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Analysis Performance of Modified Tamanu Oil Enhanced with Additives as Potential Green Alternative in Metalworking Fluids

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Abstract: Mineral oils have long been utilized in industries as machining lubricants, which contributed to their depletion and hick in price and being non-biodegradable, harmful to the environment, and risk to health. Plant-based oil is more biodegradable, renewable, and environmentally friendly as a green alternative. However, in their crude state, plant-based oils are not up to par with the standard mineral oil used in lubrication in terms of high acidity, low-temperature performance, and oxidative instability. Further chemical modification and adding additives had to be made to improve the oil properties for industrial applications. This study focused on the performance of transesterification of Tamanu plant-based oil with Trimethylolpropane (MTO) and Pentaerythritol (MTOP), which mixed with 1% of Phosphonium Ionic Liquid (PIL) and 10% of Ammonium Ionic Liquid (AIL), producing a series of oil specimens; MTO, MTO+PIL1%, MTO+AIL%, MTOP, MTOP+PIL1%, and MTOP+AIL%. These samples are then subjected to physical analysis to determine the improvement of their properties in terms of kinematic viscosity and viscosity index as well as undergo a four-ball wear test to the determine the tribology aspects of the lubrication in terms of coefficient of frictions and wear scar diameter, in accordance with ASTM standard method. All the results were compared with commercial MWF which synthetic ester (SE) as reference oil. The result of viscosity index reveals that MTOP+PIL1% had the highest value of 178.76. MTO+AIL10% exhibits the lowest average COF (0.061) compared to other MTOs lubricants. Among MTOP, MTOP+PIL1% also had the lowest average COF which is 0.082. The addition of PIL1% to MTO and MTOP lowered the average scar diameter, 730.77 μm and 674.93μm respectively. With the enhanced properties from chemical modification and additives, Tamanu oil can be proposed as a green alternative for developing the metalworking fluid industries in the future.

Keywords: Modified Tamanu oil, additives, ionic liquids, physicochemical properties, tribology

1. Introduction

Cutting fluid or lubricant is used to reduce the friction and heat between surfaces in relative motion and improve the efficiency of manufacturing processes by conserving or enhancing the performance and lowering defects [1]. It can also decrease the energy used and increase the production output, as adequate lubrication can lessen the coolant required in adsorbing the heat generated from the friction during cutting processes. Thus less energy is needed, as observed by [2].

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The widely used lubricants are mineral-based oil or petroleum oil, which have been in demand since it was established to increase the efficiency in manufacturing processes. However recent years, there has been a significant concern in finding green alternatives to replace the mineral-based lubrication in metalworking fluid because of its cons that outweigh the pros in its application for manufacturing and production industries. Several articles have highlighted the high usage consumption of mineral oil negatively impacted the environment. Nowak et al.[3] had discussed the harmful impact petroleum oil lubricants had on the aquifer, soils, plants, and animals and the risk to human health.

Statistics indicate that more than 50% of the world's lubricants end up in the environment due to spills, drops, instability, maritime accidents, illegal discharge, and tanker mishaps [4-6]. In recent years, South Korea Korean Coast Guard has reported about 2300 litres oil spills approximately had been related to maritime accidents from 2018 to 2020, which adversely affected marine lives [7]. There are also high risks for the soil and water to be polluted caused by the process of failure lubrication, as the volatile lubricants could cloud the atmosphere [8]. The general population could be exposed to the toxicity of lubrication via inhalation and dermal contact, which is caused issues such as irritation of the eyes, skin, and respiratory tract. It could also be a possible etiological factor in skin cancer, asthma and dermatitis [9]. Besides that, mineral-based oil is non-renewable, and with its continued usage, had been a scarcity of resources which led to price fluctuations [10].

Several initiatives have been made toward a green alternative to using plant-based oil based on these environmental and health concerns and prices. The justification had been made based on the plant-based oil being highly biodegradable, less toxic, and renewable as well as non-reactant to organic chemicals [11-15] .

In this study, Tamanu plant-based or Calophyllum Inophyllum was chosen to be the primary oil analysis for its physicochemical and tribological properties due to its biodegradable, renewable, and sustainable [16].Tamanu is a nonedible plant, which is a good choice in replacing lubrication oil sources from an edible plant. It eliminates the edible oil from being inconsumable [17,18]. Tamanu seeds also have high oil content and productivity compared to other plants such as sunflower and rapeseed, according to Kartika et al. [19].

