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TEMPERATURE-DEPENDENT SURFACE ENERGY AND WETTABILITY OF POLYALPHAOLEFIN WITH ORGANIC FRICTION MODIFIERS

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ABSTRACT

This study investigates the temperature-dependent wettability and surface energy behaviour of polyalphaolefin (PAO4) lubricants modified with renewable organic friction modifiers, namely fatty acid methyl ester (FAME) and glyceryl monooleate (GMO). While PAO exhibits excellent thermal stability, its inherently low polarity limits surface interaction and may reduce its effectiveness under boundary-dominated lubrication conditions. To address this limitation, PAO4 was blended with varying concentrations of FAME and GMO, and their interfacial behaviour was evaluated on steel and glass substrates at 25°C, 40°C, and 60°C using contact angle measurements. The results show that increasing additive concentration consistently reduces contact angle and interfacial energy, accompanied by reduced interfacial energy and improved spreading behaviour. On steel surfaces, the contact angle decreased from 34.21° for neat PAO4 to 19.71° for the PAO4–FAME 10 wt% blend at 60°C, corresponding to approximately 42% improvement in wettability. Similarly, PAO4 blended with GMO 7 wt% exhibited the lowest interfacial energy among the tested formulations. FAME (10 wt%) and GMO (7 wt%) exhibited optimal performance, indicating a concentration-dependent balance between polar interaction and dispersive spreading. Temperature further amplifies these effects, suggesting thermally activated molecular mobility enhances surface affinity. These findings demonstrate that wettability enhancement in PAO-based lubricants is governed by the interplay between additive polarity and temperature, providing a practical framework for screening sustainable friction modifiers. The study offers a surface-energy-based approach for screening lubricant–surface compatibility as a preliminary indicator for boundary lubrication applications.

KEYWORDS

Polyalphaolefin, Fatty Acid Methyl Ester; Glyceryl Monooleate; Temperature dependence; Surface energy

INTRODUCTION

Lubricants are essential for the optimal performance and longevity of mechanical systems, as they reduce friction, minimise wear, and enhance energy efficiency. Typically, lubricants consist of base oils and additives, with base oils accounting for 75–99% of the formulation, depending on the application [1]. Base oils are generally classified into mineral, semi-synthetic, and synthetic types. While mineral oils have historically dominated the market due to their availability and cost-effectiveness, their environmental persistence and toxicity have raised significant concerns. Mineral oils, derived from petroleum, are non-biodegradable and can persist in the environment, contaminating soil and water, and posing risks to ecosystems and human health. Their use and extraction also contribute to greenhouse gas emissions, exacerbating climate change [2].

As environmental awareness grows and regulations become stricter, there has been a shift towards the development of biodegradable lubricants, or bio-lubricants, which are derived from renewable resources and designed to minimise ecological impact. These alternatives not only address the environmental issues associated with mineral oils but also align with global sustainability goals [2]. Among synthetic base oils, polyalphaolefin (PAO) and synthetic esters have become increasingly popular, accounting for significant shares of the lubricant market [1]. PAO, classified as a Group IV base oil, is synthesised by hydrogenating polymerised alpha olefins and is valued for its high viscosity index, low pour point, high flash point, low toxicity, low volatility, and excellent thermal-oxidative stability [3]. These properties make PAO suitable for demanding applications such as engines, compressors, and hydraulic systems.

Despite these advantages, PAO's low polarity limits its wettability and tribological performance compared to organic esters, necessitating the use of additives [4]. To address PAO's limitations, various additives have been explored. Nanoparticles, such as nanodiamonds and carbon nanotubes, can significantly reduce friction and wear. Metal borates and molybdenum disulfide also exhibit promising antiwear properties. The incorporation of these additives not only improves PAO performance but also supports the industry's transition to more sustainable, environmentally friendly lubricant solutions [5]. Traditional additives like zinc dialkyl dithiophosphate (ZDDP) offer anti-wear protection but may promote micro-pitting in rolling contacts, requiring careful formulation [6].

