Jurnal Tribologi 44 (2025) 150-166

MYTRIBOS
Malaysian Tribology Society

Jurnal Tribologi
Website : jurnaltribologi.mytribos.org
e-issn: 2289-7232

Influence of propylene glycol on the tribological performance and lubrication mechanism of water-based lubricants incorporating silicon carbide, magnesium oxide and aluminium oxide nanoadditives

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KEYWORDS

Nanolubricants
Dispersants
Tribological characteristics
Lubrication mechanism
Film thickness

ABSTRACT

Water-based lubricants (WBLs) offer cooling capabilities and are environmentally friendly compared to oil-based lubricants, though they have low viscosity and can result in very thin film thicknesses. This study evaluates the effects of propylene glycol as a dispersant on the dispersion stability of various nanopowder additives, specifically magnesium oxide (MgO), aluminium oxide (Al_2O_3), and silicon carbide (SiC). The physical, mechanical, and tribological properties of the formulated WBLs were examined. Tribological assessments were performed using block-on-ring setup following ASTM 77-98 standards under different rotational speeds and contact loadings. Results shows that addition of propylene glycol improves the viscosity of WBLs with MgO have the highest viscosity of 26.32 mPa·s compared to water alone. MgO/SiC was proven to have the lowest frictional coefficient of 0.1802, as well as lowest wear loss of 0.1118% which makes it the best lubricant in this study. The study found that combination of nanoadditives with propylene glycol is an effective solution to improve the tribological performance of lubricants which potentially led to an environmentally sustainable lubricant.

Received 8 August 2024; received in revised form 12 December 2024; accepted 9 January 2025.
To cite this article: Azizan et al., (2025). Influence of propylene glycol on the tribological performance and lubrication mechanism of water-based lubricants incorporating silicon carbide, magnesium oxide and aluminium oxide nanoadditives. Jurnal Tribologi 44, pp.150-166.

1.0 INTRODUCTION

The main challenge for researchers in tribology is to reduce the friction and wear between two contact surfaces. The most commonly used are oil-based, which offer advantages such as ability to withstand high pressure, speed and temperature due to their high viscosity properties, which are important for improving tribological performance (Rawat et al., 2019). However, drawbacks such as difficulties in post-cleaning and removal due to the non-biodegradable nature of oil, the consideration of non-biodegradable additives that release toxic materials and the projected depletion of petroleum products within the next 50 years may raise concerns regarding the use of oil-based lubricants (Rajak & Menezes, 2021; Zhou et al., 2022). Therefore, water-based lubricants (WBLs) have been developed to address these issues. WBLs offer several advantages, including cost savings, excellent cooling capacity, superior thermal conductivity, and eco-friendliness (Morshed et al., 2021; S. Wang et al., 2016). Water-based lubricants have been applied in various applications, such as drilling operations which helps to cool the drill bit and carry away the cuttings from drilling site (Wang et al., 2020). WBLs also served as a cutting fluid in cutting and machining operations to help cool the cutting tool and workpiece. The use of water-glycol mixtures has been shown to improve the tool life and surface finish while being more environmentally friendly compared to petroleum-based cutting fluids (Wu et al., 2017). Additionally, water-based lubricants effectively dissipate heat, enhancing the longevity of tools and machinery (Wang et al., 2012; Ding et al., 2017). For instance, water-glycol hydraulic fluids are commonly used in hydraulic systems due to their ability to maintain operational efficiency while providing fire resistance (Wang et al., 2012; Wang et al., 2020). However, WBLs exhibit poor performance in the reducing the friction due to low viscosity and can be corrosive to metal surfaces which leads to rust (Tomala et al., 2010). Thus, to overcome the problems faced, application of nanopowder additives and surfactants were required in improving the WBLs tribological performances.

To enhance the properties of lubricants, certain additives are required to ensure an effective lubrication mechanism. The addition of nanopowders has been proven to improve the tribological properties by penetrating small gaps on surface (Alazemi et al., 2015). Nanopowders may induce lubrication of mechanism such as thin film formation, rolling effects, bearing effects and polishing effects. The combination of different nanopowders additives in lubricants can also help overcome the limitations For example, Azizan et al., (2024) demonstrated that MgO/SiC is capable to improve the formation of film thickness in lubrication. Additionally, surfactants can be incorporated into the formulation of lubricants to improve the properties and provide a stable nanopowders dispersion in the lubricants. Surfactants can be classified into various types such as cationic, anionic and non-ionic which plays an important role in improving viscosity and improving surface tension which helps in improving the lubrication and dispersion of nanopowders (Huang et al., 2021). Haribabu et al., (2023) demonstrated the use of surfactants by dispersing graphene nanopowders in different concentrations of water/propylene glycol, concluding that propylene glycol improves the dispersion stability of nanopowders in lubricants, as well as reducing the coefficient of friction.

This study focused on the lubrication effects of various nanopowders additives in water-based lubricants, which were tested on a block-on-ring test under different rotational speeds and contact loadings. In contrast of previous research done by Rui et al., (2023) which focused on the addition of polyvinylpyrrolidone (PVP) in formulating WBLs, this current research investigates the impact of propylene glycol on the tribological performance and lubrication mechanism of WBLs incorporated with SiC, MgO and Al_2O_3 nanopowder additives, employing different variables which allows more nuanced understanding of the tribological properties and lubrication mechanisms.

2.0 MATERIALS & METHODS

Deionized water was used as a base fluid in the formulation of water-based lubricants. Polyvinyl glycol obtained from Jasa Chemi Sdn. Bhd. was used as a surfactant in order to improve the viscosity and ensure a stable dispersion of lubricants. MgO and Al_2O_3 were obtained from Jiangu XFNano Materials Tech Co., while SiC were obtained from Xi'an LY Health Technology Co. MgO and Al_2O_3 nanopowders can be categorized as metal oxides, were known for its ability to withstand high temperature, and good mechanical strength with advantage on Al_2O_3 to possess excellent mechanical strength hardness and toughness (Costa et al., 2020). On the other hand, SiC which is a metal carbide known for their frictional behavior which provides tribo-corrosive aspects from their abrasive properties of nanopowder due to high hardness, wear resistance and durability, also possess high thermal conductivity. These nanopowders were added into the lubricants in a single and hybrid mixture of MgO/SiC, Al_2O_3/MgO, Al_2O_3/SiC and Al_2O_3/MgO/SiC in order to observe either improvements or drawbacks from different properties possess by the nanopowders. The content for each test lubricants formulated were listed in Table 1.

