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Evaluation of physicochemical and tribological performances of hBN/WS_2 and hBN/TiO_2 hybrid nanoparticles-MJO-based oil

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KEYWORDS

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Tribology
Physicochemical
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Titanium dioxide
Tungsten disulphide

ABSTRACT

This work aims to investigate the physicochemical and tribological performance of modified Jatropha oil (MJO) with the addition of 0.025 wt.% of hexagonal boron nitride (hBN) + Titanium dioxide (TiO_2) (MJOht) and hBN + tungsten disulphide (WS_2) (MJOhw). The physicochemical properties of the samples were evaluated through kinematic viscosity and viscosity index. Four ball test was used to determine the tribological performance of the samples. All the MJO samples were compared with the benchmark oil, Synthetic Ester (SE). The result revealed that MJOht has excellent physicochemical properties in kinematic viscosity of 20.79 mm^2/s at 40 °C and 6.29 mm^2/s at 100 °C. MJOht also had the highest viscosity index (288). For tribological performance, MJOhw has an excellent coefficient of friction (COF) (0.0574) and lowest surface roughness (Ra) (0.11 µm)., while MJOht shows a smaller mean wear scar diameter (MWSD) (718.6 µm). Overall, MJO with hybrid additives shows excellent performance on physicochemical and tribological properties and making it a feasible choice as a green metalworking fluid.

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1.0 INTRODUCTION

Over the last two centuries, metalworking fluids (MWFs) have been employed in various metal cutting processes. MWFs formed a coating of lubricant to reduce friction, act as a cooling medium to reduce heat generation, and restrict the elution of metal chips, hence preventing metal pick-up. These capabilities will help to decrease tool wear, minimize energy consumption, and provide efficient surface quality on the workpiece (Wickramasinghe et al., 2017). MWFs have often been created as either straight oils or a mix of water, oil, surfactants, and additives. Annually, factories utilize about 2 billion litres of mineral-based MWFs, producing an enormous need for non-renewable feedstock (Skerlos et al., 2008). Mineral oil was one of the sources of MWFs (Talib et al., 2022a). According to Sun et al. (2017), nearly 90% of lubricants are derived from fossil fuels. Almost half of all mineral lubricants enter the environment, causing irreversible environmental harm owing to direct contact with water and soil (Reeves et al., 2017). Mineral oil based MWFs, which are often used in metal machining sectors, have been linked to worker skin irritation, respiratory illnesses, and cancer. Because the MWF effluents had a lower biodegradability rating, it is not legal to release the waste fluid into the environment without treatment (Wickramasinghe et al., 2017, 2020). Thus, it shifts the attention of using mineral-based MWFs to green MWFs and vegetable-based oil.

In comparison to mineral oils, the saturated fatty acids in vegetable oil provide a superior layer of lubrication at the work-tool interface (Guo et al., 2017). Vegetable oil has a triglyceride structure that provides desired qualities of the lubricant, such as higher lubricity, higher viscosity index, higher shear stability, lower volatility and higher load carrying capacity (Jamaluddin et al., 2020a; Lawal et al., 2012). Mushtaq & Hanief (2021) study the tribological characteristics of jatropha oil with additives. They found that the tribological properties of Jatropha oil improved with the addition of the additive. They stated that additives form tribo-layer in the metal surface.

However, vegetable oil must be altered before usage in order to improve its limitations in terms of oxidation stability, high friction, high viscosity, thermal stability, and corrosion resistance (Jamaluddin et al., 2020a). According to Rahim et al. (2018), raw vegetable oil's physical and chemical qualities have many limitations. They need modification to improve the qualities to be used as MWFs. Various study has been done on the modification of vegetable oil. Talib & Rahim (2015) evaluated the chemically modified crude jatropha oil as a metalworking fluid for machining. They found that modified crude jatropha oil performs better than crude jatropha oil and synthetic ester, SE. In addition, Talib et al. (2018) also used modified vegetable oil and modified RBD palm olein oil in the machining process. They evaluated the tribological performances of modified RBD palm oil and found that modified RBD palm olein oil shows outstanding quality than raw oil and SE. Rahim et al. (2017) also studied the various formulation of Modified RBD Palm Olein as metalworking fluid. The ratio of methanol to RBD Palm olein were varied, 3:1, 6:1 and 9:1. They found out that the formulation of 6:1 shows excellent tribological properties and suggest that it can use as an alternative feedstock for metalworking fluid.