Specialised surfactant molecules known as Organic Friction Modifiers (OFMs) are essential for improving lubricant performance, especially in engine oils. Their main purpose is to drastically lower wear and friction under boundary lubrication conditions, which is crucial for mechanical systems to operate well. OFMs do this by creating an adsorbed layer on metal surfaces, which successfully keeps opposing surfaces from coming into direct contact while in motion [7]. Studies have focused on the use of renewable organic friction modifiers, such as Fatty Acid Methyl Ester (FAME) and glyceryl monooleate (GMO), to enhance the surface energy and wettability of PAO lubricants [8]. These modifiers are attractive due to their environmental compatibility and ability to enhance surface energy and wettability.

Although FAME and GMO have been widely investigated as renewable lubricant additives and friction modifiers, most previous studies primarily focused on friction and wear behaviour. Limited attention has been given to the temperature-dependent surface energy and wettability behaviour of PAO-based lubricant systems. In particular, the relationship between additive concentration and interfacial energetics remains insufficiently understood. Therefore, a systematic understanding of polar and dispersive interactions is needed to establish more effective preliminary screening methods for lubricant formulation prior to tribological validation.

Therefore, this study aims to further analyse the effects of organic friction modifiers on the temperature-dependent surface energy and wettability of PAO when applied to steel and glass surfaces. Unlike previous studies that focus primarily on friction and wear performance, this work emphasises temperature-dependent surface energy decomposition to provide a predictive basis for lubricant formulation. The findings are expected to provide useful information for more effective screening of additives when added to base oils. This study focuses on interfacial metrics as preliminary indicators of lubrication performance prior to full tribological validation.

METHODOLOGY

The methodology for this study was designed to systematically evaluate the temperature-dependent surface energy and wettability of polyalphaolefin (PAO4) lubricants blended with organic friction modifiers, specifically Fatty Acid Methyl Ester (FAME) and glyceryl monooleate (GMO). The GMO, with Product No. 49960, CAS69 No. 111-03-5, purity >90%, was procured from Sigma-Aldrich Malaysia and used as the commercial OFM. On the other hand, the FAME was synthesised from refined palm olein according to the procedures described by Hamdan et al. [9].

The process began with preparing lubricant samples by blending PAO4 base oil with precise weight percentages of FAME and GMO. The mixing procedure involved magnetic stirring to ensure initial dispersion, followed by ultrasonic mixing to achieve a homogeneous mixture and eliminate any agglomerates. Each lubricant sample was then stored in clean, sealed containers to maintain its integrity prior to testing.

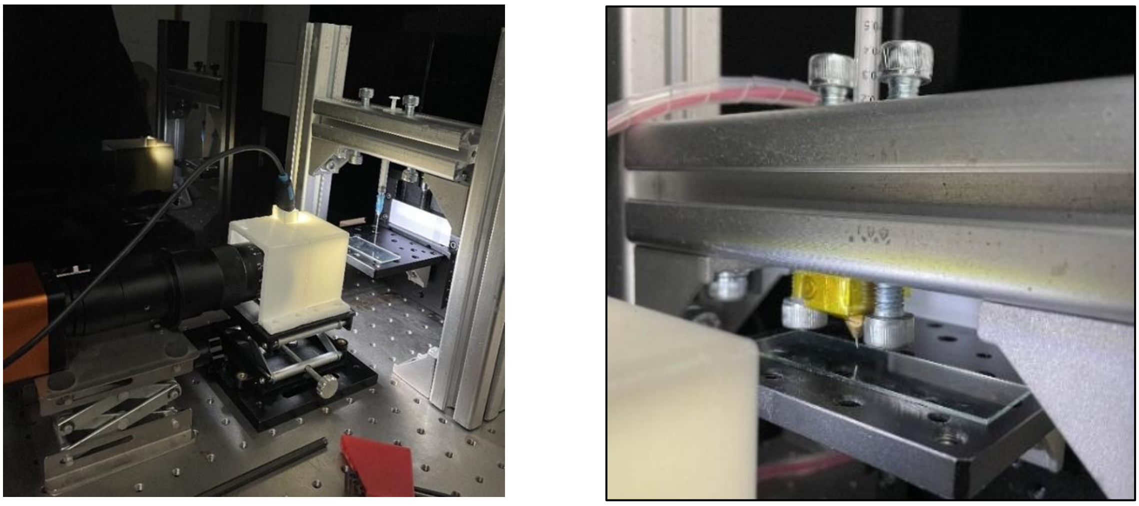
For the experimental phase, contact angle measurements were conducted to assess the wettability of each lubricant mixture on both steel and glass substrates. Prior to testing, the substrates were cleaned using acetone and dried under ambient conditions to minimise surface contamination. An in-house-built goniometer (details given in the work from Lee et al. [10]), modified to include a heating block, was used in the present study for this purpose. The lubricant temperature was controlled using the heating block integrated into the goniometer setup, while the substrate was allowed to thermally equilibrate prior to measurement. The setup is given in Figure 1.

