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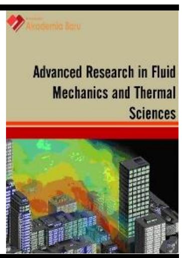

Tribological Analyses of Modified Jatropha Oil with hBN and Graphene Nanoparticles as An Alternative Lubricant for Machining Process

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ABSTRACT

The increase of health and environmental consciousness has motivated the effort of technology improvement on lubrication by finding and exploring another potential alternative to replace mineral-based metalworking fluids. Due to this concern, vegetable-based oils have been recognised as an ideal lubricating base oil in machining due to low toxicity, biodegradable, and renewable energy sources. Moreover, nanofluids have attracted enormous attention in the field of lubrication due to excellent physical and chemical properties that can enhance tribological characterisation. The objective of the current work is to develop a new formulation of nanofluids in modified jatropha oil (MJO) by adding hexagonal boron nitride (hBN) and graphene nanoparticle additives at the lowest concentration (0.01, 0.025. and 0.05 wt. %). The physicochemical tests in terms of kinematic viscosity and viscosity index were conducted and compared with synthetic ester (SE). Tribology testing was conducted through four-ball test to determine the coefficient of friction, mean wear scar diameter, and friction torque. The result shows a significant improvement of MJO samples by adding nanoparticle additives compared to the SE. MJOg2 (MJO + 0.025 wt. % of graphene) exhibited excellent tribological behaviour by providing the lowest coefficient of friction and friction torque. Meanwhile, MJOh1 (MJO + 0.01 wt. % of hBN) provided with a smaller mean wear scar diameter among other lubricant samples. Conclusively, the addition of nanoparticle additives significantly enhanced the tribological characteristics and is highly suitable as a substitute for SE.

1. Introduction

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Metalworking fluids (MWFs), which are also known as lubricants, coolants, or cutting fluids, are widely used in manufacturing and industrial processes. MWFs play a vital role in minimising friction and wear between tool and workpiece components that are in close proximity and move relatively to each other by providing a layer between both surfaces to help carry the load where the pressure is generated between the opposing surfaces [1]. Approximately 85% of MWFs in industrial applications are derived from petroleum-based oil as the source provides good lubrication capacity and high stability with a significantly prolonged lifetime [2]. However, the demand for mineral-based lubricants increased due to the rapid development of modern technology, which recorded high consumption of mineral-based lubricants for about 96 million barrels per day in 2016. A primary concern of mineral-based lubricants is the disposal management of the used oil as the oil is non-biodegradable and heavily toxic to human and the environment [3]. Due to this scenario, vegetable oil has received considerable attention among the researchers as a replacement for mineral-based lubricants due to the promising opportunities as a lubricant in machining processes [4-6]. Vegetable oils are derived from food and renewable sources that are highly biodegradable and have low toxicity towards human and the environment. However, vegetable oils are not widely used as a base stock of lubricants due to undesirable properties of low thermal and oxidation stability [7]. According to Panchal *et al.,* [8], vegetable oils can undergo chemical modification to develop a perfect biodegradable lubricant, able to withstand wide operating conditions. Additionally, a suitable nanoparticle additive can be added to the base stock to provide an enhancement in terms of physicochemical properties, hence improves the tribological performance of the lubricant. For further improvement of the physicochemical properties, nanoparticles are one of the suitable additives [9].