Moreover, reformulating additives to vegetable oil also enhances its properties as metalworking fluids. Nanofluids are commonly used in lubrication systems, where nanometer-sized particle additives are introduced to base fluids and feature anti-friction and anti-wear capabilities, promoting advances in physicochemical properties and tribological performance. The previous researcher study on the tribological performance of modified jatropha oil with the addition of hBN nanoparticles for MWFs. The concentration of hBN varied from 0.05wt.% to 0.5wt.%. They discovered that the lowest concentration of hBN at 0.05 wt.% surpassed SE in terms of friction and wear (Talib et al., 2018). Another study using MJO and hBN as additives was also conducted. The concentration of hBN added was lower than the addition of hBN in Talib et al. study, which is 0.01, 0.025 and 0.05 wt.%. They concluded that adding additives has improved the base oil's kinematic viscosity and viscosity index. Besides that, they also found that MJO with additives had a lower coefficient of friction (COF) compared to Synthetic ester (SE) and its base oil. They also conclude that the addition of hBN has anti-friction features (Jamaluddin et al., 2020a). hBN nanoparticles in the form of lamellar powder created layered crystal structures resulting in low friction. The protective boundary lubrication film that was created because of the lamellar powder adhering to the contact surface was able to reduce the amount of contact surfaces and minimize wear. In order to reduce friction, it tended to have its layers closely allied parallel to the direction of motion (Talib et al., 2017). Paturi et al. (2016) used WS_2 as additives for the turning process of Inconel 718. They mentioned that with assistance from WS_2, the surface roughness decreased by about 35%. There was also a study on WS_2 that stated that WS_2 particles were adhered to the worn areas, thus producing a tribo-layer, thus reducing the frictional force and wear (Zhang et al., 2020). Kumar et al.(2017) investigated of physicochemical properties of TiO_2 nano-lubricant oil. They found that the viscosity of the oil with additives slightly improved. In addition, the combination of TiO_2 and lubricating oil reduced the coefficient of friction (Ingole et al., 2013). From previous study, the lubrication performance was improved by adding single nanoparticles to the base oil in order to create a protective layer between the contact surface. Under these circumstances, nanoparticles may produce a rolling impact at the mating surfaces, which could transform sliding friction into both sliding and rolling friction (Talib et al., 2021). Additionally, nanoparticles trapped between mating surfaces during contact between two solid surfaces can fill the micro- and nano-gaps of the rubbing surfaces. As a result, a tribolayer will develop on the worn surfaces, which can lessen direct contact between the two surfaces and reduce COF (Omrani et al., 2019).

Due to the different tribological properties of single nanoparticles, it is worth looking into how the two particles work together as a lubricant additive. It has been discovered that hybrid nanofluids have greater strengths and potential than single nanofluids in terms of physical and chemical properties such as thermal conductivity, thermal stability, anti-wear, and friction performance. This discovery has piqued the interest of numerous researchers in this field and led to a progressive increase in the amount of research being done on hybrid nanofluids in recent years. The incorporation of hybrid nanoparticles into biobased oil might improve the capability of nanofluids. Since combining two or more components results in a more noticeable lubricating performance than individual nanoparticle performance, hybrid nanoparticles are particularly crucial for lubricant additives. Previous research on synthetic TiO_2/MoS_2 nanoclusters as lubricant additives improved tribological performances by 30.8 % and 40%, respectively, over pure MoS_2 and pure TiO_2 (Hu et al., 2011). Meng et al. (2021) examined tribological performance of single nanoparticles and SiO_2/MoS_2 hybrid nanoparticles dispersed in deionized water. They conducted a half-hour grinding test to measure the change in friction coefficient for each sample. They found that hybrid nanofluids displayed remarkable frictional characteristics because of the formation of a hybrid lubrication film, improved adsorbing properties, and the synergistic interaction between single nanoparticles. This research provided evidence that introducing hybrid nanoparticles to a base lubricant improves lubrication performance more than using single nanoparticle additives. Since two or more nano-additives can make up for the performance flaws of each individual nano-additive and those two different types of nanomaterials tend to work synergistically to increase friction performance.

Therefore, this study formulated hybrid nanofluids from modified Jatropha oil (MJO). MJO was incorporated with two types of hybrid nanoparticle additives and at 0.025 wt.% concentration of the nanoparticles to develop a sustainable MWF. The biobased oils were blended with nanoparticles of hexagonal boron nitride (hBN) hybrid with titanium dioxide (TiO_2) and tungsten disulphide (WS_2), respectively. The physicochemical of the hybrid nanofluids were evaluated through the kinematic viscosity and viscosity index of hybrid nanofluids samples, and tribological properties were investigated through a four-ball test. The outcomes were compared to pure MJO and synthesized ester (SE). These new hybrid formulation nanofluids may widen the possibility of MWFs in the lubrication industry.