The contact angle was measured at three different temperatures: 25°C, 40°C, and 60°C. In each test, a droplet of 10 µL of the lubricant mixture was carefully placed on the substrate surface, and the contact angle was determined using the sessile drop method. The droplet profiles were captured with a high-resolution camera attached to the goniometer. Each condition was measured at least six times at different locations, and the reported values represent the average measurements.”

The captured images were analysed using ImageJ to determine the contact angles for each sample, temperature, and substrate combination. These measurements formed the basis for calculating surface energy, work of adhesion, and interfacial energy by solving the Owens–Wendt–Kaelble (OWK) equation:

$$\gamma_L\,(\cos\theta + 1) = 2(\gamma_L^d\ \gamma_S^d)^{\frac{1}{2}} + 2(\ \gamma_S^p \gamma_L^p)^{\frac{1}{2}} \qquad (1)$$

It is noted that the solid surface energy was determined using standard liquids with known polar and dispersive components, namely distilled water and diiodomethane, based on the OWK approach. All experiments were performed under controlled laboratory conditions, with equipment calibrated prior to use and each measurement repeated multiple times to ensure data reliability and reproducibility. It is noted that the repeated measurements showed consistent trends across all tested conditions.



(a) Goniometer setup

(b) Heating block for test liquid temperature control

Figure 1: In-house-built goniometer

RESULTS AND DISCUSSION

The results compare the wettability behaviour of both mixtures on two different substrates: glass and stainless steel. Contact angles were measured at three temperatures, which are 25°C (room temperature), 40°C, and 60°C, to observe the influence of temperature on the surface interaction.

Contact angle for the mixtures

At 25°C, the contact angle values were compared with pure PAO4 (without additives) in Table 1. The highest contact angle recorded was 24.68° for the PAO4 FAME 1 wt% mixture, while the lowest was 20.57° for the 10 wt% mixture, indicating improved wettability at higher FAME concentrations. At 40°C, the contact angle ranged from 23.62° (3 wt%) to 19.94° (10 wt%). Similarly, at 60°C, the highest contact angle was 23.39° for the 1 wt% mixture, and the lowest was 19.88° for the 10 wt% mixture. These results show a consistent decrease in contact angle with increasing additive concentration across all temperatures, suggesting that the 10 wt% FAME blend promotes the best wettability. The improved spreading behaviour at higher concentrations supports its suitability for enhancing surface coverage in lubricant formulations. This behaviour suggests that the reduction in contact angle is driven by improved alignment between the polar component of the lubricant and the surface energy of the substrate, consistent with Owens–Wendt interfacial interactions.

