain. Energy Rev.*, **16**, 764 (2012); https://doi.org/10.1016/j.rser.2011.09.002
25. S.C. Cermak, K.B. Brandon and T.A. Isbell, *Ind. Crops Prod.*, **23**, 54 (2006); https://doi.org/10.1016/j.indcrop.2005.04.001
26. O. Ocholi, M. Menkiti, M. Auta and I. Ezemagu, *Egypt J. Pet.*, **27**, 265 (2018); https://doi.org/10.1016/j.ejpe.2017.04.001
27. Q.A. Rizvi, Comprehensive Review of Lubricant Chemistry, Technology, Selection and Design, USA, ASTM International (2008).
28. F.J. Owuna, M.U. Dabai, M.A. Sokoto, M.S. Dangoggo, B.U. Bagudo, U.A. Birnin-Yauri, L.G. Hassan, I. Sada, A.L. Abubakar and M.S. Jibrin, *Egyptian J. Pet.*, **29**, 75 (2020); https://doi.org/10.1016/j.ejpe.2019.11.004
29. S. Qiao, Y. Shi, X. Wang, Z. Lin and Y. Jiang, *Energy Fuels*, **31**, 7185 (2017); https://doi.org/10.1021/acs.energyfuels.7b00876
30. T.V.V.L.N. Rao, A.M.A. Rani, M. Awang and M. Baharom, *J. Tribol.*, **18**, 40 (2018).
31. C.J. Reeves, A. Siddaiah and P.L. Menezes, *J. Bio. Tribo. Corros.*, **3**, 11 (2017); https://doi.org/10.1007/s40735-016-0069-5
32. M. Bahadi, N. Salih and J. Salimon, *Biointerface Res. Appl. Chem.*, **11**, 14359 (2021); https://doi.org/10.33263/BRIAC116.1435914371
33. H. Kubouchi, H. Kai, K. Miyashita and K. Matsuda, *J. Am. Oil Chem. Soc.*, **79**, 567 (2002); https://doi.org/10.1007/s11746-002-0523-5
34. É.C.G. Aguieiras, E.D.C. Cavalcanti, P.R. da Silva, V.F. Soares, R. Fernandez-Lafuente, C.L. Bessa Assunção, J.A.C. da Silva and D.M.G. Freire, *Renew. Energy*, **148**, 689 (2020); https://doi.org/10.1016/j.renene.2019.10.156