2.0 EXPERIMENTAL PROCEDURE

2.1 Preparation of Hybrid Bio-based Metalworking Fluids

The sample preparation starts with the chemical modification of Crude Jatropha oil (CJO) to improve the oxidation, thermal and viscosity stability. The first step is the esterification process, which focuses on lowering the concentration of free fatty acids (FFA) to less than 1% to produce a high JME yield. The FFA contents in CJO are in the range of 2.5- 65% according to Talib et al. (2017). CJO undergo two-step acid-based catalyst transesterification which is the first step was the esterification process of CJO with methanol (CH_4O) with the presence of sulphuric acid(H_2SO_4) as a catalyst. The first process produces esterified Jatropha oil (EJO) as the product. After that, EJO reacts with methanol and sodium hydroxide (NaOH) to produce Jatropha methyl ester (JME). This reaction takes place for 2 hours and at a constant temperature of 60°C in water bath conditions. The ideal reaction temperature leads to efficient rate of reaction, thus produce higher yield of JME. The viscosity of CJO was reduced at 60°C, resulting in better mixing of oil with alcohol and speed up the separation of glycerol from the JME (Chozhavendhan et al., 2020). The next step was the transesterification process of JME with trimethylolpropane (TMP) with the presence of 1 wt.% sodium methoxide ($NaOCH_3$) as a catalyst and produced Modified jatropha oil (MJO). The molar ratio of MJO and TMP was 3.5:1. The temperature of the reaction was maintained at 120 °C in the oil bath and carried out in vacuum condition for 24 hours at a constant pressure of 20 kPa as shown in the set-up in Figure 1.

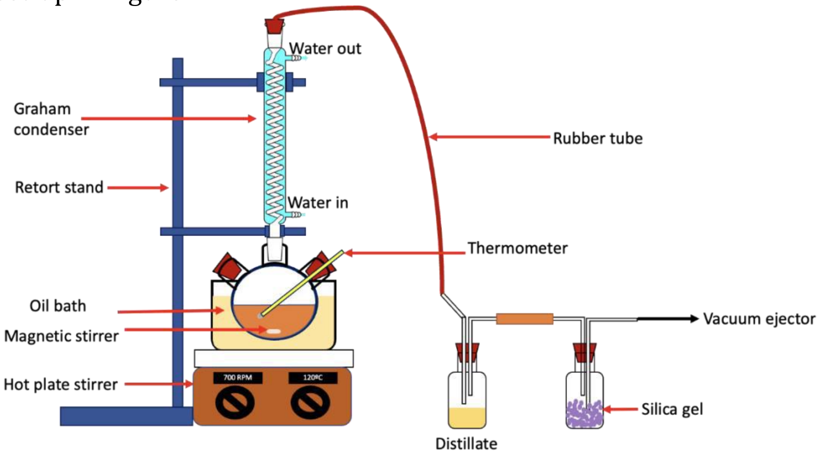


Figure 1: Set-up of transesterification process for reaction of JME with TMP.

Then, MJO was blended with 0.025% by weight of hybrid nanoparticles: hexagonal boron nitride (hBN) with tungsten disulphide (WS_2) and titanium dioxide (TiO_2), respectively, as shown in Table 1. The nanoparticles' properties are listed in Table 2. A magnetic stirrer was used to mix the MJO with the 0.025 wt.% of hybrid nanoparticles for 30 minutes at 700 rpm and 60°C. In the next phase, a Bandelin HD3200 model ultrasonic homogenizer was used for 30 minutes to homogenize the mixture (at 20 kHz frequency and 200 W). In the last step, any precipitation or layer separation in the combinations was visually observed. This procedure ensured that hybrid nanoparticles were evenly distributed in MJO. The sample was examined over time using the sedimentation technique to assess the dispersion stability. Since the density of hybrid nanoparticles is greater than that of MJO, hybrid nanoparticles tend to settle over time. However, the dispersion of hybrid nanoparticles in MJO is deemed to have high dispersion stability, as there is no noticeable sedimentation over a brief observation period.

Table 1: Samples description.