Table 1: Contact angle for the mixture PAO4 with FAME oil at 25°C, 40°C and 60°C on the glass surface

Mixture	Contact Angle, °		
	25°C	40°C	60°C
PAO4		48.64	
FAME 1wt%	24.68	23.40	23.39
FAME 3wt%	23.07	23.62	22.40
FAME 5wt%	22.23	22.36	22.59
FAME 7wt%	21.41	21.54	20.55
FAME 10wt%	20.57	19.94	19.88

Table 2 presents the contact angle measurements of PAO4 blended with FAME oil at 25°C, 40°C, and 60°C, conducted on the glass surface. At 25°C, the highest contact angle was 25.19° for the PAO4 FAME 1 wt% mixture, while the lowest was 21.47° for the 10 wt% mixture. At 40°C, the maximum contact angle was 22.60° (1 wt%), and the minimum was 19.75° (10 wt%). At 60°C, the highest value was 21.92° (1 wt%), and the lowest was 19.71° (10 wt%). The results show a consistent decrease in contact angle with increasing FAME concentration, indicating improved wettability at higher additive levels. Notably, the lowest contact angle of 19.71° at 60°C for the 10 wt% mixture on the glass surface demonstrates excellent spreading behaviour.

Table 3 presents the contact angles of PAO4 blended with glyceryl monooleate (GMO) on the steel surface at 25°C, 40°C, and 60°C. At 25°C, the highest contact angle was 28.38° for the 1 wt% mixture, while the lowest was 20.38° for the 7 wt% mixture. At 40°C, the contact angle ranged from 25.56° (1 wt%) to 20.46° (7 wt%). At 60°C, the highest contact angle was 23.62° (1 wt%) and the lowest was 19.53° (7 wt%). These results indicate that the PAO4–GMO 7 wt% mixture consistently exhibited the lowest contact angle across all temperatures, demonstrating superior wettability on the steel surface. The significant reduction in contact angle with increasing GMO concentration suggests enhanced lubricant spreading on metallic surfaces.

Table 2: Contact angle for the mixture PAO4 with FAME oil at 25°C, 40°C and 60°C on the steel surface

Mixture	Contact Angle, °		
	25°C	40°C	60°C
PAO4		34.21	
FAME 1wt%	25.19	22.60	21.92
FAME 3wt%	24.40	21.22	20.31
FAME 5wt%	23.39	21.78	21.61
FAME 7wt%	22.56	21.51	20.61
FAME 10wt%	21.47	19.75	19.71

Table 3: Contact angle for the mixture PAO4 with GMO at 25°C, 40°C and 60°C on the glass surface

Mixture	Contact Angle, °		
	25°C	40°C	60°C
PAO4		48.64	
GMO 1wt%	29.53	27.52	25.54
GMO 3wt%	24.40	26.48	24.47
GMO 5wt%	25.47	23.20	22.33
GMO 7wt%	22.53	21.56	20.57
GMO 10wt%	21.58	21.56	20.55

Compared to the values observed on the glass surface (as shown in Table 4), the steel surface exhibited even lower contact angles for the 7 wt% mixture, indicating that steel offers better wettability for this formulation. Overall, the PAO4–GMO 7 wt% mixture showed the most favourable wetting characteristics on both substrates, particularly on steel.

Table 4: Contact angle for the mixture PAO4 with GMO at 25°C, 40°C and 60°C on the steel surface

Mixture	Contact Angle, °		
	25°C	40°C	60°C
PAO4		34.21	
GMO 1wt%	28.38	25.56	23.62
GMO 3wt%	26.56	23.58	22.56
GMO 5wt%	24.54	22.68	21.36
GMO 7wt%	20.38	20.46	19.53
GMO 10wt%	20.48	20.61	19.58

The contact angles of PAO4 blended with 7 wt% GMO and PAO4 with 10 wt% FAME oil on the steel surface at 60°C indicate good wettability, as evidenced by the reduced contact angle and improved spreading behaviour on the substrate. For the PAO4–FAME mixtures, higher temperature decreases the contact angle, indicating enhanced wettability of the steel surface. This trend suggests that higher temperatures promote better interaction between the lubricant and the substrate. The temperature-dependent decrease in contact angle is illustrated in Figure 2, highlighting the correlation between thermal conditions and surface spreading characteristics.