The water-based lubricants were formulated via two-step method (Opia et al., 2024) as shown in Figure 1. The formulation of lubricants involved mixing DI water with propylene glycol using a magnetic stirrer at 500 rpm, followed by addition of nanopowders into the solution. The mixture then underwent vigorous stirring using a high-speed homogenizer at 10000rpm to remove any agglomerations of nanopowders in the lubricants.

The tribological properties of the lubricants were studied on a block-on ring system following the ASTM G77-98 with the setup were illustrated in Figure 2. Table 2 and 3 describes the test parameters and properties of block and ring used when the test being performed. Before the test began, the block and ring were cleaned using acetone to remove any oil, dust or dirt on the surface and were weight prior the test. During the test, the speed and temperature were recorded while the contact loading were kept constant for each test. For each test done, excess lubricant remaining will be disposed and refilled with a new lubricant, block and ring for each test lubricants. The weight of block and ring were also measured after the test to record any weight loss due to friction and wear occurring on the contact surfaces. The blocks were then observed under Dino-Lite microscope to study the wear area for each test lubricants.

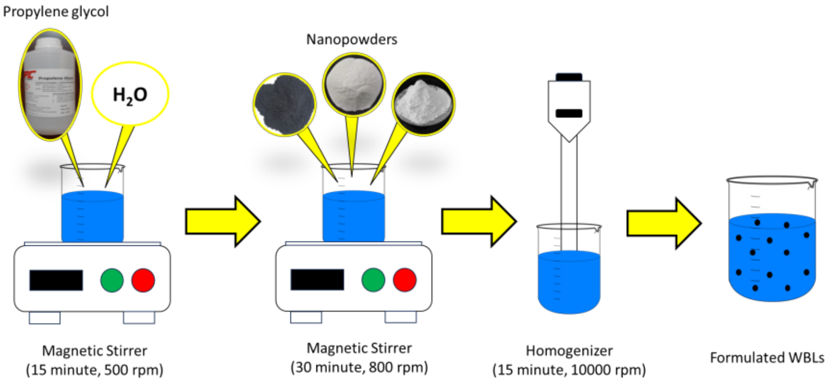


Figure 1: Formulation of water-based lubricants.

Table 1: Composition in formulated test lubricants.

Sample Name	**Nanopowder**	**Surfactant**	**Water Content**
PG-MgO PG-SiC PG-Al_2O_3 PG-MgO/SiC	0.5%wt	75%	25%
PG- Al_2O_3/MgO	0.25%wt MgO + 0.25%wt Al_2O_3		
PG- Al_2O_3/SiC	0.25%wt SiC + 0.25%wt Al_2O_3		
PG-Al_2O_3/MgO/SiC	0.167%wt MgO + 0.167%wt SiC + 0.167%wt Al_2O_3		

Table 2: Test parameters.

Parameter	**Details**
Block	Ø14mmxL14mm,67HRC R_a=0.0363µm, R_q=0.0455µm
Ring	Ø40mmxW12mm,68.1HRC R_a=0.6453µm, R_q=0.8317µm
Contact Load	40N
Temperature, T	30°C (room temperature)

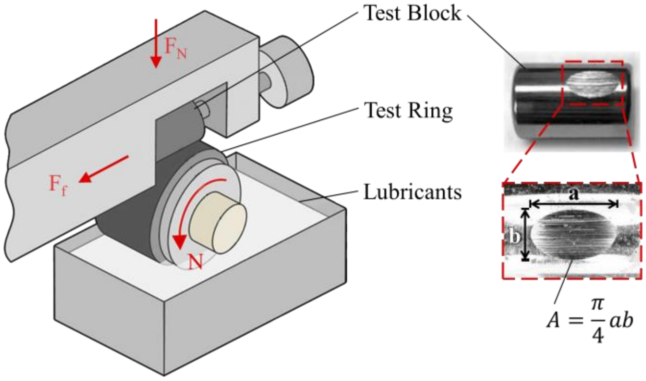


Figure 2: Block-on-ring setup.

Table 3: Properties of block and ring used in the test.

Properties	AISI 52100 bearing steel
Density	7.81 g/cm3
Vickers Hardness	690 HV
Elastic modulus E	210 GPa
Yield strength	1800 MPa
Ultimate strength	2400 MPa
Chemical composition	Fe 96.5%, Cr 1.6%, C 1.1%, Mn 0.45%, Si 0.3%, P 0.025%, S 0.025%

3.0 RESULTS AND DISCUSSION

3.1 Morphological Properties of Nanopowders

The physical properties and morphology of the nanopowders are shown in Figure 3. Based on the figure, it is observed that SiC and Al_2O_3 have an irregular and angular shapes which may possess abrasive properties. MgO however is observed to have a nearly spherical shaped. The sizes of the nanopowders are approximately 20nm.

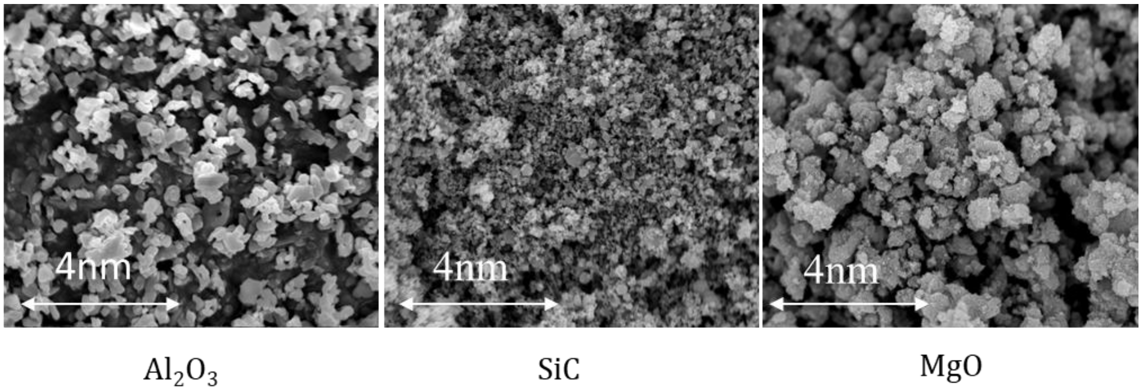


Figure 3 : FESEM images of nanopowder additives.