A recent review by Ajithkumar and Xavior [10] stated that a nanolubricant is attributed to the rolling, sliding, or filming of nanoparticles between the interface of tool and workpiece. The inclusion of nanoparticles in lubricants improves the wettability, lubricant properties, and heat transfer efficiency, which increase the tool life and surface finish. Vegetable oil with the addition of nanoparticles as a nanofluid is more effective in machining compared to pure oil due to the significant reduction of temperature and friction at the cutting zone. Singh *et al.,* [9] conducted an experiment on the tribological behaviour of macadamia oil with the addition of copper nanoparticles at the concentration range of 0.1%–0.5%. The result revealed that the tribological properties in terms of wear scar obtained in the presence of macadamia nanofluids are smoother compared to pure macadamia oil. The optimum concentration of copper nanoparticles (0.3%) in macadamia oil exhibited a minimum coefficient of friction and specific wear rate, as well as smoother worn surfaces after sliding Lv *et al.,* [11] proved that the coefficient of friction (COF) and worn scar diameter (WSD) decreased by the addition of nanoparticles in the base oil. In their study, tribological and machining performance were evaluated using commercial vegetable oil (LB-2000) with the addition of graphene nanoplatelets (GPLs) as the minimum quantity lubricant (MQL) cutting fluid at the concentration of 0.02 to 0.08 wt.%. From the result, the optimum concentration of GPL of 0.06 wt. % demonstrated the lowest COF and WSD. They emphasised that a self-lubricating film was formed as GPLs penetrated into the worn surface to make up the furrows and pits on the rubbing interface and thus, prohibited the rough faces from direct contact. Furthermore, Chatha *et al.,* [12] studied the performance of drilling aluminium 6063 under minimum quantity lubrication using a nanofluid from refined soy bean oil as the cutting fluid with the addition of Al_2O_3 nanoparticles at the concentration of 1.5 wt. %. Based on the results, the nanofluid is very effective and showed better results in drilling with respect to pure MQL, flood cooling, and dry cutting in terms of cutting force, surface roughness, and tool wear. This is because the nanoparticles create a thin lubricating film on the workpiece and tool. The nanoparticles flow at the interface with the vegetable oil and decrease the plastic contact, leading to the reduction of flank wear.

Thus, the present study aims to obtain a new formulation of a biodegradable lubricant from vegetable oil with the addition of green solid additives to enhance the tribological performance outcomes. In this study, modified jatropha oil (MJO) was selected as a sustainable metalworking fluid for machining process. The effectiveness of the green solid additives, namely hexagonal boron nitride (hBN) and graphene nanoparticles, was evaluated at various concentrations. The tribological performance of MJO at various concentrations of hBN and graphene nanoparticles was investigated using four-ball test to determine the potential of the newly developed MWF.

2. Experimental Procedures

2.1 Preparation of Bio-Based Lubricant Samples

The bio-based MWF was developed from crude jatropha oil (CJO) through chemical modification and addition of additives to improve the performance of lubricant in terms of thermal and stability characteristics of the oil. The CJO was chemically altered via a two-step acid-based catalyst transesterification process to produce jatropha methyl ester (JME). Then, the chemical reaction of JME with trimethylolpropane TMP (JME:TMP) was carried out in the presence of 1% (wt./wt.) sodium methoxide ($NaOCH_3$) at a molar ratio of 3.5:1 to produce TMP triester, which is also known as MJO [13]. As illustrated in Figure 1, the reaction was conducted in a three-necked round-bottom flask and capped with a Graham condenser. MJO was mixed with hBN and graphene nanoparticles by varying the concentration of the additives at 0.01, 0.025, and 0.05 wt. % (based on the weight of the oil sample). Table 1 indicates the prepared sample with and without the addition of hBN and graphene additives.

Subsequently, the physicochemical properties of MJO samples in terms of kinematic viscosity and viscosity index were performed according to the procedure of the American Society for Testing and Materials (ASTM) standard and compared with a commercial SE (Unicut Jinen MQL). The kinematic viscosity was measured at 40 and 100 °C based on ASTM D445 by using a portable viscometer. The lubricant sample was heated at the desired temperature before being immersed in the viscometer. Furthermore, the viscosity index was calculated according to ASTM D2270, which was related to the data of kinematic viscosity obtained at 40 and 100 °C.

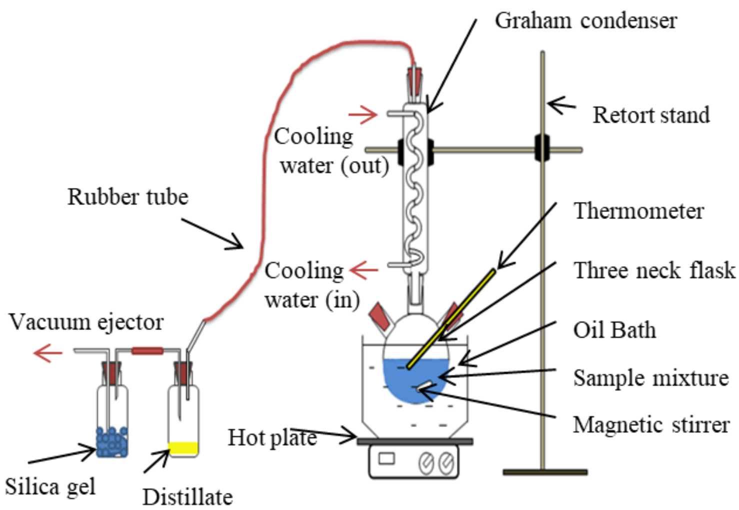

Fig. 1. Setup for chemical modification process

Table 1
Description of the MJO samples

Name of sample	Concentration of additives (wt.%)	Type of additive nanoparticles
MJO	-	No additive
MJOh1	0.01	
MJOh2	0.025	hBN
MJOh3	0.05	
MJOg1	0.01	
MJOg2	0.025	Graphene
MJOg3	0.05	

