Jurnal Tribologi 43 (2024) 71-85

MYTRIBOS Malaysian Tribology Society
Jurnal Tribologi
Website : jurnaltribologi.mytribos.org e-issn: 2289-7232

Tribological evaluation of a novel scallop-shell $CaCO_3$ additive for extracted banana peel oil lubricant

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KEYWORDS

$CaCO_3$
Banana peel
Bio-based oil
Nanoparticles
Tribology

ABSTRACT

Nowadays, many sectors use lubrication derived from petroleum oil, resulting in severe environmental pollution. Hence, as a countermeasure, developing lubricants from organic sources is gaining attention. This research aimed to formulate a new bio-based banana peel oil by adding scallop shell nanoparticles (SSNs) as an additive. The process involved extracting banana peel oil (BPO) using a Szf-06A Soxhlet extraction fat analyser. The extracted oil was mixed with 0.5 vol% of three types of SSNs as an additive, and the mixture was then homogenised using ultrasonication. A tribological test using a four-ball tester, according to ASTM D-4172, was performed to determine the coefficient of friction (CoF) and the wear scar diameter (WSD). Scanning electron microscopy (SEM) was conducted to verify the wear mechanism. Samples with commercial-sized SSN additives yielded the best CoF (0.08385) and WSD (0.67 mm) results, indicating that commercial SSN additives can improve the physical properties and enhance the excellent anti-wear and anti-friction characteristics of lubricant mixtures compared to mineral oil.

Received 17 July 2024; received in revised form 23 September 2024; accepted 3 November 2024.
To cite this article: Mohamad Fazdhli et al., (2024). Tribological evaluation of a novel scallop-shell $CaCO_3$ additive for extracted banana peel oil lubricant. Jurnal Tribologi 43, pp.71-85.

1.0 INTRODUCTION

Lubricating oil is widely used in aviation, automobile, mechanical processing, transportation, metallurgy, coal, construction, and light industries. Traditional lubricating oils provide benefits such as excellent lubrication, resistance to high temperatures, and protection against corrosion (Hamnas & Unnikrishnan, 2023). Lubricants play a critical role in the industrial sector by reducing friction and wear, enhancing heat transfer, mitigating impurities, and prolonging the life of machines (Quaranta, 2023). Approximately 50% of the mineral lubricants used in automobiles ultimately enter the environment and cause irreversible damage to the ecosystem on coming into direct contact with water and soil. Thereafter, they destroy the ecological balance in the environment and cause severe contamination of soil and water resources (Wathi et al., 2023). Traditional mineral-based lubricants are not entirely biodegradable and are highly toxic compared to bio-based options from plant oils (Hassan et al., 2022). In addition, the main concern with petroleum-based lubricant oils is their potential to release harmful volatile organic compounds (VOCs) into the air. Among the primary VOCs released during the refining of crude oil are benzene, toluene, ethylbenzene, and xylene. These VOCs have been linked to harmful health outcomes, including cancer, respiratory problems, and neurological damage (Rajabi et al., 2020). Therefore, the search for novel bio-derived oils is essential in fostering the sustainability, mitigating environmental damage, and propelling the progress of eco-friendly lubricant alternatives (Mortier et al., 2010).

The use of fruit oils as a bio-lubricant has a long history dating back to ancient times, with sesame and olive oils being used as lubricants in India and Rome, respectively. Vegetable oils are used as lubricants, but their use is limited due to poor performance and stability at high temperatures (Erhan et al., 2006). Compared to traditional lubricants, vegetable oils have weaker tribological characteristics and lower thermal and oxidative stabilities, making them unsuitable for use as lubricants (Uppar et al., 2023). However, with the growing demand for sustainable alternatives, there has been a renewed interest in the use of fruit oils such as palm, coconut, and castor oils. Today, fruit oils are widely used in the production of biological lubricants. An example of a fruit oil is banana peel oil (BPO), which is eco-friendly due to its excellent lubricity, renewability, biodegradability, and low toxicity (Masripan et al., 2020). Fruit oil is oil that is extracted from the fleshy part of a fruit. Fruits such as palm, coconut, olive, avocado, and others are familiar sources of fruit oils. These oils are extracted from the fruit using various methods, such as cold-pressing or solvent extraction.

Banana peel oil (BPO), also known as isoamyl acetate, has a high concentration of fatty acids, free sterols, sterol esters, and steryl glucosides, which are found in banana fruit species such as the 'Dwarf Cavendish', making BPO a promising lubricant (Oliveira et al., 2008). Moreover, the improved properties of BPO, such as its density, viscosity, fire point, pour point, and flash point, make it an eco-friendly lubricant substitute that works well for machining tasks (Deshpande & Jyothi, 2022). Banana peel oil (BPO) is biodegradable and non-toxic, making it an environmentally friendly alternative to traditional petroleum-based lubricants. In recent years, BPO has demonstrated its effectiveness as an additive for extreme pressure and anti-wear purposes in lubricants, making it suitable for gear and engine oils (Masripan et al., 2019).