3.1 Viscosity of Formulated WBLs

Figure 3 shows the kinematic viscosity of single and hybrid WBLs at room temperature. Based on the figure, it is observed that MgO have the highest kinematic viscosity for single (26.32mPA·s) and MgO/SiC/Al_2O_3 for hybrid WBLs (21.8 mPA·s). The value of the kinematic viscosity was influenced by the type of nanopowder used as metal oxide tends to improve the viscosity of lubricants.

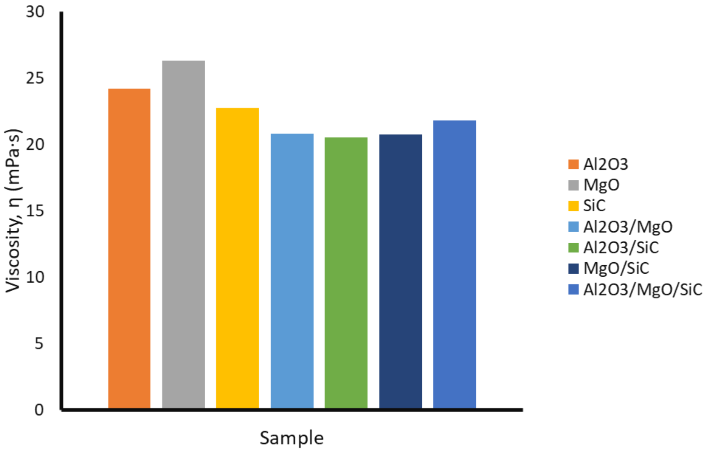


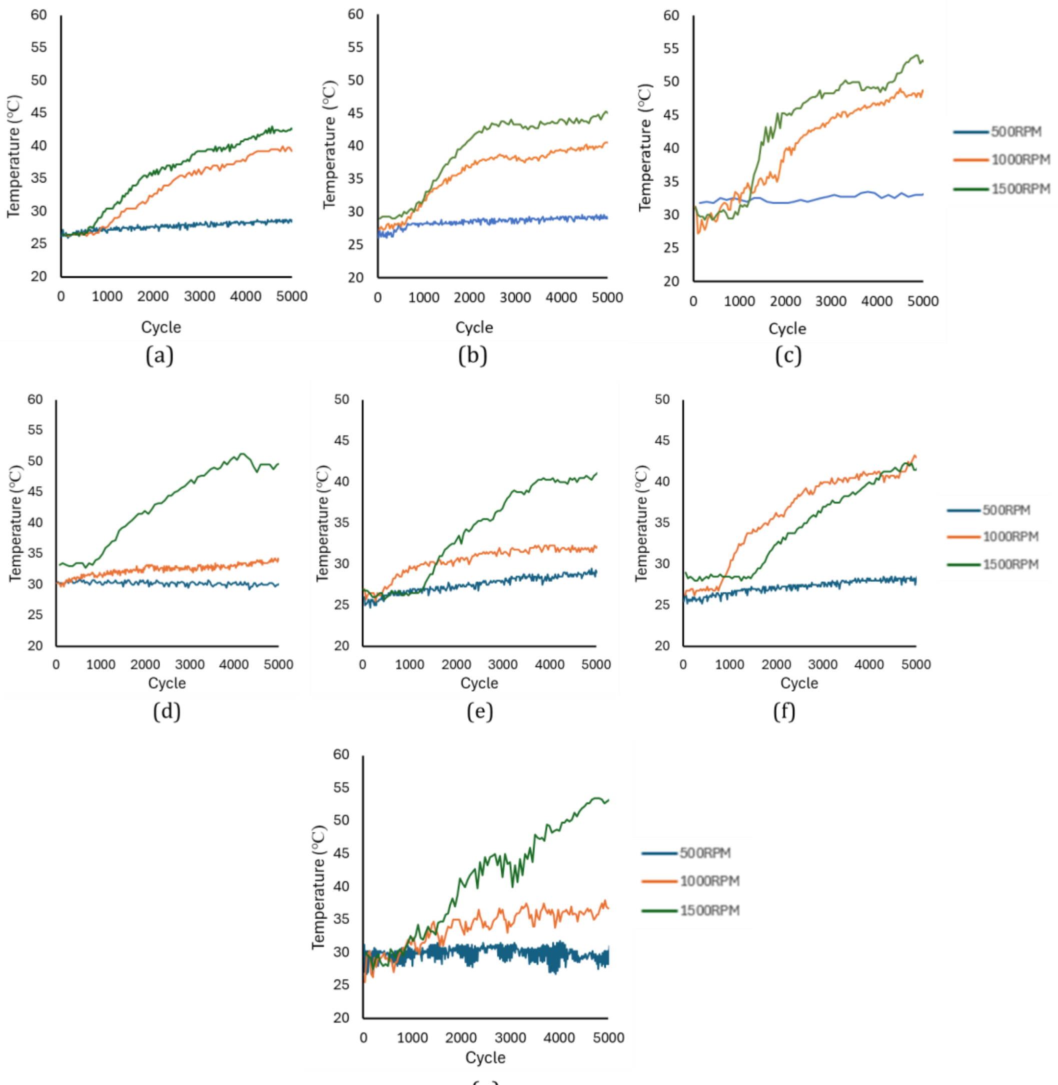
Figure 4: Kinematic viscosity of lubricants.