2.2 Four-ball Tribological Testing

The study of tribological characteristics of a MWF mainly focuses on friction and wear mechanisms. In this study, the tribology test was carried out using the four-ball test according to the standard procedure of ASTM D4172 using DUCOM four-ball tribotester. The ball material used for this testing is a chrome steel ball (AISI 52100) with the diameter of 12.7 mm and range of hardness be-tween 64 and 66 HRC. The lower ball of the test consists of three stationary steel balls clamped together in the ball pot assembly. Approximately 10 ml of the lubricant sample was poured into the ball pot assembly in each testing session. The role of the fourth ball in this test is as a rotating ball. It is locked inside the collet and tightened into the spindle. The ball pot assembly was installed in the four-ball machine and pressed slowly at the normal load of 392 ± 2N to prevent any concentrated stress. The constant speed of 0.461 m/s (i.e., 1,200 rpm) was used to rotate the upper rotating ball for 60 min when the designated temperature was reached. Figure 2 displays the setup for the four-ball tribological test. The COF was automatically calculated based on the friction torque data recorded using Winducom 2010. The mean wear scar diameter (MWSD) of the stationary steel balls was obtained from the average of all the steel ball scar captured with an optical camera and measured using an image acquisition system according to the average length of the horizontal and vertical scar. The friction torque was calculated ac-cording to IP-239 standard as pointed out in Eq. (1).

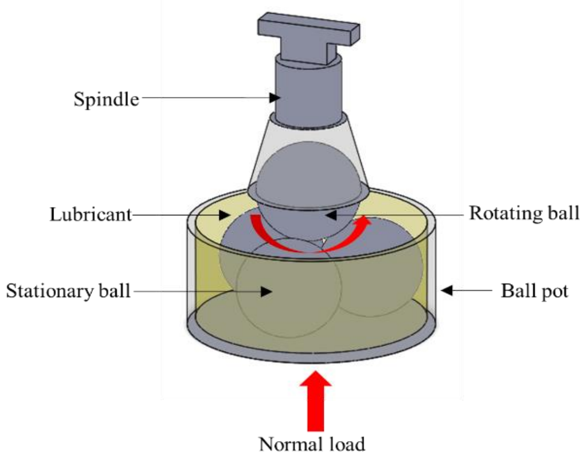

Fig. 2. Schematic diagram for four-ball tribo tester

$$Friction\ torque, T = \frac{\mu \times 3W \times r}{\sqrt{6}} \tag{1}$$

where, μ = Coefficient of friction, W = Applied load (N), r = The distance from the centre of the contact surface on the lower balls to the rotation axis, which is 3.67mm.

3. Results and Discussion

3.1 Physicochemical Properties of Bio-Based Lubricant

Figure 3 indicates the kinematic viscosity at 40 and 100 °C, as well as the viscosity index for all lubricant samples. It was observed that the SE provided the highest kinematic viscosity of 19.12 mm2/s at 40°C in comparison with MJO samples. This result proved that the SE had shorter carbon chains with C8 to C10 compared to MJOs that have long carbon chains between C16 and C18 [7]. Concurrently, the composition of MJOs changed due to the chemical modification via transesterification process. As a result, the intermolecular forces on the hydrogen bond had weakened, which significantly reduced the viscosity of the oil product [14]. The inclusion of nanoparticle additives showed the possibility of enhancing the values of kinematic viscosity. The result showed that the kinematic viscosity at 40 and 100 °C increased as the concentration of the additives increased. It can be seen that the kinematic viscosity of MJO samples blended with hBN nanoparticles was much lower compared to MJO samples with graphene nanoparticles. MJOg3 recorded the highest kinematic viscosity value at 40 °C (17.38 mm2/s) among MJO samples. Rani *et al.,* [14] emphasised that the viscosity should be optimum for all applications as low viscosity might cause more wear whereas high viscosity might cause critical frictional loss. Furthermore, it was also found that MJOg3 had the highest kinematic viscosity at 100 °C (i.e., 5.23 mm2/s) among other samples and the value was correlated with its highest viscosity index of 269. The sample was significantly affected by the negative coefficient of thermal expansion of graphene (-8 × 10-6/°C) compared to hBN [15]. MJOh3 had the second highest viscosity index as it also had a lower coefficient of thermal expansion with the value of 1 × 10-6/°C. Additionally, it can be observed that the viscosity index increased as the concentration of nanoparticles increased due to the higher level of interaction that maintained a larger thermal network [7].