However, crude Tamanu oil (CTO) cannot compete with the synthetic ester in the current market due to its natural plant state's properties such as high viscosity, poor low thermal-oxidative stability, and high acidity [20-23]. Further structural modification process, such as esterification processes and the addition of additives, is required to improve their current physical and chemical properties to use in industrial applications.

In this study, CTO is divided into two chemically modified samples: MTO and MTOP, with Trimethylolpropane (TMP) ester and Pentaerythritol (Penta) ester, respectively. Furthermore, MTO and MTOP are mixed with Phosphonium Ionic Liquid (PIL) and Ammonium Ionic Liquid (AIL), two low toxicity ionic liquids. These two ionic liquids are expected to improve the physicochemical and tribology performance of the Tamanu oil. Amiril et al.[24] had conducted research on modified Jatropha oil (MJO) with PIL and AIL additives, where the lubrication performance of MJO+AIL10% and MJO+PIL1% samples provide competitive lubrication performance to other lubricant samples used herein. The ionic liquids used had enhanced the corrosion inhibition, superior friction reduction, lower worn surface area, excellent surface finish, and increased tapping torque efficiency, which is also highlighted in previous research [25, 26]. The objective of this study is to compare the physical and tribological properties of crude state oil, modified oil, and added additive. The modification of crude oil and the addition of additives might improve the properties of the crude oil. A conclusion was drawn based on the comparison between the MTO, MTO-PIL1%, MTO-AIL10%, MTOP, MTOP-PIL1%, and MTOP-AIL10%.

2. Methodology

2.1 Sample Preparation

Material used - Crude Tamanu oil (CTO), are the primary sources for this research. CTO used in this research is produced in-house, where the fruits are procured locally in Pekan of Pahang, Malaysia. The fruits are then converted into crude oil, which undergoes esterification processes, with the final phase using Trimethylolpropane Ester (TMP-ester). After that, the final phase product will be known as modified Tamanu oil (MTO). The two MTO samples are then mixed with 1% of Phosphonium Ionic Liquid (PIL) and 10% of Ammonium Ionic Liquid (AIL). These ionic liquids are anticipated to enhance the properties and performance of the MTO and MTOP. Other chemicals used are included in the degumming process, such as phosphoric acid (H_3PO_4) and esterification process, such as sulphuric acid (H_2SO_4) with 96% purity, methanol (CH_3OH), Sodium Hydroxide (NaOH) with 99% purity and Sodium Methoxide (CH_3NaO).

Drying process - The drying process used in the mechanical extraction of the Calophyllum Inophyllum for this study used Method 1 [16]. This method has two drying processes: drying the fruits first, followed by the seeds. The drying manner for the fruits used 12 bulbs of a hundred-watt, with room temperature surrounding. The husk of the fruits turns from green to blackish, akin to rotting, which is then easy to crush using the fruit crushing machine to obtain the seeds. The seeds are then cut into four-part and dried in an oven when the texture of the seed will turn into a peanut-like surface. This heating process removes the water and rubber inside the kernel. The time taken for drying the result and seeds are one of a kind which can be 24 hours and 48 hours, respectively.

Oil extraction process- The dried fruits are crushed using a fruit crushing machine, which separates the seed from the exocarp, mesocarp and endocarp. The obtained seeds are then sliced before being dried. The preliminary texture of the seeds is soft before it emerges into a peanut-like surface because of the drying method. The dried seed is then fed into an electric-powered small automated oil press machine to extract the raw Tamanu oil from the seeds. The dried seeds are compressed and then filtered using Whatman filter paper at Grade 4 (20- 25 µm), resulting in oil and seeds residue output, as in Fig. 1.

(a)

(b)

Fig. 1 - (a) Oil extraction; (b) Crude Tamanu Oil (CTO)

Degumming process- The degumming process is the step to turn the raw Tamanu oil into crude oil or degummed oil. This process is necessary to separate the oils from resin and enhance the oxidization balance of the oil. The following process used the setup as illustrated in Fig. 2, where the weighted raw Tamanu oil is heated in a three-necked ball flask inside a water pot on top of Cole-Parmer advanced digital stirring hot plates and stirred using a magnetic stirrer. When the temperature of the water pot, consequently the oil inside the flask, reaches 60°C, it is then mixed with phosphoric acid (3%) with 1% of raw Tamanu oil. The reaction time for the degumming process starts at 1 hour.