On the other hand, the contact angle of PAO4 blended with 7 wt% GMO on the steel surface shows a decreasing trend with increasing temperature, indicating improved wettability. This suggests that the steel surface allows the lubricant mixture to spread better at elevated temperatures. As illustrated in Figure 2 and further confirmed in Figure 3, the contact angle decreases progressively, demonstrating enhanced spreading behaviour of the mixture on the steel surface.

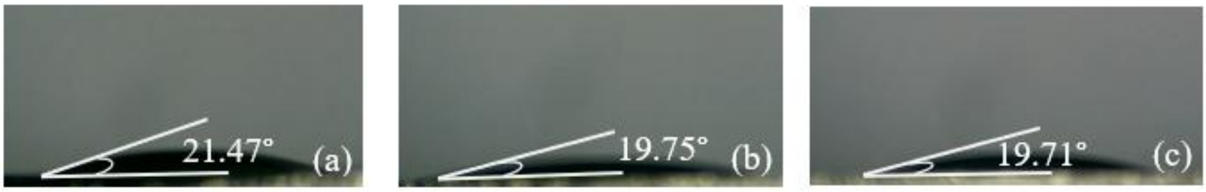


Figure 2: Contact angle of PAO4 with FAME oil 10wt% on the steel surface in temperatures (a) 25°C, (b) 40°C, and (c) 60°C

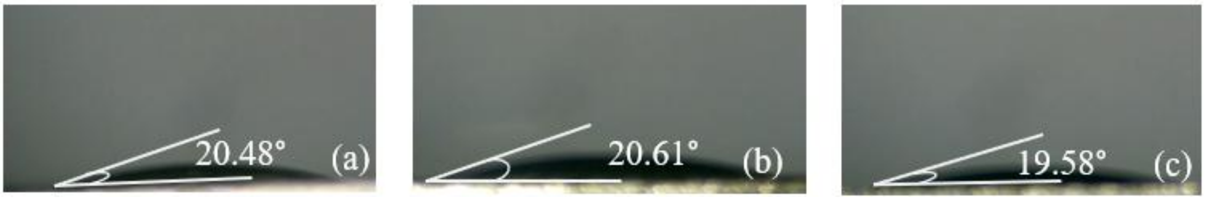


Figure 3: Contact angle of PAO4 with GMO 7wt% on the steel surface in temperatures (a) 25°C, (b) 40°C, and (c) 60°C

Surface Energy for the mixtures

For the PAO4-FAME mixtures, the results indicated a clear increase in surface energy with the addition of FAME compared to pure PAO4. At 25°C, the surface energy of the PAO4-FAME mixture on the glass substrate was higher than that of the unmodified base oil, and this trend persisted at 40°C and 60°C. Similar enhancements were observed on the steel substrate, demonstrating FAME's effectiveness in improving adhesive interaction between the lubricant and solid surfaces. The increase in surface energy with temperature suggests that higher thermal energy promotes better spreading and interaction of the lubricant on both glass and steel surfaces, which is favourable for lubricant film formation and stability. This trend indicates that increased temperature enhances molecular mobility, allowing polar functional groups from the additives to interact more effectively with the substrate, thereby strengthening interfacial adhesion.

Table 5 presents the solid surface energies of steel and glass measured using standard liquids with known surface energies. The data show that the lowest polar component of the solid surface energy was observed on the glass surface, while the lowest dispersive component was recorded on the steel surface. These variations indicate substrate-dependent differences in surface interactions between the lubricant and the solid materials.

Table 5: Surface energy on solid for the mixture PAO4 with FAME oil in 25°C

Surface	Surface Energy on solid, (mJ/m^2)	
	25°C	
	γ_S^P	γ_S^D
Steel	23.41	33.93
Glass	15.74	38.86

Using the surface energy of the steel surface above, Table 6 presents the polar and dispersive surface energy components of the tested liquid mixtures at three different temperatures. At 25°C, the highest polar component recorded was 9.93 mJ/m² (FAME 1wt%), while the lowest was 9.33 mJ/m² (FAME 7wt%). For the dispersive component, values ranged from a high of 48.80 mJ/m² (FAME 1wt%) to a low of 47.53 mJ/m² (FAME 10wt%).