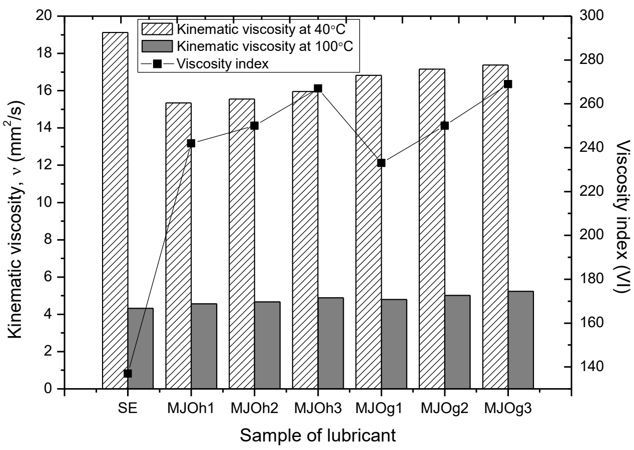

Fig. 3. Variation of kinematic viscosity and viscosity index of lubricant sample

3.2 Four-Ball Test Result
3.2.1 Coefficient of friction

Figure 4 illustrates the results of COF for various MJO samples in comparison to SE. The SE recorded the highest COF value of 0.0894 compared to MJO samples, which recorded lower COF values with 22%–62% improvement of COF. This is due to the formation of molecular chains in MJO, which produced lower friction at the contact surface. It was observed that the MJO samples blended with nanoparticle additives had improved COF. These results showed the significant role of nanoparticles that reduced the COF by filling inter-asperity valleys. The mechanism provided by the nanoparticles by filling the asperities formed thin transfer films that provided more effective lubrication by allowing the particles to align themselves parallel to the relative motion and slide between the two surfaces [16]. The addition of 0.025 wt. % graphene was considered as the optimum concentration that demonstrated the best performance by providing the lowest COF of 0.0332, which improved COF more than 62% compared to SE. This scenario proved that graphene has a two-dimensional structure that provides ease of movement of particles to slide between the two mating surfaces, hence enhanced the tribology behaviour in terms of friction and wear. However, a higher concentration of graphene caused some coagulation, thus promoting poor frictional properties due to the unstable friction, and also exhibited vibration in the friction zone [11]. Furthermore, the trend of hBN was slightly different from graphene nanofluids as the lowest concentration (0.01 wt. %) at MJOh1 recorded the lowest COF of 0.0509 compared to MJOh2 and MJOh3. In the study by Talib *et al.,* [7], it was shown that the abrasive particle was considered at a high concentration of hBN as it could damage the worn surface due to plastic deformation.