After 1 hour, the oil is then transferred into a separating funnel. A slight separation occurred on the separating funnel for a few minutes when we needed to remove the bottom layer. The oil is then filtered using a clothed paper to separate the dirt residue. The filter oil is then dried in the vacuum oven at 110°C for 16 hours. The outcome of this process is degummed oil.

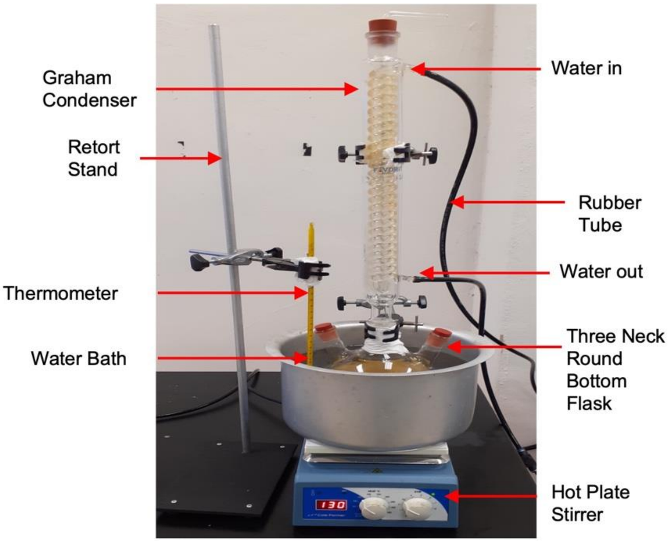

Fig. 2 - Degumming process setup

Esterification process- The esterification process is an essential method to lessen free fatty acid in crude Tamanu oils, as a high free fatty acid will cause an excessive soap formation and low yield of oil methyl ester. There are three phases in the esterification process: acid esterification, base transesterification, and transesterification. These processes used the same setup as the degumming process with a minor change and the difference in terms of catalyst used, reaction time, and drying time.

In acid esterification, the objective is to lower the acidity level in the oil, as a high acidity level in lubricants could cause oxidative instability and corrosion to the part machine. Using the same setup, the crude oil is mixed with 0.5% wt of sulphuric acid and methanol with a molar ratio to oil (24:1) when the temperature reaches 60°C. After the reaction time of 5 hours ends, the mixture is filtered inside a separating funnel to remove the methanol. After that, it dried in the vacuum at 80°C for overnight to further remove the excess methanol. Free fatty acid tests are then conducted on the esterified oil to determine its reduced acidity level.

In base esterification, the esterified oil is mixed with 1% wt of sodium hydroxide and methanol with a molar ratio to oil (6:1) at 60°C and set to finish in 3 hours. The filter process involved a washing process to remove the glyceride, where warm water (60°C - 70°C) is used to purify the ester with added three drops of phosphoric acid (86%) until the acidic range in ph7. The remaining alcohol and water are then dried in the oven at 110°C overnight. This process produces Fatty Acid Methyl Ester (FAME), also known as biodiesel.

In base transesterification, the setup is similar to previous processes, except the water bath is replaced with an oil bath, as this process involves 24 hours' reaction time at 120°C. The setup used in the previous cycle is also attached with a vacuum pressure set at 0.1 bar. In this study, two reactants are used to produce Tamanu-based ester, Trimethylolpropane (TMP) ester and Pentaerythritol (Penta) ester, respectively. For each process, 1% wt of sodium methoxide was added. The number of reactants in solid form is determined by the molar ratio of ester to oil (3.5:1). This process produced two chemically modified Tamanu oil, MTO and MTOP, using TMP-ester and Penta-ester, respectively.

Additive mixing- Ionic liquids involved in this process are Phosphonium Ionic Liquid (PIL) and Ammonium Ionic Liquid (AIL). These ionic liquids have low toxicity and are shown to reduce friction and wear. MTO and MTOP are heated on a hot plate for 70°C, then mixed with either 1% of PIL or 10% of AIL, then stirred for 30 min. The output of this process produced MTO+PIL1%, MTO+AIL10%, MTOP+PIL1%, and MTOP+AIL10%, which completed the samples needed to proceed with the following process of this study.