Table 6: Surface energy (liquid) for the mixture PAO4 with FAME oil at 25, 40, and 60°C

Mixture	Surface energy on liquid (mJ/m^2)					
	25°C		40°C		60°C	
	γ_L^P	γ_L^D	γ_L^P	γ_L^D	γ_L^P	γ_L^D
PAO4	11.86	52.09	-	-	-	-
FAME 1wt%	9.93	48.80	10.77	47.72	11.20	47.41
FAME 3wt%	9.25	48.58	11.81	47.05	11.43	46.91
FAME 5wt%	9.35	48.20	10.52	47.48	10.80	47.39
FAME 7wt%	9.33	47.91	10.11	47.46	9.99	47.19
FAME 10wt%	9.47	47.53	10.10	46.92	10.08	46.85

Table 7 presents the polar and dispersive components of the liquid surface energy for PAO4 blended with GMO at 25°C, 40°C, and 60°C. At 25°C, the highest polar surface energy was observed for PAO4 (11.86 mJ/m²), while among the GMO mixtures, GMO 1 wt% had the highest polar value at 11.79 mJ/m², and GMO 10 wt% had the lowest at 10.77 mJ/m². For the dispersive component, GMO 1 wt% showed the highest value of 49.85 mJ/m², while the lowest was recorded for GMO 7 wt% at 46.85 mJ/m².

At 40°C, GMO 3 wt% had the highest polar value of 12.62 mJ/m², and GMO 5 wt% had the lowest at 10.59 mJ/m². Regarding dispersive energy, the highest was 48.56 mJ/m² (GMO 1 wt%), and the lowest was 47.02 mJ/m² (GMO 7 wt%). At 60°C, the polar component ranged from a high of 11.79 mJ/m² (GMO 1 wt%) to a low of 10.59 mJ/m² (GMO 10 wt%). For the dispersive component, the highest was 47.88 mJ/m² (GMO 1 wt%), and the lowest was 46.76 mJ/m² (GMO 7 wt%). Overall, the PAO4–GMO 7 wt% mixture consistently exhibited the lowest dispersive surface energy across all three temperatures, indicating strong wettability and enhanced spreading behaviour compared to other concentrations.

Table 7: Surface liquid for the mixture PAO4 with GMO at 25°C, 40°C and 60°C

Mixture	Surface energy on liquid (mJ/m^2)					
	25°C		40°C		60°C	
	γ_L^P	γ_L^D	γ_L^P	γ_L^D	γ_L^P	γ_L^D
PAO4	11.86	52.09	-	-	-	-
GMO 1wt%	11.79	49.85	12.13	48.56	11.79	47.88
GMO 3wt%	11.24	49.15	12.62	47.68	11.63	47.54
GMO 5wt%	11.08	48.37	10.59	47.76	10.77	47.30
GMO 7wt%	11.50	46.85	10.76	47.02	10.63	46.76
GMO 10wt%	10.77	47.02	10.68	47.08	10.59	46.79

The temperature-dependent surface energy components for neat PAO4 were not separately evaluated in the present study. The primary objective was to comparatively assess the relative influence of additive concentration on interfacial behaviour under elevated temperatures. Therefore, the analysis emphasises comparative trends among additive-containing formulations.

Work of adhesion and liquid surface tension

Figure 4 shows the work of adhesion of the PAO4/GMO mixtures on the steel surface at 25°C. The work of adhesion generally decreases with increasing GMO concentration from 1 wt% to 10 wt%. Since work of adhesion represents the energy required to separate the lubricant film from the solid surface, lower values indicate reduced adhesive interaction at the lubricant–steel interface.

The simultaneous reduction in contact angle and interfacial energy suggests that increasing GMO concentration promotes improved spreading behaviour despite the lower adhesive interaction. This behaviour may be associated with the surface-active nature of the GMO, which reduces the energetic barrier at the solid–liquid interface and enhances the steel surface's wettability. From a thermodynamic perspective, the reduction in the work of adhesion, together with a lower interfacial energy, indicates a more energetically favourable spreading condition for the lubricant film.