Aside from bio-based oils, nanoparticles are also beneficial in enhancing the tribological properties of base lubricants by enhancing the viscosity and forming chemical bonds with the oil (Singh et al., 2019). Along with improving severe pressure, anti-wear, and friction-reducing qualities, they also form protective layers that help to extend the life and effectiveness of the machine (Patil et al., 2014). In addition, studies by Rahmadiawan et al. (2023) reported that nanoparticles, like MXene, can form a protective film on surfaces to reduce direct contact between moving parts. This film acts as a barrier, minimising wear and friction during mechanical operations, thereby improving the coefficient of friction (CoF) by 25% compared to lubricated water. Moreover, nanoparticles have good sliding and delamination properties, which can also reduce friction and wear, thereby improving the lubrication effect (Khan et al., 2019). Therefore, nanoparticles can be used as additives to improve the performance and efficiency of bio-lubricants by enhancing their lubrication properties and reducing friction and wear.

In 2021, the lubricant market worldwide was valued at US$164.9 billion and was projected to grow steadily at a rate of 4.0% annually to reach US$205.9 billion by 2028. Bio-lubricants were worth around $2.92 billion in 2021, and this figure is anticipated to grow at a rate of 4.7% annually, reaching approximately $4.26 billion by 2029 (Moses et al., 2023). The global bio-based lubricants market is expected to grow significantly due to the rising demand for eco-friendly lubricants in the automotive industry (Syahir et al., 2017). While bio-based lubricants have many benefits in terms of the environment and sustainable development, they may only perform as well as traditional lubricants in specific applications. Hence, this can be a barrier to adoption, particularly in high-performance industries such as the aerospace and automotive industries.

Adding scallop shell nanoparticles (SSNs) could significantly improve the performance of lubricating oils. This is mainly due to the chemical components of scallop shell powder, which consists of calcium oxide (CaO), silicon dioxide (SiO_2), aluminium oxide (Al_2O_3), and iron (III) oxide (Fe_2O_3). Calcium oxide (CaO) makes up 53.72% of scallop shells (Her et al., 2021), and it can be used as an additive to improve the wear resistance of lubricating oils, but not to enhance their anti-friction performance (Arinbjarnar et al., 2022). Smaller particles can be used to stabilise the contact interface and have the potential to form a protective film (Rahmadiawan et al., 2023). Hence, SSNs can be used as an additive for bio-lubricants to improve their performance and make them more efficient by enhancing their lubricating properties.

As research is continuing on the use of BPO infused with nanoparticles as a lubricant, there are still insufficient studies on BPO with SSNs. Therefore, this study aimed to investigate the potential of a newly developed lubricant from a renewable energy source. The study focused on the tribological performance of the newly developed BPO. The addition of SSNs is expected to boost the performance of the base oil, hence contributing to the discovery of a new lubricant that can achieve better lubricity performance.

2.0 EXPERIMENTAL PROCEDURE

The study began with the extraction of BPO and included several other methods, such as the infusion of SSNs into BPO, four-ball testing, and the verification of wear mechanisms using scanning electron microscopy (SEM).

2.1 Extracting the Banana Peel Oil (BPO)

Ripe banana peels were collected from stalls as agro- or domestic waste. These were washed under clean water and air-dried until a constant mass was recorded. Once dried, the peels were crushed using a pestle and mortar to provide a larger surface area for more efficient oil extraction. The specimen was then extracted by means of a Szf-06A Soxhlet extraction fat analyser (Figure 1), and hexane was used as a solvent for the extracted BPO. The extraction process involved breaking down the banana peels and separating the oil or aromatic compounds from the solid material. Soxhlet extraction is a time-honoured method in which a sample is placed in a thimble holder that is progressively filled with fresh concentrated solvent from a distillation flask. The entire extraction process took two hours, after which, the specimen was removed from the Soxhlet extraction machine and cooled for 30 minutes. This resting process was to ensure that the sediment was left at the bottom of the bottle and only the liquid sample was taken. The specimen was then prepared for the second bathtub to extract the oil from the hexane mixture. Finally, pure BPO was obtained.

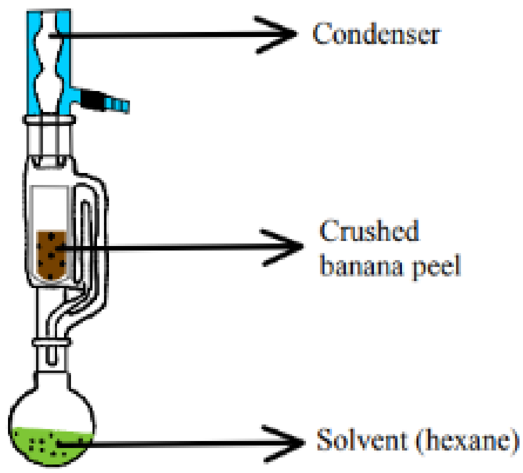

Figure 1: A diagram of a Szf-06A Soxhlet extraction fat analyser setup.