Biobased oil	**Symbol**	**Description of hybrid nanofluids**
MJO	MJOht	MJO + 0.025 wt.%hBN + TiO_2
	MJOhw	MJO + 0.025 wt.%hBN + WS_2

Table 2: Properties of nanoparticles.

Properties	**hBN**	**WS_2**	**TiO_2**
Appearance	Colourless crystal powder	Blue Gray powder	White solid powder
Density (g/cm^3)	2.3	7.5	4.23
Thermal expansion coefficient (10^{-6}/K)	1	10	8.4
Thermal conductivity (W/m K)	8.4	53	4.8

2.2 Physicochemical Test

The physicochemical performances were evaluated by kinematic viscosity and dynamic viscosity of the hybrid nanofluids samples. The kinematic viscosity of the samples was measured at 40 °C and 100 °C using a Viscometer machine, Viscolite 700, according to ASTM D445. The viscosity index (VI) was calculated using the kinematic viscosity of the lubricant following the calculation method provided by the interpolation data from the ASTM D2270 standard. The samples were compared with the benchmark Synthetic Ester (SE), Unicut Jinen MQL.

2.3 Tribological Test

The samples' tribological performance was analysed using a four-ball wear tribotester machine (Ducom TR-30 L) in accordance with ASTM D4172. The ball used in this experiment was a chrome steel ball (AISI 52100) with a diameter of 12.7mm and hardness between 64 to 66 HRC. The steel balls had an average surface roughness (Ra) of roughly 0.02 μm. Four new sets of balls were used for each of the experiments. From the schematic diagram in Figure 2, three stationary balls were placed inside the ball pot and the collet held one rotating ball. Approximately 10ml of the hybrid sample was poured inside the ball pot. After that, the ball pot assembly was placed inside the four-ball machine. For each set, the upper ball rotates against three stationary balls at 392 N load, a speed of 1200 rpm, and a temperature of 75°C for 60 minutes.

The coefficient of friction (COF) was calculated using Winducom software based on the testing results. The stationary steel balls' average wear scar diameter (MWSD) was determined using a scanning electron microscope (SEM, Toshiba S-3000N). Finally, a surface roughness tester in accordance with ISO4288:1996 was used to measure the surface roughness of the steel ball's worn surfaces. The hybrid nanofluid samples were compared to SE and biobased oil in terms of lubricating performance.

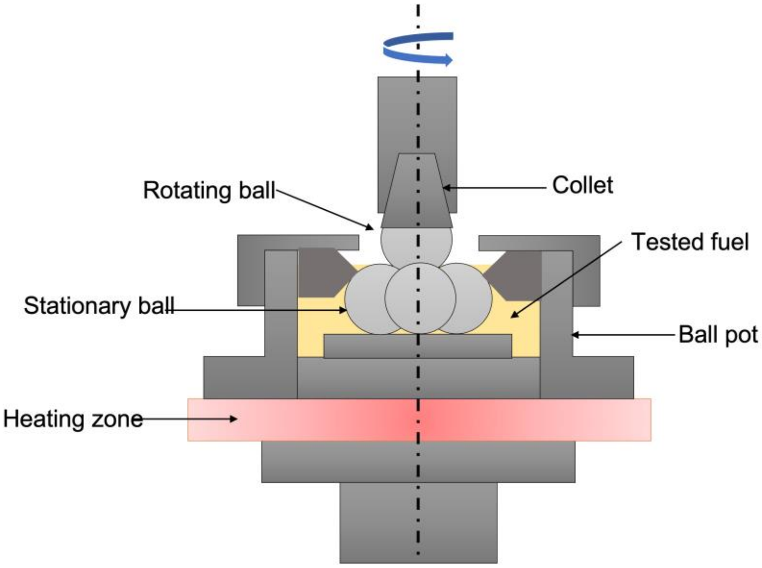


Figure 2: Schematic diagram of four-ball tester.

3.0 RESULTS AND DISCUSSION

3.1 Kinematic Viscosity and Viscosity Index

The oil's viscosity affected the production of a thin protective layer at metal contacts, which works as friction and wear reduction throughout the machining process (Amiril et al., 2017). The kinematic viscosity at 40 °C and 100 °C and the viscosity index of all samples were shown in Figure 3. The kinematic viscosity of all samples was higher at 40 °C compared to at 100 °C. This is because the rate of intermolecular change was higher at 100 °C and the reduced cohesive force between molecules, thus lowering the samples' viscosity (Wenhao, 2021). From the graph, SE provided the highest kinematic viscosity of 23.12 mm^2/s at 40 °C compared to the MJO samples. This is due to the number of the carbon chain of SE (C8 to C10) being shorter than the number of MJO carbon chains (C16 to C18) (Jamaluddin et al., 2020b; Talib et al., 2019). Due to the transesterification process, MJOs underwent a chemical alteration that altered their structure. Consequently, the intermolecular tensions on the hydrogen bond had reduced, resulting in a considerable decrease in the viscosity of the oil product (Rani et al., 2015).