2.2 Physical Testing

Kinematic viscosity followed the ASTM D445 test methods, which determine both transparent and opaque liquid petroleum products. Both parameters determine the internal resistance flow of oil fluid internal under gravitational forces, as dynamic viscosity is a measure of force, while kinematic viscosity is a measure of velocity.

Viscosity index (VI) is used to determine the changes of fluid in viscosity relative to temperature change and used to characterize the viscosity-temperature behaviour of lubricating oils. The method used is ASTM D2270, which calculates the viscosity index based on kinematic viscosity at 40 °C and 100 °C.

2.3 Tribological Testing

This four-ball wear test was done according to the ASTM D4172 method, which investigates lubricant's efficiency in reducing wear and friction of the sliding surfaces. This test was carried out using a four-ball test rig (Ducom TR-30L) and four chrome steel balls (AISI 52100) with a diameter of 12.7 mm, as shown in Fig. 3. The top ball contains one ball bearing and is pressed using 392 N force and rotates at 1200 rpm for 60 minutes at a temperature of 75°C, while the ball pot has three balls coated in oil samples.

(a) (b)

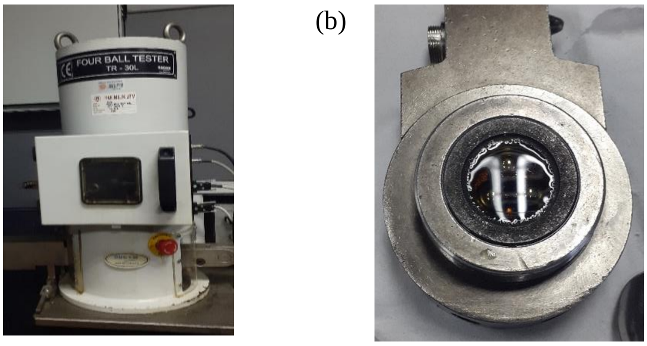

Fig. 3 - (a) Ducom RR-30L; (b) ball pot

Wear scar diameter (WSD) was observed to investigate the average scar on the three steel balls bearing inside the ball pot to dictate the efficiency of the oil used in reducing wear. This test is done using an optical microscope (Olympus BX51M Metallurgical Microscope) and a Scanning Electron Microscope machine (FEI, Quanta 450) to identify the topological surfaces of the worn area of the three balls bearing after the test.

3. Result and Discussion

3.1 Physical properties

Table 1 and Fig. 4 presents the kinematic viscosity and viscosity index of the samples. From the graph, SE had the highest value of kinematic viscosity at temperatures 40°C and 100°C with 21.5 mm²/s and 5.5 mm²/s compared to MTO and MTOP. This situation portrayed that SE was created with a variety of additive packages, including viscosity improvers, anti-wear additives, and corrosion inhibitors, resulting in an increase in SE viscosity [27]. MTO and MTOP has the lowest value of kinematic viscosity among MTOs and MTOPs itself at temperatures 40°C and 100°C with 16.856 mm²/s and 3.6801 mm²/s and 5.7315 mm²/s and 2.0186 mm²/s, respectively due to a change in the composition of MTO and MTOP upon a chemical alteration. The hydrogen bond's intermolecular interactions were lessened, lowering the oil's viscosity [28]. Kinematic viscosity plays a role in the use of crude oil in the machining process in terms of oil fluidity with the changes in temperatures. Moreover, the addition of additives improves the kinematic viscosity of MTO and MTOP. MTO+AIL10% shows the highest kinematic viscosity among MTOs with 20.539 mm²/s and 4.2439 mm²/s at 40°C and 100°C respectively. Among MTOPs, the addition of AIL10% additives improves the kinematic viscosity of MTOPs.

Furthermore, the result of the viscosity index shows that MTOP+PIL1% had the highest value of 178.76 compared to MTOPs and SE. Interestingly, MTO+PIL1% shows the highest viscosity index compared to MTOs and SE. PIL's properties include low volatility and an almost non-flammable nature. PIL's increased viscosity is also due to its high alkyl chain length [4]. In addition, the thermal conductivity of the additives plays important role, the thermal conductivity of PIL varies relatively small with temperature and pressure. Thus, it resulting in good thermal stability and maintains a high evaporation temperature. Therefore, it giving the lubricant combination a high VI value [29]. A lesser viscosity index means more viscosity affected by temperature changes