3.2 Temperature and Tribological Characteristics

The temperature against cycle of single and hybrid WBLs were plotted as shown in Figure 5. Each lubricant maintains the temperature below 60°C. The sudden increase during the initial period and stabilizes after can be describe as plateau effect. At 500rpm, MgO shows the lowest reading in frictional coefficient for lubricants for single WBLs, as for hybrid WBLs, MgO/Al_2O_3 have the lowest heat generation compared to other hybrid WBLs while Al_2O_3/SiC and MgO/Al_2O_3/SiC have a very high heat generation. Due to abrasive properties possess by SiC and Al_2O_3, it influenced the increase in temperature due to high friction between the nanopowder and the contact surfaces in the system. MgO presents the lowest reading in temperature due to excellent thermal conductivity which contributed by the pore size and volume as well as high specific surface area, thus helps to dissipate the heat faster (Granja et al., 2024; Hornak et al., 2018; Vu et al., 2014).

However, as the speed increases, Al_2O_3 have the lowest reading for temperature compared to MgO while SiC remains to have the highest temperature. The increase in temperature for MgO at a higher speed can be identified as a weakening mechanism for the nanopowder as MgO has the potential to generate higher temperature due to flash heating, referring to the rapid and localized increase in temperature at the contact points between surfaces in relative motion (Yao et al., 2016). The increase in temperature can also be contributed by increasing in friction and impact between the contact surfaces is as the rotational speed increases (Bai et al., 2022).

Figure 6 illustrates the coefficient of friction against cycle of the WBLs. The coefficient of friction for single WBLs show MgO having the lowest value compared to others. The physical properties of MgO may contribute to the improvement of the tribological properties. On the other hand, SiC however have the highest coefficient of friction due to irregular shapes of nanopowders and angular morphological properties which may introduce abrasive effects onto the surface (Rui et al., 2023). The coefficient of friction also may be influence by the kinematic viscosity of lubricants. The higher kinematic viscosity represents a thicker liquid film which may increase the relative motion, led to a higher coefficient of friction (Nirmal, 2018). Hence, a suitable viscosity is required in order to improve the tribological properties of formulated WBLs.

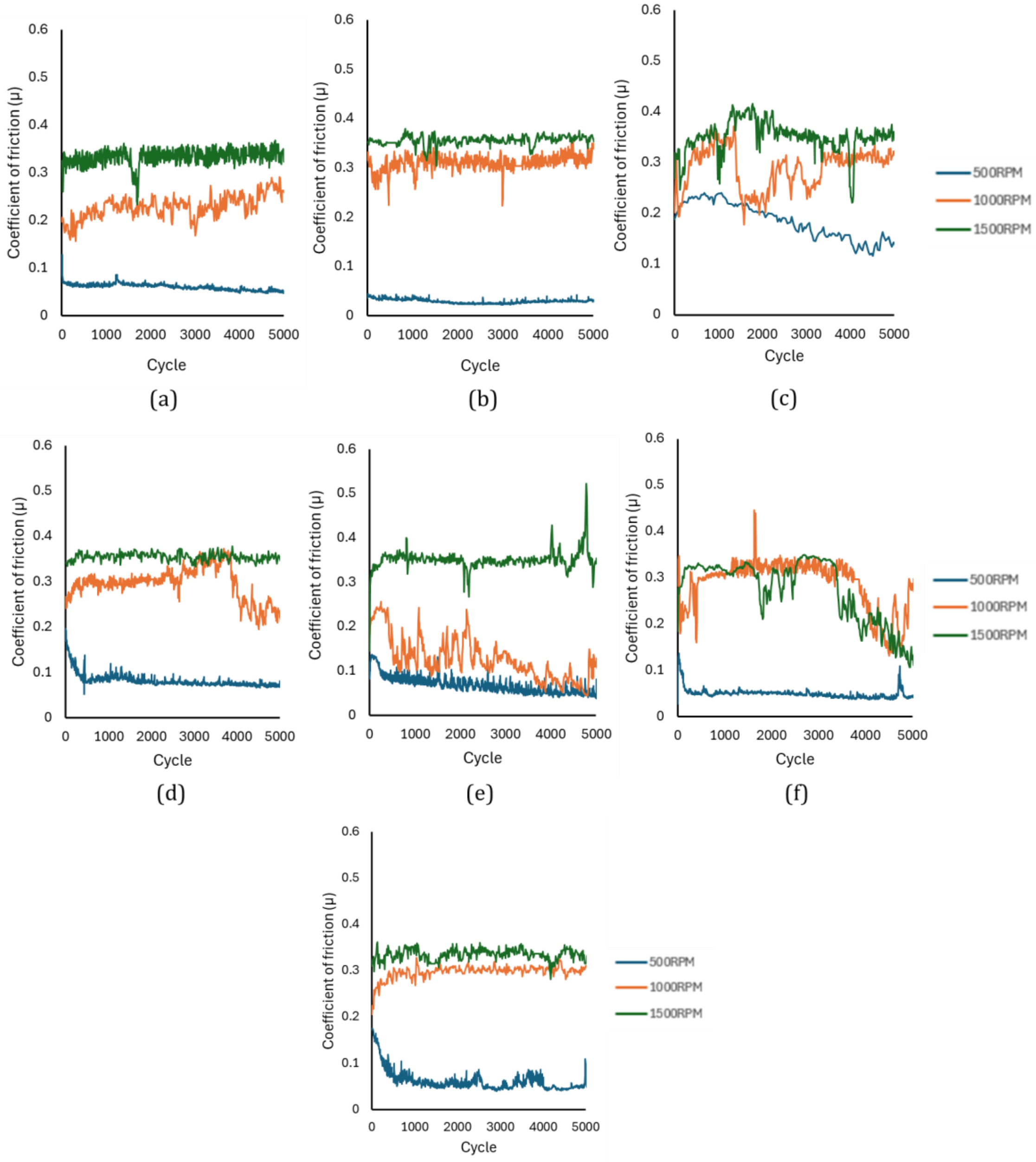


(a) (b) (c)

(d) (e) (f)

(g)

Figure 5: Temperature against cycle for (a) Al_2O_3, (b) MgO, (c) SiC, (d) Al_2O_3/SiC, (e) Al_2O_3/MgO, (f) MgO/SiC and (g) Al_2O_3/MgO/SiC nanopowder additives in WBLs at various rotational speeds.