2.2 Preparing the Scallop Shell Nanoparticles (SSNs)

The preparation of SSNs involved the process of synthesising or creating nanoparticles from scallop shells. Scallop shell powder is comprised of CaO, SiO_2, Al_2O_3, and Fe_2O_3. Calcium oxide (CaO) is the primary constituent of scallop shell powder (53.72%) (Her et al., 2021a) (Table 1). The crystallite size of the calcium carbonate ($CaCO_3$) powder was identified as 52.18 nm. This was strongly supported by the Fourier-transform infrared (FTIR) spectroscopy and SEM results (Figures 2 & 3), which showed the crystallite structure of the $CaCO_3$ and the absorption peaks of the pure $CaCO_3$ at 881.46 and 713.66 cm^{-1} (Puspitasari et al., 2024).

Table 1: The chemical composition of the SSNs.

Chemical Components	Value (%)
CaO	53.72
SiO_2	0.28
Al_2O_3	0.12
Fe_2O_3	0.02

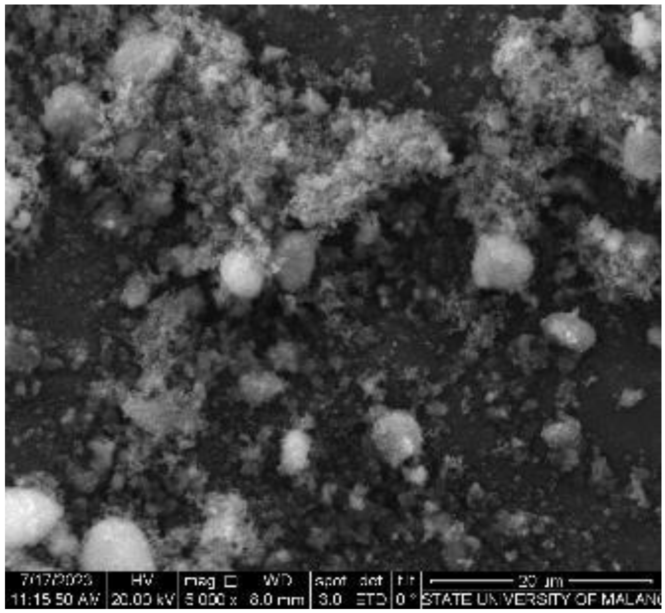

Figure 2: The crystallite structure of the $CaCo_3$ nanoparticles.

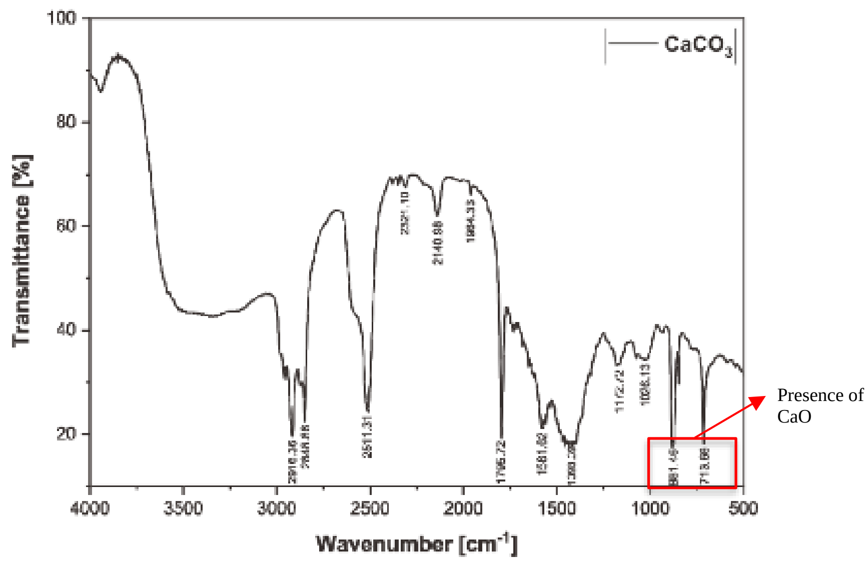

Figure 3: The FTIR of the $CaCo_3$ nanoparticles.

The detailed flow of the preparation of the SSNs is shown in Figure 4. The scallop shells were collected from the shore or obtained from a seafood processing plant. These were then cleaned and ground for five hours using the ball milling method before being calcinated for 60 minutes at 110°C. This process of calcinating at a high temperature was to remove any organic matter and convert it into CaO. Next, the ground scallop shells were subjected to a milling process to obtain micro-sized particles or nanoparticles. A surface modification process can be performed by treating the nanoparticles with surfactants or by conducting chemical modifications. This process helps to improve their compatibility and dispersion in extracted oils. Lastly, the scallop shells underwent an oil-mixing process, where the modified SSNs were added to the extracted BPO.