Table 1 - Physical properties of samples

Physical Properties	Unit	Temp.	SE	MTO	MTO+ PIL1%	MTO+ AIL10 %	MTOP	MTOP+ PIL1%	MTOP+ AIL10 %
Kinematic viscosity, v	mm^2/s	40°C	21.5	16.856	18.613	20.539	5.7315	5.9481	7.4930
		100°C	5.5	3.6701	3.9225	4.2439	2.0186	2.0943	2.4164
Viscosity Index, VI	-	-	116	101.53	104.26	111.44	170.02	178.76	159.85

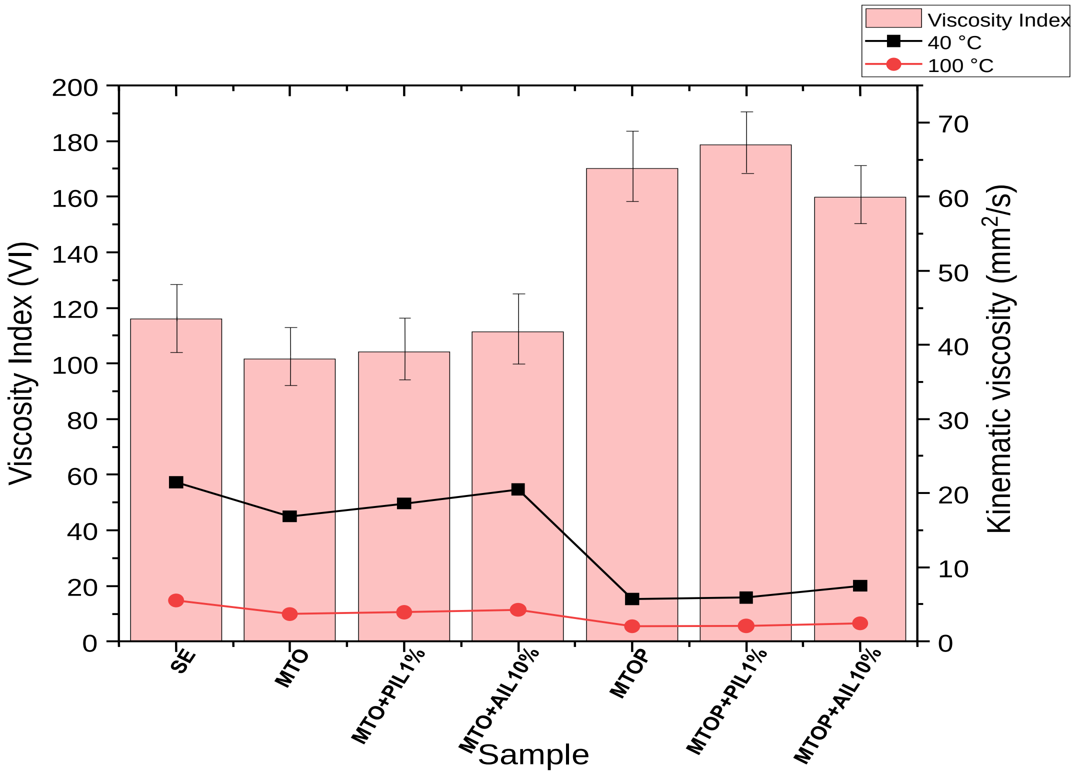

Fig. 4 - Graph of kinematic viscosity and viscosity index

3.2 Tribological properties

Table 2 and Fig. 5 shows the tribological behavior of COF and WSD of the studied samples. MTOs and MTOPs shows excellent tribological performance compared to SE. This phenomenon was because of MTOs and MTOPs formed a stable and strong protective lubrication film compared to SE. The average COF and WSD shows a reduction with the addition of additives It should be noted that the lowest coefficient of friction tends to have better lubrication in reducing wear. MTO+AIL10% exhibits the lowest average COF compared to other MTOs lubricants. As for MTOPs lubricant, MTOP+PIL1% had the lowest average COF which is 0.08207. Average mean wear scar diameter (WSD) of MTO+PIL1% reduced approximately 24% from average WSD of MTO. In addition, MTOP+PIL1% had great reduction in average wear scar diameter (WSD) approximately 30% compared to MTOP. The addition of PIL to MTO and MTOP shows an improvement of average WSD reduction of 24.8% and 30.6% compared to commercial reference oil SE. During the tribology test, PIL interacts with the base oil and the steel surface and form a protective boundary film that decreases friction and wear through chemical adsorption of the PIL anion on the steel surface [30].