(a) (b) (c) (d) (e) (f) (g)

Figure 6: Coefficient of friction against cycle for (a) Al_2O_3, (b) MgO, (c) SiC, (d) Al_2O_3/SiC, (e) Al_2O_3/MgO, (f) MgO/SiC and (g) Al_2O_3/MgO/SiC nanopowder additives in WBLs at various rotational speeds.

The average frictional coefficient against the rotational speed has been illustrated in Figure 7. At 500rpm, MgO proven to have the lowest average frictional coefficient (0.0318) while SiC have the highest value (0.0931) compared to all lubricants which can be explained by the bearing effect and polishing effect possessed by each nanopowders respectively. However, MgO have the highest average frictional coefficient at 1500rpm (0.3539) which can be explained by the enhanced interaction between nanopowders and water which leads to formation of densely packed particles, leading to a greater resistance against sliding motion due to higher effective viscosity (Wang et al., 2019). At 1000rpm, Al_2O_3 have the highest average coefficient of friction of 0.3328, but then dropped to 0.2916 at 1500rpm. MgO/SiC also shows the same pattern of declining from 0.2862 to 0.2187 at the rotational speed of 1000rpm to 1500rpm. The reduction of friction coefficient at 1500 rpm for Al_2O_3 and MgO/SiC can be attributed by the load-carrying capacity of the additives itself which causing the separation of contact surfaces (Autay et al., 2011; Naveed, 2023; Shrivastava et al., 2018; C. Wang et al., 2022). As the rotational speed increases, the shear forces acting on the fluid film also increase, allowing for better separation of the contact surfaces and thus reducing the friction coefficient (Na et al., 2022). Furthermore, the presence of SiC in the MgO/SiC composite may contribute to a synergistic effect that enhances the lubricating properties of the fluid, leading to a decrease in friction as the speed increases (Guo et al., 2020).

The sudden increase in the frictional coefficient were influenced by plateau effect which caused by reduction of surface roughness or alteration of the surface condition as the rotational speed increases (Johansson et al., 2017). Plateau effect can be described as the friction of system stabilizes after a period of high wear or friction due to the alteration of surface roughness. Duan et al. highlights that the friction coefficient can initially fluctuate due to the interlocking of rough surface features, but as sliding continues, these features become smoothed, leading to a reduction in friction and the establishment of a plateau (Duan et al., 2023). This observation is consistent with findings from Hu et al., who noted that the presence of plateau components directly affects sliding properties and friction behavior, indicating that smoother surfaces can lead to more stable frictional performance (Hu et al., 2017, Primožič et al., 2022).

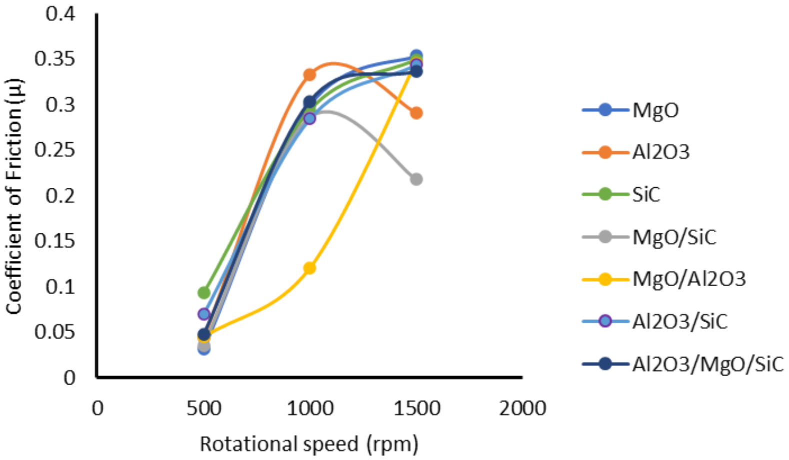


Figure 7: Rotational speed against coefficient of friction of test sample.

3.2 Wear Analysis

The wear area of the blocks surface was observed using Dino-Lite microscope as shown in Figure 8 and 9. At lower speed, MgO eexhibitsthe smallest wear area, followed by Al_2O_3 and MgO.

Al_2O_3 and SiC widely known to be abrasive due to high hardness possess by the nanopowders itself (Arafat et al., 2023; Rui et al., 2023), while MgO additives lead to a homogenous and denser microstructure which increase the wear resistance (Singh et al., 2022). However, at a higher speed, Al_2O_3 have a smaller wear area, followed by SiC which have slightly smaller area than MgO. As for hybrid WBLs, SiC/MgO demonstrates the smallest wear area, followed by SiC/Al_2O_3, MgO/SiC/Al_2O_3 and MgO/Al_2O_3. Since SiC and Al_2O_3 both act as abrasive agents due to high hardness of the nanopowders, it will create a polishing effect which will form a larger worn area by corroding the metal surfaces when in contact from sliding.