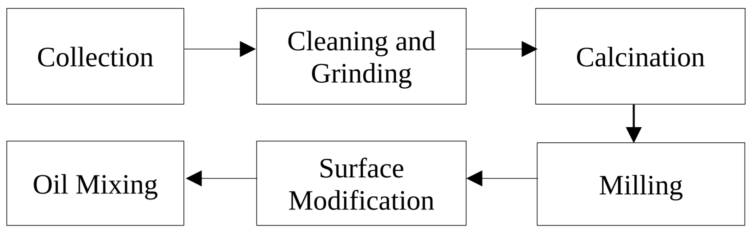

Figure 4: A diagram of the preparation of the SSNs.

2.3 Mixing the Sample Oils and The Ultrasonication Process

The homogenisation of SSNs involved the process of adding the tiny scallop shell particles to the bio-lubricant oil formulation. The SSN concentration was 0.05 vol%, which has been proven to be the optimal composition for the mixing of nanoparticles with oil (Abdullah et al., 2014). The required weight of the SSNs was calculated using the density equation, given that the density of the SSNs was 0.26 g/cm³.

$$\rho = \frac{m}{v}$$

$$\rho = Density\ (0.26\ g/cm^3)$$
$$m = Mass\ (g)$$
$$v = Volume\ (ml)$$

Samples B, C, and D were prepared by dispersing 0.05 ml of the SSNs in 9.95 ml of BPO, while for Sample A, which consisted of 10 ml of 100% pure BPO, no SSNs were used. Sample A was set as the benchmark for the other samples. The four samples, A, B, C, and D, that were prepared, are shown in Table 2.

Table 2: The different sizes of the SSN additives

Sample	Quantity (ml)	
	BPO	**SSN Size**
A	10	0
B	9.95	0.05 (Micro $CaCO_3$)
C	9.95	0.05 (Nano $CaCO_3$)
D	9.95	0.05 (Commercial $CaCO_3$)

The mixture of nanoparticles and BPO was homogenised for 30 minutes using an ultrasonic homogeniser (Figure 5) (Abdullah et al., 2014). An ultrasonic homogeniser was used in the incorporation process to reduce the small particles in the liquid. The frequency and duty cycle were set at 60 Hz and 0.5, respectively. An MS 72 micro tip probe size was used to prevent changes in the properties of the oil due to the rise in temperature. Thermocouples were also used to ensure that the temperature of the oil remained within 60–70°C during the homogenising process. The oil mixture was placed inside a water bath with water and ice to control the temperature of the water, thereby preventing a spike in the temperature. The ultrasonication process served to improve the stability and uniformity of the mixture.

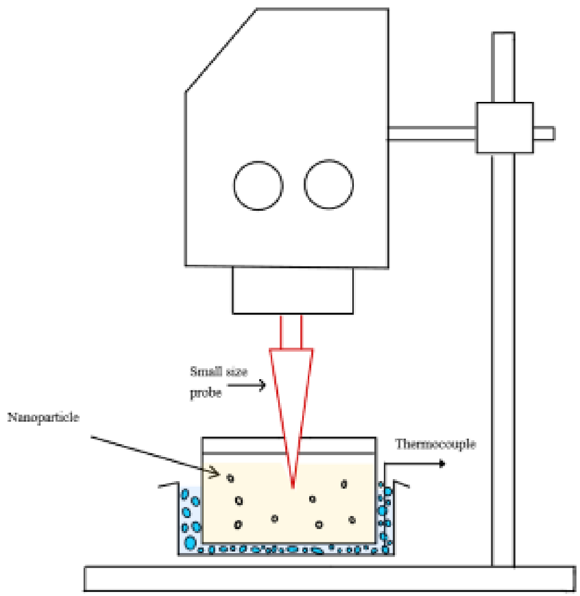

Figure 5: A schematic of the sample preparation using the ultrasonic method.

2.4 Four-Ball Tester

This test used the ASTM D4172 as the designated test technique to determine the CoF and the wear scar diameter (WSD). According to the ASTM D4172, the parameters to be used were a rotational speed of 1200 rpm, a load of 392.4 N, a duration of 3600 seconds, and a temperature of 75°C. The four-ball test involved arranging carbon chromium steel balls in a triangular pattern (Figure 6), while a fourth steel ball was placed against these with a constant force and rotational speed. The test chamber was filled with 10 ml of BPO enhanced with SSNs, completely submerging the three fixed balls. The fourth ball was rotated in contact with the three balls immersed in BPO under specified conditions and time. During the test, the revolving ball generated friction and scars on coming into contact with the fixed balls. The CoF was measured using a data terminal processing device.