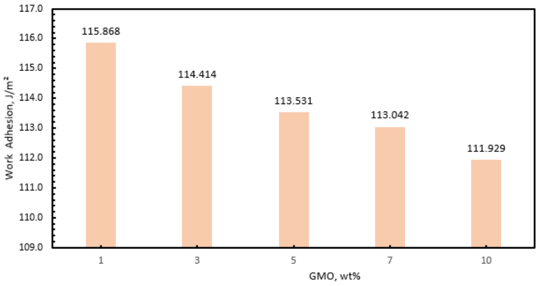


Figure 4: Work of adhesion for the PAO4 with GMO on the steel surface at 25°C

Similarly, for the PAO4–FAME mixtures, the work of adhesion generally decreased with increasing FAME concentration, with the lowest value observed at 10 wt%. Although the values at 1 wt% were higher than those at some intermediate concentrations, the overall trend suggests reduced adhesive interaction at higher FAME content. As illustrated in Figure 5, increasing FAME concentration corresponds with improved spreading behaviour and reduced interfacial energy on the steel surface.

These observations suggest that FAME modifies the interfacial characteristics of PAO4 by reducing the energy barrier at the solid–liquid interface, thereby enhancing wettability. The combined trends in contact angle, interfacial energy, and work of adhesion indicate that the improvement in wettability is primarily governed by interfacial energy minimisation rather than stronger adhesive bonding alone.

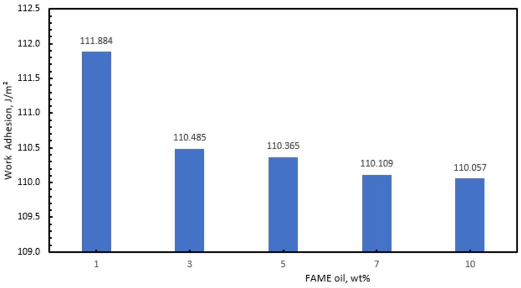


Figure 5: Work of adhesion for the PAO4 with FAME on the steel surface at 25°C

Figure 6 illustrates the effect of GMO concentration on liquid surface tension in PAO4 mixtures. Surface tension, the elastic property of a liquid that minimises surface area, plays a critical role in wetting behaviour. As the concentration of GMOs increases, the surface tension of the mixture decreases, especially at higher concentrations. This reduction in surface tension corresponds with earlier observations of increased wettability and higher work of adhesion, indicating that GMO acts as a surface-active agent that enhances the spreading and bonding of the lubricant to the substrate.

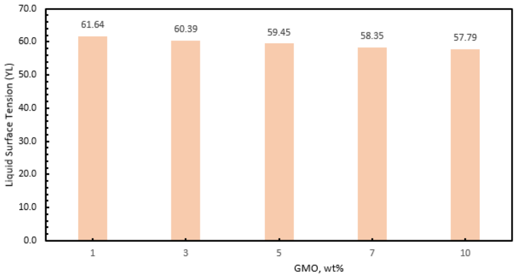


Figure 6: Liquid surface tension versus PAO4 with GMO wt% on the steel surface at the 25°C

Similarly, Figure 7 shows that increasing FAME concentration also reduces the surface tension of the PAO4 mixture, further improving its ability to spread across and adhere to the steel surface. This reduction aligns with previously reported contact angle and work of adhesion data, confirming that FAME improves the lubricant's interfacial performance.