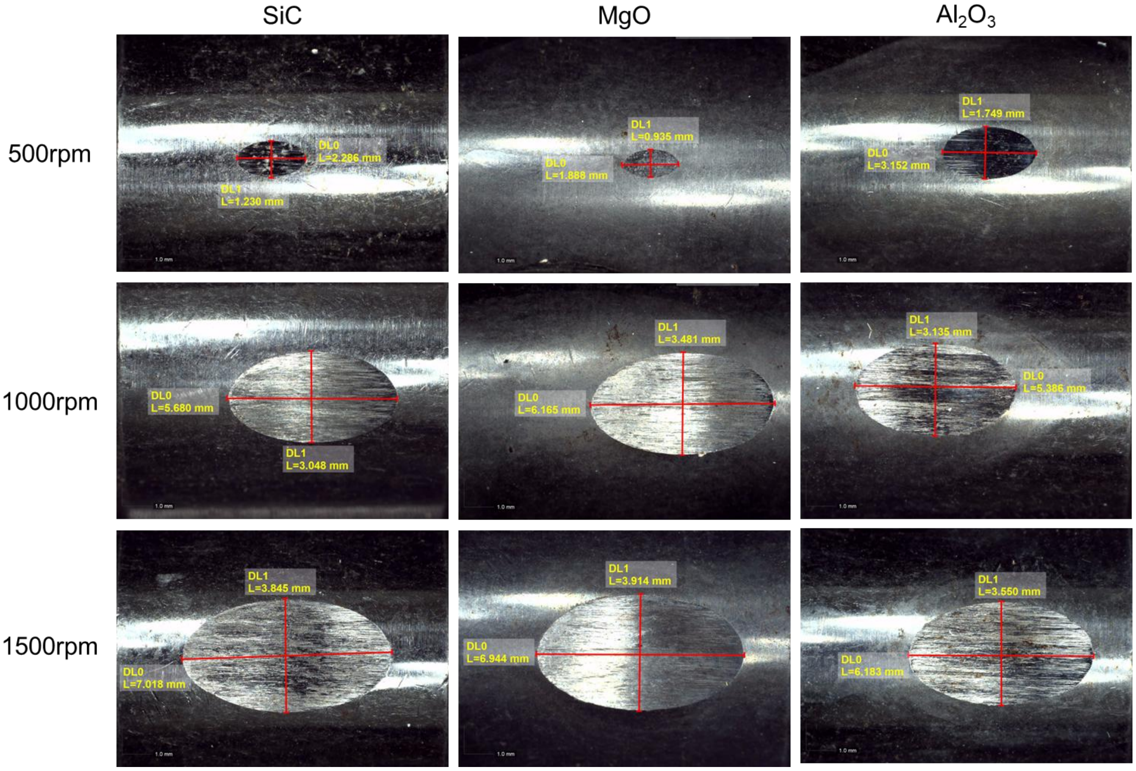


Figure 8: Wear surface of single additives nanopowder in WBLs.

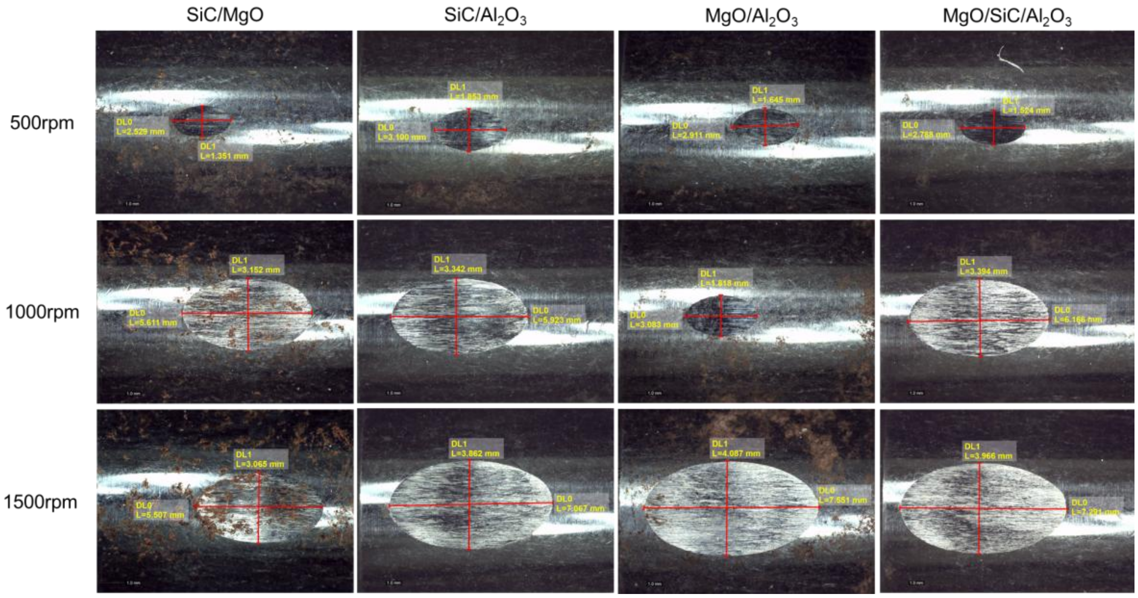


Figure 9: Wear surface of hybrid additives nanopowder in WBLs.

Table 4: Wear area for single and hybrid WBLs.

	MgO	**SiC**	**Al_2O_3**	**MgO/SiC**	**Al_2O_3/SiC**	**Al_2O_3/MgO**	**Al_2O_3/MgO/SiC**
500 rpm	$1.4mm^2$	$2.2mm^2$	$4.3mm^2$	$2.7mm^2$	$4.5mm^2$	$3.8mm^2$	$3.3mm^2$
1000 rpm	$16.9mm^2$	$13.6mm^2$	$13.3mm^2$	$13.9mm^2$	$15.5mm^2$	$4.4mm^2$	$16.4mm^2$
1500 rpm	$21.5mm^2$	$21.2mm^2$	$17.2mm^2$	$13.3mm^2$	$21.4mm^2$	$24.2mm^2$	$22.7mm^2$

The wear loss of each block-on-ring test were determined by calculating the difference of the weight of the block before and after the test has been done. The wear loss of the blocks were tabulated as shown in Table 3. MgO/SiC provides the lowest wear loss with an average percentage loss of 0.0482% while Al_2O3/SiC have the highest wear loss with an average of 0.1118%. The increase in wear loss for Al_2O3/SiC due to the incorporation of both abrasive nanopowders which will corrodes more surface of the metal (Ahmed Najar et al., 2016). In comparison to MgO/SiC, the MgO nanopowders initiated bearing effects with the help of SiC which acts as polishing effects helps in reducing the friction coefficient and wear. Plus, softer nanopowders such as MgO may deform and squeezed into the thin film under high load to maintain the lubrication without damaging the surface as illustrated in Figure 11 (Wang et al., 2017).