Table 2 - Tribological properties of samples

Tribological Properties	Unit	SE	MTO	MTO+ PIL1%	MTO+ AIL10 %	MTOP	MTOP+ PIL1%	MTOP+ AIL10 %
Average Coefficient of Friction	-	0.112	0.083	0.064	0.061	0.078	0.082	0.088
Average Wear Scar Diameter	μm	971.86	965.57	730.77	936.17	962.13	674.93	882.90

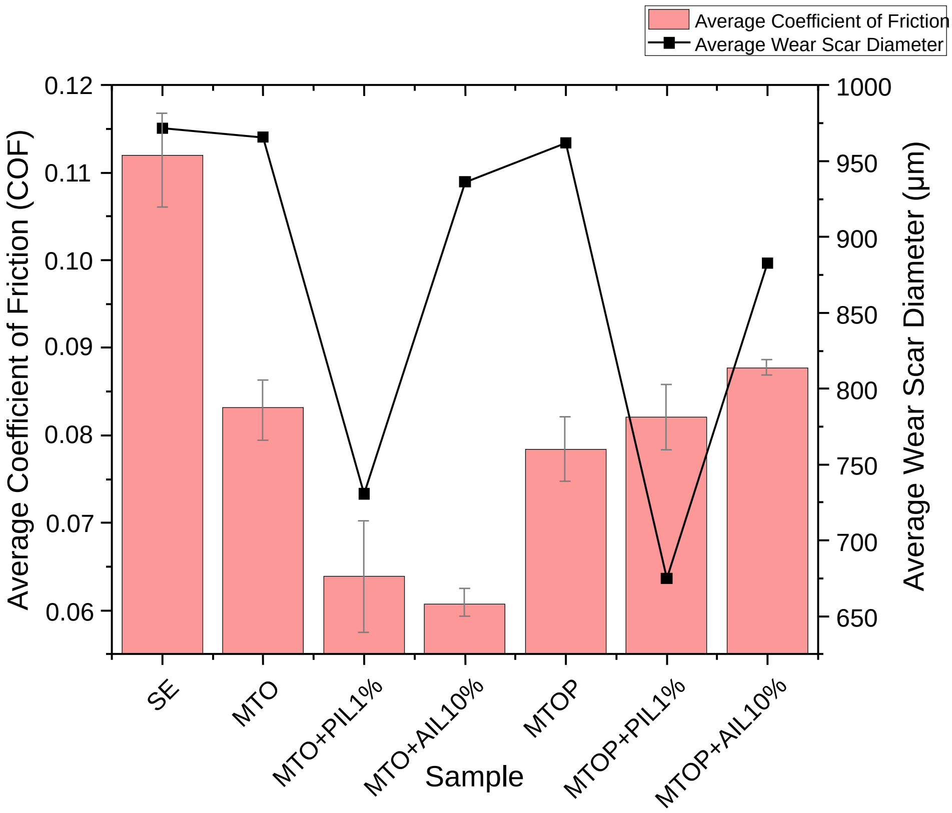

Fig. 5 - Graph of average coefficient of friction (COF) and average wear scar diameter (WSD)

4. Conclusion

This study highlights the used Tamanu oil as potential green alternative toward the sustainable metalworking fluid industries by means of chemical modification and adding additive. It can be concluded that;

1. The added ionic liquid also able to bring changes towards the properties of the Tamanu oil based on TMP-ester and Penta-ester. The addition of ionic liquid improves the physical properties of Tamanu oil based.
2. The result of viscosity index shows that MTOP+PIL1% had the highest value of 178.76. High viscosity index means, that the viscosity remains more stable across a wider temperature range. This means that sample MTOP+PIL1% lubricant with a higher viscosity index is more desirable, because it provides a more stable lubricating film over a wider temperature range.

3. The addition of ionic liquid into Tamanu oil based also shows improvement in average COF and WSD. The addition of 10% AIL and 1% PIL increase the anti-wear ability of the base oil. It provides good antifriction performance compares to SE.
4. The chemically modified processes and addition of ionic liquids had improved the crude states of Tamanu oil. With the used of sustainable resources and low toxicity additive, MTO and MTOP can be proposed for as a green alternative toward the development of the metalworking fluid industries in the future.

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