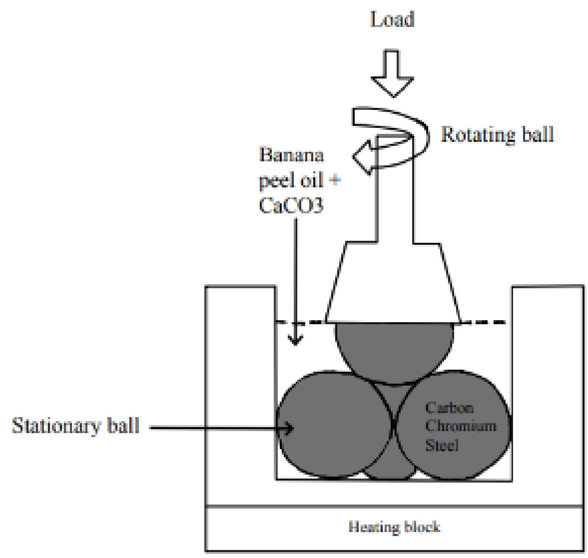

Figure 6: A schematic diagram of the ball pot assembly.

2.5 Scanning Electron Microscopy (SEM) Surface Analysis

After the four samples of BPO with various particle-sized additives had been subjected to the four-ball test, a wear scar emerged on the surface of the steel balls. The WSD was determined by measuring the scar diameter on the three fixed steel balls immersed in the BPO. The steel balls that encountered each other were left with a spherical-shaped scar. Firstly, the pre-image of the scar was measured using a portable mini-electronic microscope connected to a computer. Then, the image of the wear was captured using dimension software to identify the WSD.

Next, SEM was employed to analyse the morphologies of the surfaces of the worn steel balls. The SEM micrographs precisely depicted the centre of the worn scar on the disks, thus assessing the anti-wear capability of the oil sample.

3.0 RESULTS AND DISCUSSION

The data were analysed for the effect of the four-ball tribo-tester on the performance of the extracted BPO infused with SSNs. The subsequent sections present the findings for the CoF, WSD, and SEM.

3.1 Effects of The Scallop Shell Nanoparticle (SSN) Additives on The Coefficient of Friction (COF)

Table 3 shows the average CoF of Sample A and Samples B to D with different sizes of SSN additives. Figure 7 shows the four-ball tribo-testing results. It can be observed that Sample D had the best CoF result of 0.08385, while Sample A had the worst CoF result of 0.13794. In addition, the standard deviations for Samples A, B, C, and D were 0.0124, 0.0096, 0.0056, and 0.0036, respectively. This shows that the performance of Sample D was stable across the steady state. According to Hamid et al. (2020), a lower CoF is beneficial for the proper operation of equipment and to minimise energy loss. Figure 8 displays the overall test results for all the samples. It was shown that the extracted BPO with the addition of $CaCO_3$ had the potential to reduce the unsteady state of the CoF at an early stage. The extracted BPO with the addition of micro and nano $CaCO_3$ particles showed insignificant differences, probably because the powder was produced in the laboratory. Nanoparticles synthesised in the laboratory may exhibit drawbacks in terms of variations in particle size. This can be seen in Figure 7, where there was a sudden fluctuation in the steady state area, indicating the non-homogeneity of the nanoparticles. Fluctuations in particle size can affect the characteristics and effectiveness of nanomaterials, resulting in irregular behaviour (Magdalena & Gabriela, 2020). The BPO with commercial SSNs presented the best result among all the other three samples. This result concurred with the finding of Guo et al., (2021) that commercial SSNs are stable and well-suited for oil additives due to their biocompatible properties, resistance to acid degradation, and ability to form Pickering emulsions.

Table 3: The average CoF of the BPOs with different sized SSN additives.

Sample	$CaCO_3$	Average CoF
A	-	0.13794
B	Micro	0.10102
C	Nano	0.10176
D	Commercial	0.08385