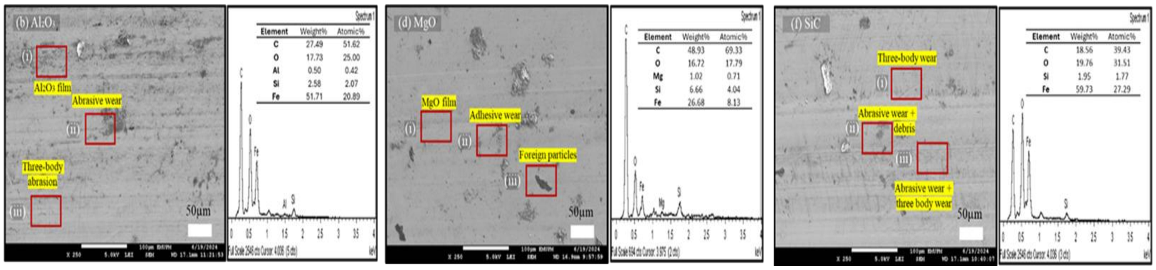
Table 5: Wear loss for single and hybrid WBLs.

Sample	Rotational speed (rpm)	Initial weight (g)	Final weight (g)	Mass difference (g)
Al_2O_3	500	16.5079	16.5053	0.0026
	1000	16.5053	16.4881	0.0172
	1500	16.4881	16.4752	0.0129
MgO	500	16.5679	16.5669	0.0010
	1000	16.5669	16.5468	0.0201
	1500	16.5468	16.5134	0.0334
SiC	500	16.5799	16.5785	0.0014
	1000	16.5785	16.5713	0.0072
	1500	16.5713	16.5575	0.0138
Al_2O_3/MgO	500	16.4832	16.4820	0.0012
	1000	16.4820	16.4814	0.0006
	1500	16.4814	16.4407	0.0407
Al_2O_3/SiC	500	16.5534	16.5505	0.0029
	1000	16.5505	16.5311	0.0194
	1500	16.5311	16.4991	0.0332
MgO/SiC	500	16.5490	16.5485	0.0005
	1000	16.5485	16.5356	0.0129
	1500	16.5356	16.5251	0.0105
Al_2O_3/MgO/SiC	500	16.4993	16.4983	0.0010
	1000	16.4983	16.4815	0.0168
	1500	16.4815	16.4682	0.0133

3.3 Lubrication Mechanism

The FESEM-EDX analysis of the block were shown in Figure 10. It is observed that Al_2O_3 and MgO nanopowders were able to form mending effect by filling the small gas between the surfaces. For SiC and Al_2O_3, there are formation of abrasive wear mainly described as a three-body abrasive wear which caused by loose abrasive particles sliding on the wearing surface. The darker appearance of the wear scar is likely due to the lubricant contributing to the oxidation of steel during the wear process. The incorporation of nanopowders like SiC and Al_2O_3 into the lubricant primarily resulted in abrasive wear, as evidenced by plough lines of varying intensities (Gara & Zou, 2012). The predominant wear mechanism remained abrasive, likely due to the inherent irregularity and high hardness of the nanopowders, which functioned as abrasive grains. On the other hand, it is observed that formation of adhesive wear occured for MgO nanopowders, which refers to wear generated by sliding of one solid surface to another which leads to material transfer or loss from either surfaces.

The abrasive particles of Al_2O_3 and SiC hinders their functions as bearings. The angular shapes of the nanopowders contributed to polishing effects on surfaces which increase the wear of surface. Al2O3's molecular structure has a robust adsorption capacity, allowing for the ingress of lubricant at the interface creating mending mechanism (Bai et al., 2023). The nanopowders also formed a protective film which prevent direct contact between two friction surfaces. The spherical MgO nanopowders function as bearings, transforming sliding friction into rolling friction, mitigating the direct interaction between the tribological pair's surface. The malleable nature of MgO nanopowders allows them to undergo deformation when subjected to sustained loads and pressures (Ledoux et al., 2022). The lubrication mechanism of the nanopowders were illustrated in Figure 11.

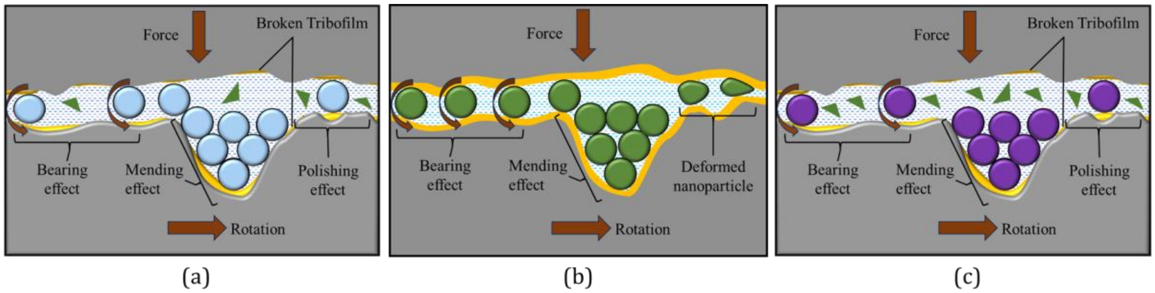


Element	Weight%	Atomic%
C	27.49	51.62
O	17.73	25.00
Al	0.50	0.42
Si	2.58	2.07
Fe	51.71	20.89

Element	Weight%	Atomic%
C	48.93	69.33
O	16.72	17.79
Mg	1.02	0.71
Si	6.66	4.04
Fe	26.68	8.13

Element	Weight%	Atomic%
C	18.56	39.43
O	19.76	31.51
Si	1.95	1.77
Fe	59.73	27.29

Figure 10 : FESEM-EDX of block samples after the test of Al_2O_3, MgO and SiC.



(a) (b) (c)

Figure 11: Lubrication mechanism for **(a)** Al_2O_3, **(b)** MgO and **(c)** SiC.