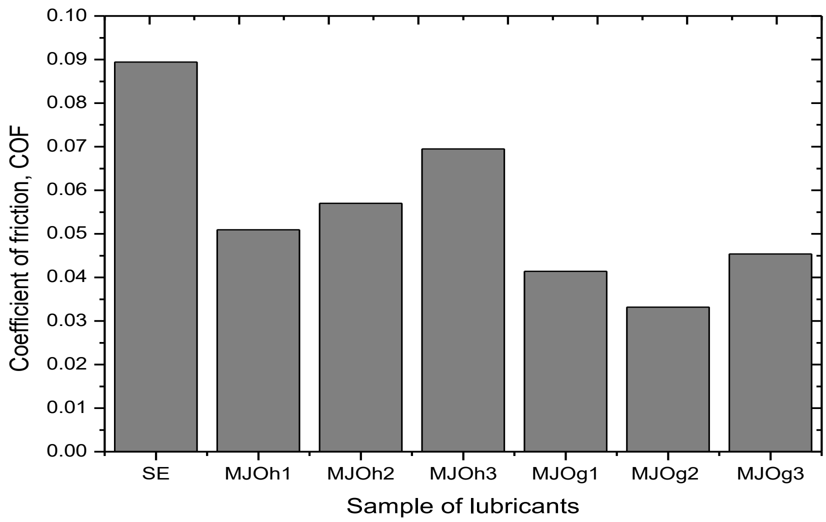

Fig. 4. Variation of coefficient of friction for lubricant sample

3.2.2 Wear scar diameter

Figure 5 illustrates the variation of MWSD of the SE and MJO samples. The MWSD represents the damage occurred caused by the material removal during the sliding contact between the two surfaces. It was found that SE had the highest MWSD of 940.87 µm among other samples. These results proved that very high viscosity of SE produced worse worn surface. It indicated that MJO samples provided a better lubrication film compared to SE. This is due to the formation of TMP ester as longer fatty acid chains of TMP ester tended to increase the adsorbed film thickness and provided higher protection to the contact surface. Additionally, as the binding of the molecules became greater, the resistance to shear forces was greater [17]. The presence of hBN and graphene nanoparticles in MJO improved the MWSD by 38% to 52% compared to SE. MJOh2 exhibited the smallest MWSD among MJO samples with the value of 443.35 µm. The inclusion of 0.025 wt. % hBN nanoparticles in MJO was sufficient to fill the asperity valleys of the contacting surface, hence resulting in the reduction of WSD. However, MJOh1 that contained the lowest amount of hBN nanoparticles possessed higher MWSD of 552.13 µm than MJOh3 with a higher concentration of hBN. In contrast to MJO with graphene particles, the addition of 0.025 wt. % graphene in MJO (MJOg2) exhibited the highest MWSD of 581.56 µm compared to MJOg1 and MJOg3. This finding indicated that the smooth worn surface and lower frictional force obtained is because the nanoparticles had filled the valleys and the shearing of trapped nanoparticles at the interface of the contacting surfaces [18].

Figure 6 presents the surface morphology of the worn surface of the lubricant sample taken using an optical microscope at 5× magnification. MJOh2 had a small round shape of MWSD because it provided sufficient lubricant film to protect the surface area. The worn surfaces for MJOh1, MJOh2, and MJO3 were smoother compared to MJOg1, MJOg2, and MJOg3. However, there was some material transfer on the worn surface of MJOh1. This was because the sedimentation and re-agglomeration of nanoparticles occurred due to the lower viscosity [18]. The results for graphene nanofluids of MJO showed the formation of deep parallel grooves (deep region) as graphene contained high percentages of carbon element compared to hBN.