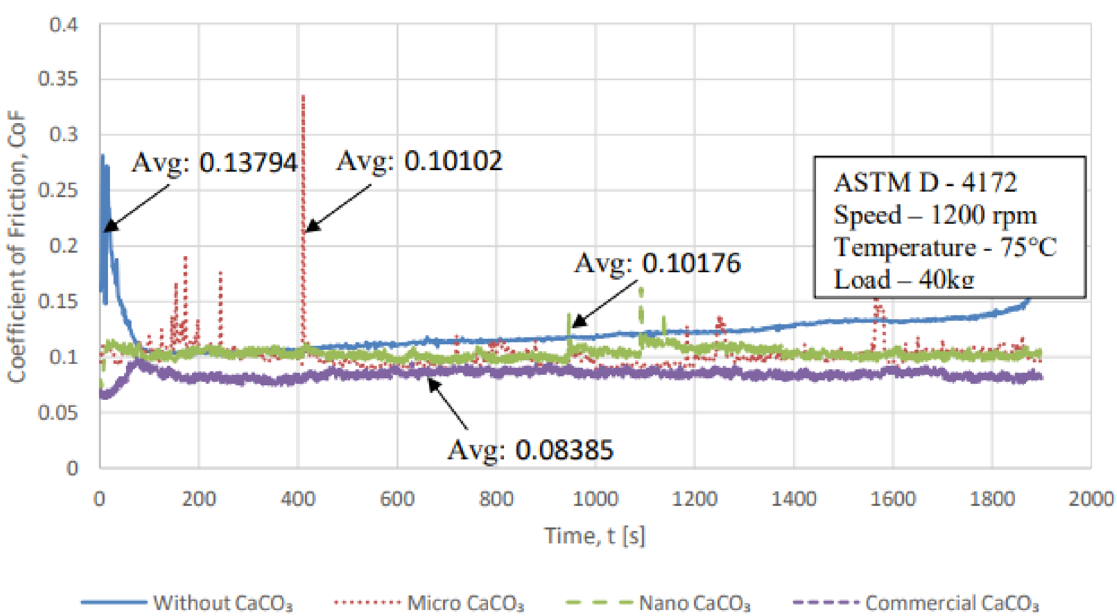

Figure 7: The CoF of the BPOs with different sized SSN additives.

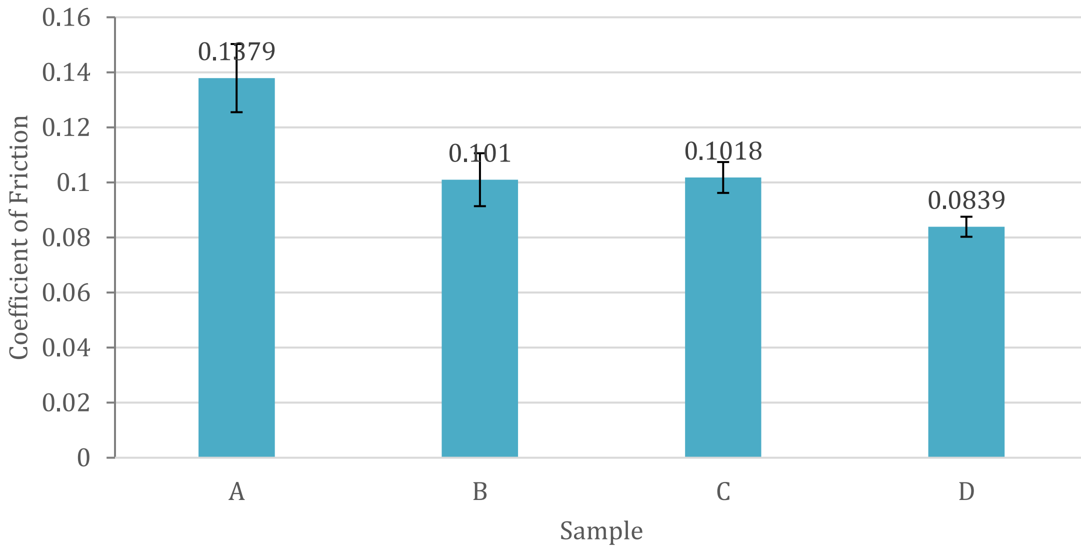

Figure: 8 A comparison of the CoF of the BPOs with different sized SSN additives.

3.2 Effect of The Scallop Shell Nanoparticle (SSN) Additives on The Wear Scar Diameter (WSD)

The WSD measures the anti-wear property of lubricating oils. A lower value of WSD indicates better lubricant performance. The result of the proposed SSN concentration blended with BPO was anticipated. The scar diameters of all the three balls were examined under a microscope following the test. Table 4 shows the average WSD for each additive used.

Table 4: The WSDs of the BPOs with different sized SSN additives

Sample	SSN Size	Average WSD (mm)
A	-	1.47
B	Micro	0.77
C	Nano	0.69
D	Commercial	0.67