In addition, MJO samples with the addition of hybrid nanoparticles show increments in kinematic viscosity. This phenomenon arises because nanoparticles produce larger nanoclusters that impede the movement of fluid layers across one another. Consequently, the intermolecular forces increase and the layers move closer together, causing the viscosity to increase (Esfe & Esfandeh, 2020). Among MJO samples, MJOht shows a higher kinematic viscosity of 20.79 mm^2/s at 40 °C and 6.29 mm^2/s at 100 °C. This proved that the addition of TiO_2 improves the viscosity of nanofluid. Nik et al. (2020) came to a similar conclusion, stating that incorporating well-dispersed TiO_2 nanoparticles increases flows resistance, increasing kinematic viscosity.

From Figure 3, the graph also shows the calculated viscosity index of all samples based on kinematic viscosity at 40°C and 100°C. The viscosity index of a lubricating fluid reveals how significantly the viscosity varies with temperature. A high viscosity index implies that the viscosity of the fluid is not impacted by changes in temperature, whereas a low viscosity index suggests a significant viscosity change. From the graph, MJO had a higher viscosity index of 227 and showed a 52% improvement compared to SE. The viscosity index increased with the addition of hybrid nanoparticles. MJOht and MJOhw had a higher viscosity index of 288 and 281, respectively and improved the viscosity index of MJO by 24% and 27%, respectively. This due to the presence of hBN as additives improved the viscosity index of MJO. The low thermal expansion coefficient of hBN (1X 10^{-6}/°C) provide thermal stability to oil and maintained bigger thermal network (Talib et al., 2019; Yıldırım et al., 2019). The presence of TiO_2 hybrid with hBN also assist in improvement of viscosity index. Among MJO samples, MJOht has the highest viscosity index This is because of TiO_2 has thermal expansion coefficient of 8.4X 10^{-6}/°C lower than thermal expansion coefficient of WS_2. The combination of hybrid hBN with TiO2 and hybrid hBN with WS_2 demonstrates that the presence of two distinct additions improves the characteristics of the vegetable-based oil. The similar result was seen in a research by Moghaddam & Motahari (2017), who reported that the viscosity of based oil including two additives was greater than the viscosity of based oil alone.

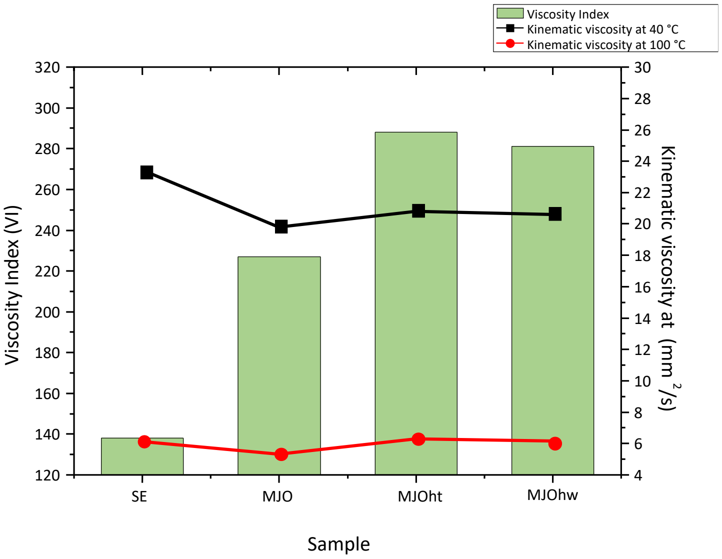


Figure 3: Kinematic viscosity and viscosity index of all samples.