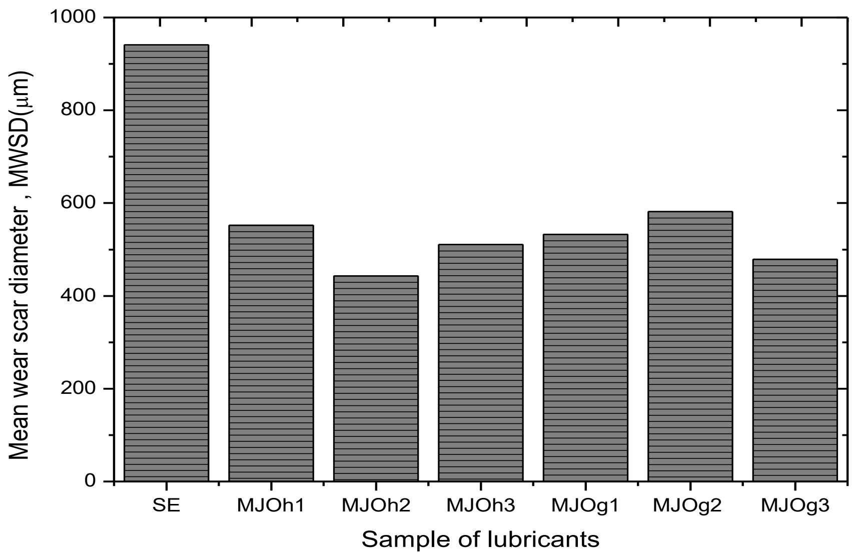

Fig. 5. Variation of mean wear scar diameter of lubricant sample

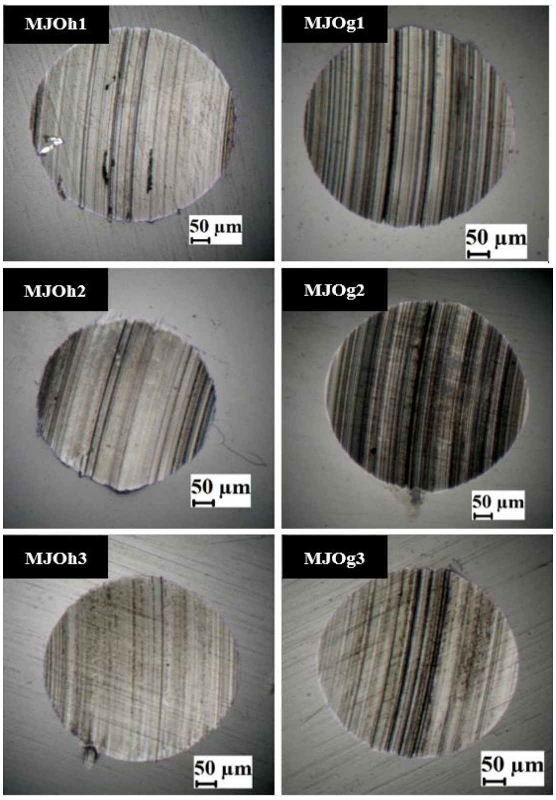

Fig. 6. Worn surface of lubricant sample using optical microscope at magnification 5x

3.2.3 Friction torque

Figure 7 displays the result of the friction torque of the lubricant samples. The friction torque had similar trends as the results of COF calculated using Eq. (1). The data showed that the SE had the maximum friction torque of 0.1575 Nm. MJO samples demonstrated lower friction torque compared to the SE, where MJOg3 had the lowest friction torque among the MJO samples at 0.0585 Nm, as well as the lowest COF. A slight increase of friction torque was shown by MJO with hbN nanoparticles. MJOh1 exhibited the lowest friction torque of 0.0897 Nm compared to MJOh2 and MJOh3. This proved the ability of hBN particles acting as a friction reducer at a lower concentration [13]. Talib *et al.,* [7] emphasised that a higher concentration of hBN particles broke down lube boundary and exhibited inadequate prevention towards friction, thus produced high friction torque and also COF.

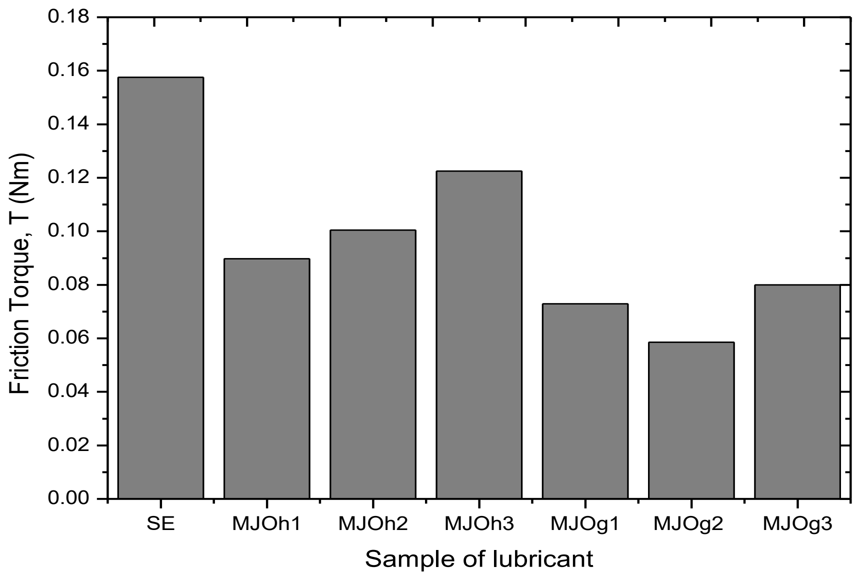

Fig. 7. Variation friction torque of lubricant sample

4. Conclusions

In this study, the relative performance of seven MJO samples as a sustainable lubricant was examined by performing physicochemical properties and tribology tests using four-ball test and the performance was compared with the SE. The presence of hBN and graphene nanoparticle additives significantly improved the physicochemical properties. The inclusion of 0.05 wt. % of hBN and graphene in MJO samples (i.e., MJOh3 and MJOg3) exhibited the highest viscosity index of 267 and 269, respectively. The addition of 0.025 wt. % graphene in MJOg2 produced the optimum concentration that provided the lowest COF among other samples. However, MJOh2 that contained 0.025 wt. % hBN demonstrated the smallest MWSD in comparison to the SE and MJO samples. Based on the overall result, the addition of nanoparticle additives significantly enhanced the tribological characteristics and the modified MJO samples were highly suitable as substitutes for SE.

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