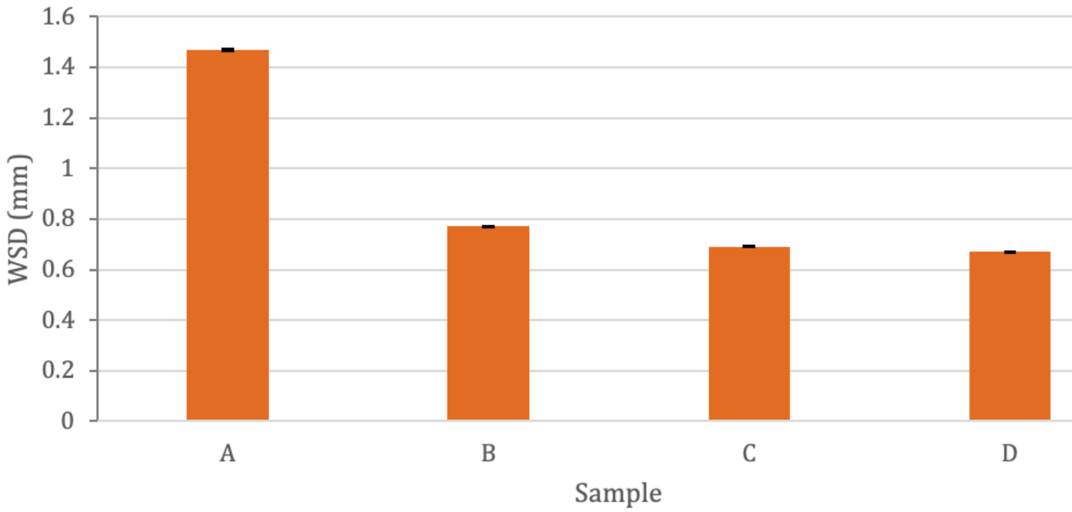

Figure 9: A comparison of the WSDs of the BPOs with different sized SSN additives.

As shown in Figure 9, Sample D performed better than the other samples as it had the lowest WSD of 0.67. This indicated the significant impact of commercial SSNs in improving the WSD by 48% compared to Sample A. Sample B had a higher WSD due to its smaller particle size compared to Samples C and D. As stated by Rhadiawan et al. (2024), smaller nanoparticles may not possess the load-bearing capacity required to provide efficient protection in situations of high-stress conditions. In addition, the higher WSD of Sample B could also be attributed to the sudden spike in the CoF in the steady-state region (Figure 7), which contributed to the slightly higher WSD than in Samples C and D. Although there were no significant differences between Samples C and D, Sample C did not provide as consistent a performance as Sample D in terms of the standard deviations of the CoF, which were 0.0056 and 0.0036, respectively. This strongly suggested that Sample D gave a better performance than Sample C. The improved performance of Sample D was due to the high percentage of $CaCO_3$ (53.72%) (Her et al., 2021b). The presence of $CaCO_3$ enabled a film to be formed on the rubbing surface, which suggests that the bio-based performance of BPO enhanced with commercial-sized SSNs can improve the tribological properties of an oil.

3.3 Surface Analysis of The Different Sized Scallop Shell Nanoparticle (SSN) Additives

Figure 10 shows the SEM analysis of balls in Samples A, B, C, and D. This result was obtained using SEM images magnified by 50X and 246X. Most of the resultant images showed abrasive wear during the test. The SEM micrographs in these figures precisely depicted the centre of the worn scar on the disks. These micrographs were used to assess the anti-wear capability of the oil samples.