Figure 11 illustrate the Stribeck's curve for each lubricant, which evaluates the relation between the coefficient of friction and the Hersey number. Stribeck's curve starts with high friction in the boundary lubrication regime, decreases through the mixed lubrication regime, and then increases slightly in the hydrodynamic lubrication regime.

Each lubricant shows an increasing trend at a lower speed. In the initial stage of the friction coefficient, the system operates in a mixed-lubrication state. This state basically characterized by the coexistence of adsorbed fluid layers dominating the process, encompassing both boundary lubrication and hydrodynamic lubrication which involved shear flow between the surface and asperities (Stephan, 2023). In case of Al_2O_3 and MgO/SiC, there is a decline trend at a higher speed which shows a Hersey number of 68.119 and 58.303 at the frictional coefficient of 0.2916 and 0.2187 respectively. This may be described as improvement in the lubrication performance due to reduction in friction as the rotational speed. These changes may also be influenced by the properties of nanopowders added which improve the surface roughness of the surface via polishing and bearing effects. The reduction of the coefficient of friction was contributed by the formation of film by the lubricants which the load will be carried by the fluid.

Table 6 represents the rank summary of the tribological test analysis. At 500 rpm, MgO performs the best followed by MgO/SiC and Al_2O_3. For 1000rpm, it is observed that Al_2O3/SiC performs the best followed by Al_2O_3/MgO and MgO/SiC. At 1500rpm, MgO/SiC performs the best followed by Al_2O_3 and Al_2O_3/SiC. Hence, it is clearly observed that MgO/SiC was the best nanopowders to be used compared to other alternative nanopowders since it able to perform well in all rotational speed compared to others.

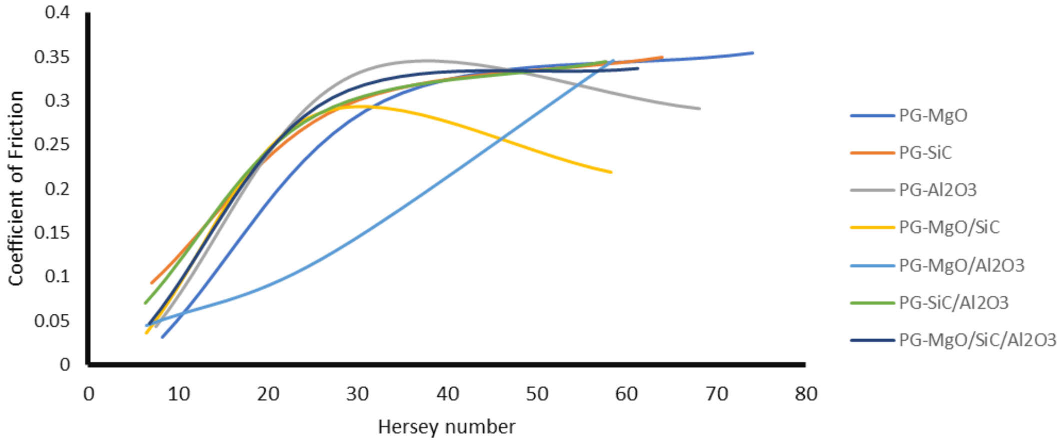


Figure 12: Stribeck's curve for single and hybrid WBLs.

Table 6: Rank of WBLs in every analysis and overall rank.

Speed	**Sample**	**Average frictional coefficient**	**Average temperature (^oC)**	**Wear area (mm^2)**	**Total**	**Overall rank**
500	MgO	1	4	1	6	1
	SiC	7	7	2	16	6
	Al_2O_3	3	1	6	10	3
	MgO/SiC	2	2	3	7	2
	Al_2O_3/SiC	4	3	5	12	4
	Al_2O_3/MgO	6	5	7	18	7
	Al_2O_3/MgO/SiC	5	6	4	15	5
1000	MgO	5	6	7	18	5
	SiC	4	7	3	14	4
	Al_2O_3	7	5	2	14	4
	MgO/SiC	3	4	4	11	3
	Al_2O_3/SiC	2	1	1	4	1
	Al_2O_3/MgO	1	3	5	9	2
	Al_2O_3/MgO/SiC	6	2	6	14	6
1500	MgO	7	4	5	16	5
	SiC	6	6	3	15	4
	Al_2O_3	2	2	2	6	2
	MgO/SiC	1	1	1	3	1
	Al_2O_3/SiC	5	3	6	14	3
	Al_2O_3/MgO	4	7	4	15	4
	Al_2O_3/MgO/SiC	3	5	7	15	4

CONCLUSIONS

In this study, the evaluation of WBLs formulated with addition of MgO, SiC, Al_2O_3 and hybrid of these lubricants together with addition of propylene glycol in a block-on-ring setup has been evaluated. The rotational speed for the system were varies throughout the session in order to observe the changes in performance of each WBLs. Based on the results, we can conclude that all lubricants able to maintain a low temperature at a lower speed. However, as the speed increases, the temperature shows an increasing trend but still able to maintain the temperature under 60°C. The frictional coefficient shows an increasing trend as the rotational speed increases except for MgO/SiC and Al_2O_3 due to the properties of the lubricants which demonstrates polishing effect and bearing effects. Among all the WBLs formulated, MgO/SiC proven to be as the most desirable lubricants as it has the lowest frictional coefficient and wear loss with an average of 0.1802 and 0.118% respectively as the rotational speed increases which contributed by the combination of bearing effect of MgO and polishing effects of SiC as well as mending effect in preventing direct contact and smoothen the surface between two contact surfaces. It is recommended to use different loads in order to study the load bearing capability of the lubricants.

ACKNOWLEDGEMENTS

The authors would like to declare that this study and publication were supported by the Ministry Higher Education under Fundamental Research Grant Scheme (FRGS/1/2022/TK10/UPM/02/9) and the Universiti Putra Malaysia GP-IPM (UPM/GP IPM/2022/9717500). The authors would like to extend their gratitude to the Department of Mechanical and Manufacturing Engineering, Faculty of Engineering, Universiti Putra Malaysia for their assistance and for making their facilities available at every stage of this research.

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