3.2 Coefficient of Friction (COF) and Mean Wear Scar Diameter (MWSD)

Figure 4 and Figure 5 displays the tribological performances of coefficient of friction (COF) and mean wear scar diameter of hybrid nanofluids samples. Modified Jatropha Oil (MJO) samples offer superior tribological properties compared to synthetic ester (SE) nanofluids. The COF of MJO samples improves from 31%to 48%, while the MWSD improves from 4 % to 28%. This is because of the viscosity index of SE increased the flow resistance thus resulting in high COF and MWSD. The lubrication form from SE was unstable in wide range of temperature due to its low viscosity index value (Latif et al., 2019; Talib et al., 2022b). Moreover, the long molecular chain and branches in MJOs produced strong lubrication films(Talib et al., 2017). Specifically, the polar functional groups of fatty acid molecules in the MJO samples lead to their ability to attach firmly to the metal surface (Lubis et al., 2017). In addition, the polar carboxyl group was densely packed and produced a lubricating coating that was adequate to minimize friction (Kashyap & Harsha, 2016).

In Figure 4, the COF MJOht shows insignificantly improvement compared to MJO. MJOhw shows improvement in terms of COF among all MJO samples by 48%. The addition of WS_2 combine with hBN nanoparticles to produce hybrid nanoparticles as additives to MJO based improved the COF. The asperity valleys at the sliding interface were filled with hBN nanoparticles, which formed a thin lubrication coating that allowed the particles to align themselves parallel to the relative motion. The particles slid over one another, which decreased the stress concentration at the contact surfaces and contributed to a low number of forces, which decreased the friction coefficient (Sani et al., 2017). In addition, WS_2 generates a depositional coating on the surface, which fills and evens out surface roughness and sliding friction. When the load increases and friction is intensified, the chemical reaction between WS_2 and the friction pair's base material results in the production of a lubricating FeS layer. It can prevent friction pair surfaces from coming into direct contact, considerably minimizing coefficient of friction(Chen et al., 2007).Moreover, The lamellar structure of WS_2 solid lubricant allows it to easily shear along the friction sliding direction, resulting in minimal heat production and frictional impacts on the ball surfaces (Paturi et al., 2016). This proved by Zhang et al. (2020) stated in their research, the addition of WS_2 reduced the frictional force and wear, thus reduced the COF.

As shown in Figure 5, MJOht and MJOhw had smaller mean wear scar diameter compared to MJO and SE. The MWSD of MJOht was 718.6 µm and MWSD for MJOhw was 727.2 µm. The viscosity of the MJOht and MJOhw was higher than MJO produced uniform and stable lubrication film, thus reduced the MWSD. The addition of hBN with TiO_2 and WS_2 respectively improved the MWSD. This was proved by previous research of (Jamaluddin et al., 2020a, 2020b) shows the addition of hBN improved the MWSD.

Figure 6 shows the morphology of the wear scars on the surfaces of ball steel for MJO samples and SE. On the worn surface of each sample, parallel, deep, parallel grooves were noticed. On the SE wear scar, there was material transfer, abrasive wear, and adhesive wear. MJO samples have a shallow groove compared to SE. On the MJOht, darker concentric grooves indicating an abrasive wear surface were found, however on the MJOhw, the wear track surface was smoother. This is attributed to hBN lowering friction between the ball (Talib et al., 2019), and WS_2 generating an effective lubricating layer (Chen & Daheng, 2009). Compared to SE, the inclusion of nanoparticles produced a worn surface with fewer grooves and material transfer spots. During lubrication, the fatty acid chain generated a thin layer of soap film, which was removed, resulting in a deep groove on the worn surfaces (Jamaluddin et al., 2020a).

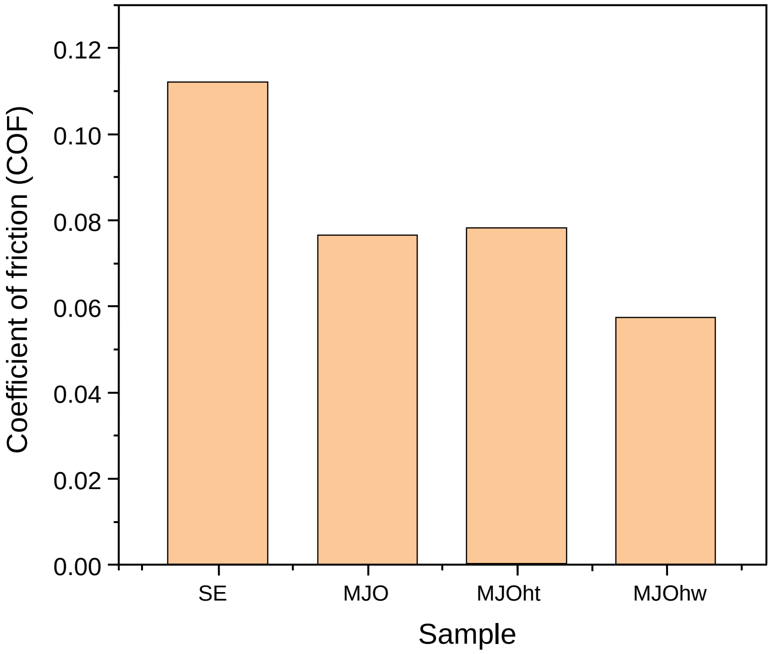


Figure 4: Coefficient of friction (COF) of all samples.