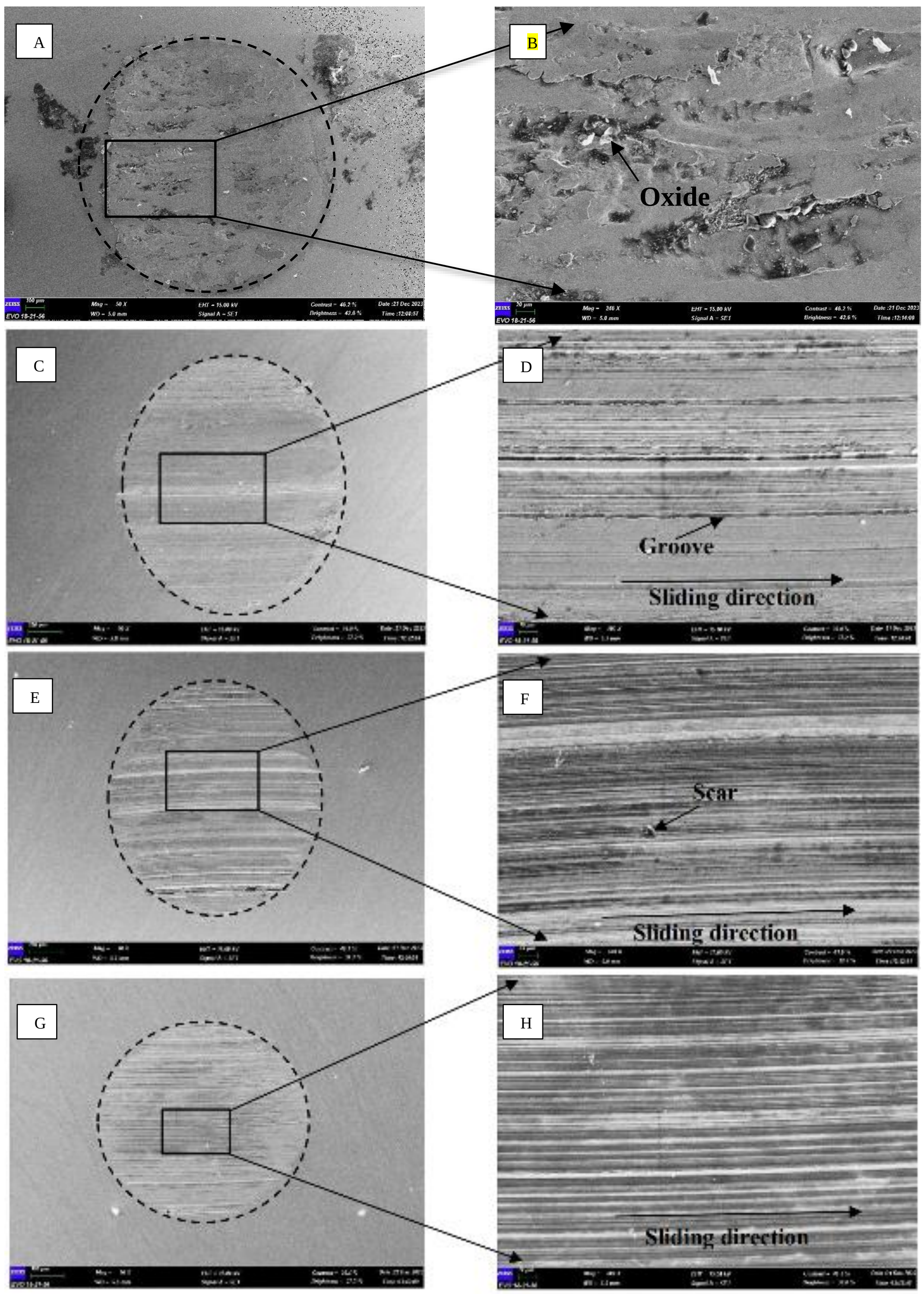

Figure 10: SEM balls image: (A&B) Sample A, (C&D) Sample B, (E&F) Sample C, and (G&H) Sample.

The SEM images of the ball from Sample A are displayed in Figures 10A and 10B. The surface of the steel ball was covered with a large amount of dirt and scars, suggesting that Sample A lacked significant lubricating qualities. These scratches and debris that appeared on the surface of the steel ball were due to increased friction, inadequate wear protection, and reduced heat dissipation capabilities. This resulted in increased mechanical stress and surface damage. The SEM images of the ball from Sample B are shown in Figures 10C and 10D. Sample B performed better in lubrication than Sample A because the surface of the steel ball had less dirt and scars than that of Sample A. This was because the additive was beginning to form a lubricating film over the surface. Figures 10E and 10F show the SEM images of the ball from Sample C. Compared with the surface of the steel ball from Sample B, the surface of the steel ball from Sample C had less debris and scars. This was because the nano-sized particles of the additive were smaller than the micro-sized particles. The smaller-sized particles were able to penetrate the microscopic gaps and irregularities in the contacting surfaces more effectively. Figures 10G and 10H show the SEM images of the ball from Sample D. Compared with the wear surfaces of the other oil samples, the wear surface of Sample D showed fewer signs of severe wear and was the smoothest and flattest, indicating that Sample D had the best lubrication properties.

In summary, the lubricating performance of commercial additives is better than that of nano- and micro-additives despite their bigger size. This could be because larger-sized additives offer more robust film formation and are better load carriers, particularly in heavy duty applications. Sample D was the most efficacious, with Sample B, C, and D demonstrating progressively improved lubrication, while Sample A performed the worst.

CONCLUSIONS

The samples with commercial SSN additives showed great potential to be developed as new bio-lubricants with the best CoF (0.08385) and WSD (0.67mm). Consequently, the SSNs displayed improved physical properties, enhancing the excellent anti-wear and anti-friction characteristics of the lubricant mixtures compared to mineral oils. Even though the characteristics of the BPO are still anticipated, it is highly recommended that BPO be subjected to physicochemical tests to further study it as a lubricating oil. In addition, further research is needed using statistical methods to ensure BPO with SSN additives are perfectly analysed and optimised. The limitation in terms of nanoparticle agglomeration in bio-based lubricant oils is also anticipated, with the idea that a surfactant can potentially solve this problem. Overall, this research has demonstrated that the addition of 0.5 vol.% of SSNs to BPO can effectively enhance its tribological qualities by reducing the WSD and CoF.

ACKNOWLEDGEMENT

The study is funded by the Ministry of Higher Education Malaysia through the Fundamental Research Grant Scheme (FRGS), No: FRGS/1/2023/TK10/UTEM/02/10. This study is also funded by Universiti Teknikal Malaysia Melaka and Universitas Negeri Malang, Indonesia, through the University Research Matching Grant (URMG), No: ANTARABANGSA (URMG)-UM/2023/FTKMP/A00057.

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