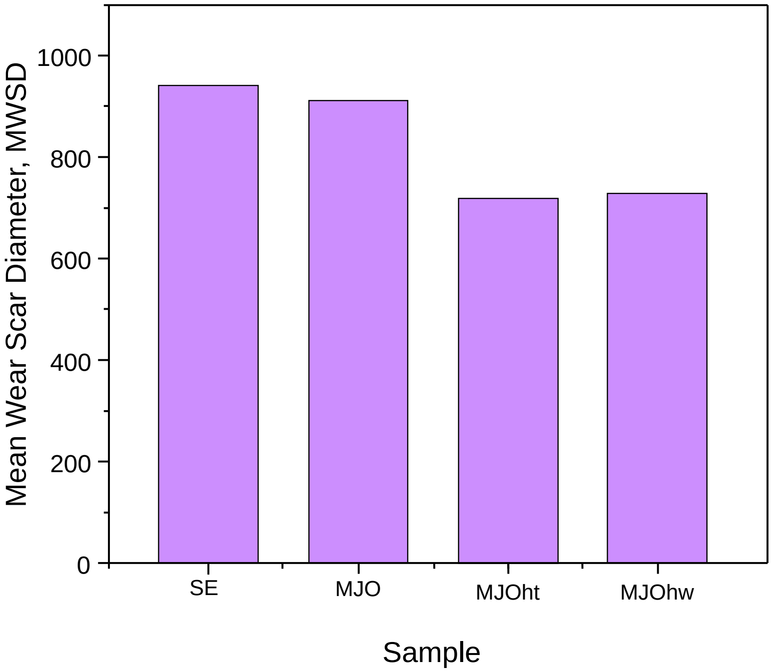
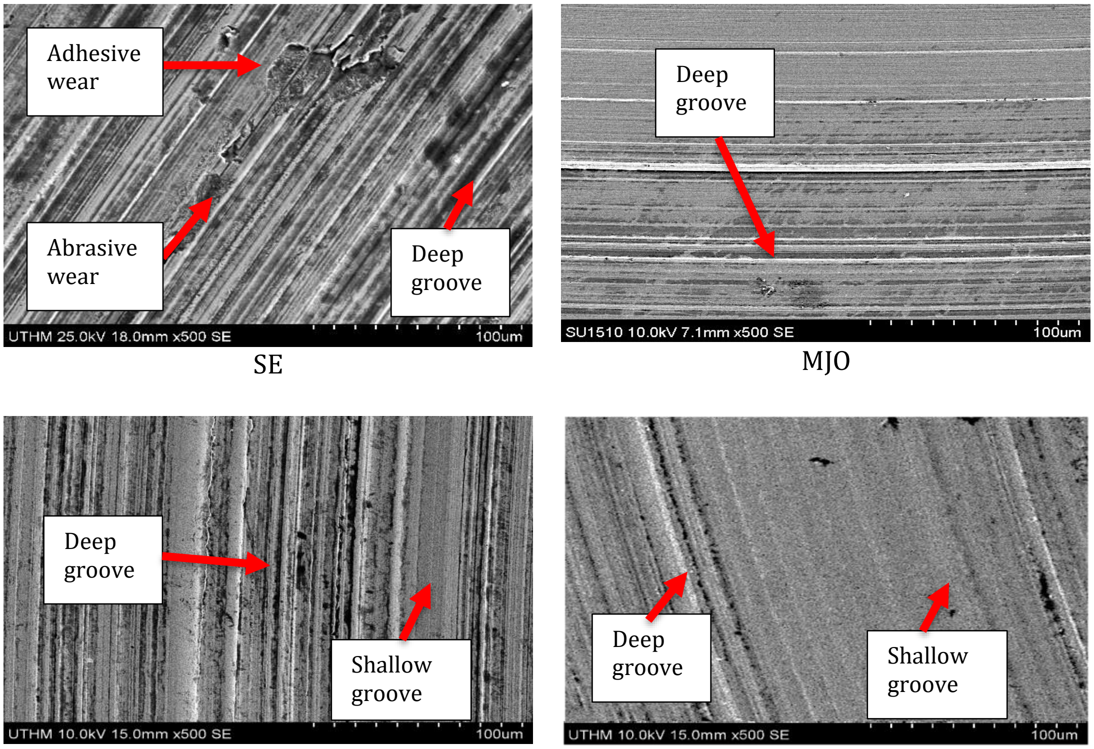


Figure 5: Mean wear scar diameter (MWSD) of all samples.



SE MJO

MJOht MJOhw

Figure 6: SEM morphology of the wear scars on the surfaces of ball steel at a magnification of 500x.

3.3 Surface Roughness

Figure 7 displayed the surface roughness distribution for all lubricant samples. Following the four-ball test, the steel ball's surface roughness was measured to observe its wear. According to the results, SE's sample had a rougher surface than the MJOs sample, measuring 0.31µm. This demonstrated that the SE offered an inadequate lubricating layer since the surface wear produced was harsh in comparison to the other MJO samples. The presence of hybrid nanoparticles in MJO improved the surface roughness of steel ball's worn surface by 52% to 65% compared to SE. MJOhw provided lowest surface roughness of 0.11 µm and followed by MJOht provided 0.15 µm. This demonstrated that the nanoparticle additions have a role in wear reduction by filling the asperities between the contact surfaces, resulting in the production of a greater protective transfer layer. The addition of hBN nanoparticles to MJO based oil improve the surface roughness as the structure of lamellar powder adhered to the contact surface (Jamaluddin et al., 2020b). The presence of WS_2 nanoparticles fill up the low valleys of the friction's pair surface form a surface depositional film, filling and levelling up the surface roughness and sliding friction (Zhu et al., 2019). In addition, the synergy effect of two different nanoparticle additives cover up each other flaws in lubrication process, resulting in better lubrication performances.

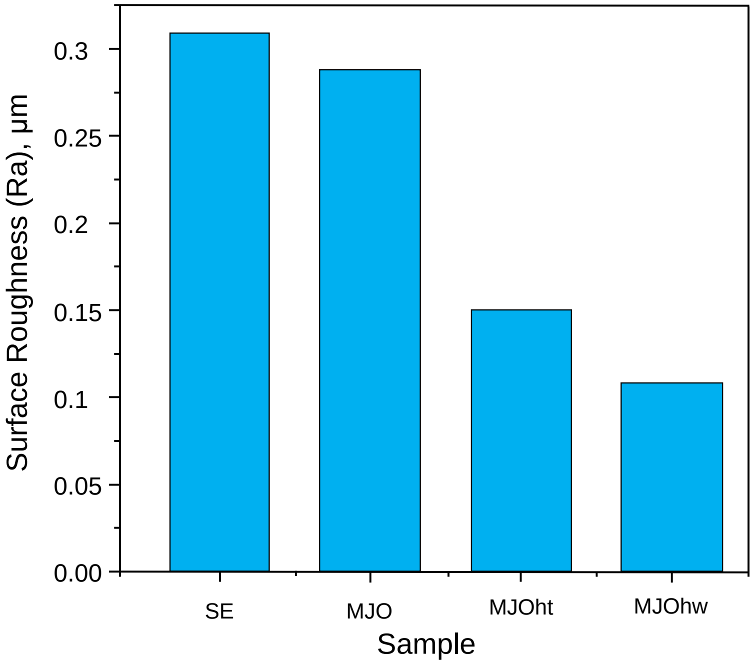


Figure 7: Surface Roughness of all samples.

CONCLUSION

In the present study, physicochemical characteristics and tribological tests were conducted to evaluate the relative effectiveness of each hybrid nanofluids sample as a sustainable lubricant by comparing the results to commercial SE. It was found that the chemical modification and the addition of hybrid nanoparticles improve the physicochemical and tribological properties of MJO. SE. MJO with 0.025 wt.% of hBN and TiO_2 (MJOht) had the highest VI of 288. It has been demonstrated that the viscosity of the lubricant affects its performance because it changes the thickness of the lubrication film. MJO with 0.025 wt.% of hBN and WS_2 (MJOhw) has the lowest coefficient of friction (COF) (0.0574) and surface roughness (Ra) (0.11 µm). It also has a smoother worn surface. MJOht has the lowest mean wear scar diameter (MWSD) (718.6 µm). MJO with hybrid additives shows outstanding result and suitable to substitute benchmark oil, Synthetic Ester (SE).

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