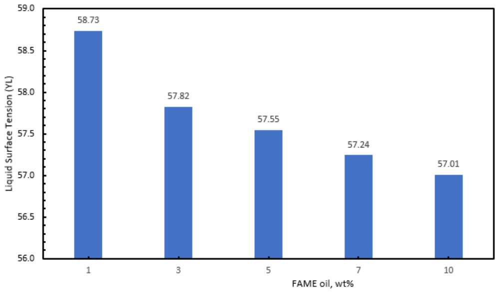


Figure 7: Liquid surface tension versus PAO4 with FAME wt% on the steel surface at the 25°C

Interfacial Energy

Interfacial energy represents the thermodynamic stability of the interface between the solid substrate and the lubricant mixture. A lower interfacial energy indicates a more stable, favourable interaction, enhancing both lubricant adhesion and spreading. As illustrated in Figure 8, increasing GMO content significantly reduces interfacial energy, especially at elevated temperatures. This trend aligns with enhanced wettability and adhesive strength, suggesting that adjusting GMO concentration in PAO4 formulations can substantially improve the energy stability of lubricant films on metal surfaces.

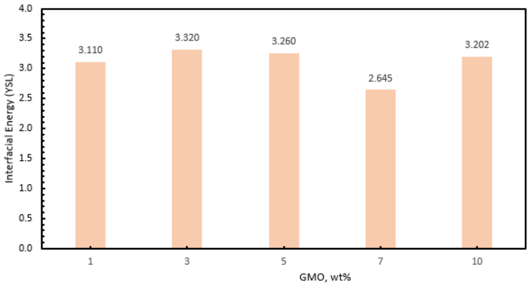


Figure 8: Interfacial Energy versus PAO4 with GMO wt% on the steel surface in the 25°C

Figure 9 shows that higher FAME content results in lower interfacial energy, indicating a more stable, adherent lubricant film. This reinforces the conclusion that FAME effectively enhances the interfacial characteristics of PAO4 on steel, particularly at higher concentrations. The observed trend, a decrease in interfacial energy with increasing FAME content, is consistent with that of GMO-based formulations. Both additives help reduce the energy barrier at the solid–liquid interface, thereby improving adhesion and wettability. Among all tested combinations, PAO4 with 10 wt% FAME exhibited the lowest interfacial energy, confirming its superior performance in promoting lubricant spreading and surface interaction. Overall, the combined trends in contact angle, surface tension, and work of adhesion demonstrate that additive-induced wettability enhancement is governed by interfacial energy minimisation.

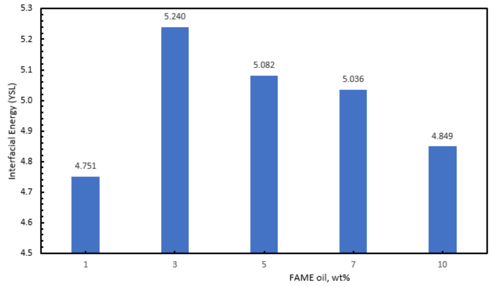


Figure 9: Interfacial Energy versus PAO4 with FAME wt% on the steel surface in the 25°C

Conclusion

This study demonstrates that the wettability and interfacial behaviour of PAO4 can be systematically enhanced by incorporating renewable organic friction modifiers, with both concentration and temperature playing critical roles. The PAO4–FAME 10 wt% blend reduced the contact angle on steel from 34.21° for neat PAO4 to 19.71° at 60°C, while the PAO4–GMO 7 wt% blend consistently exhibited the lowest interfacial energy among the tested formulations. The results show that FAME (10 wt%) and GMO (7 wt%) provide optimal performance, characterised by reduced contact angle, lower interfacial energy, and improved wettability on steel surfaces.

More importantly, the findings suggest that wettability improvement is governed by a balance between polar interaction and dispersive spreading. While increasing the additive concentration enhances surface affinity, excessive polarity does not necessarily yield proportional improvements, suggesting an optimal formulation window. Temperature further enhances wettability by increasing molecular mobility and strengthening interfacial interactions.

These results show that surface energy metrics can serve as effective proxies for screening lubricant formulations before tribological testing. However, the absence of direct friction and wear measurements limits the ability to fully correlate interfacial behaviour with tribological performance. Future work should integrate friction testing and surface characterisation.

Overall, this study provides a practical framework for designing environmentally friendly PAO-based lubricants with improved interfacial characteristics relevant to boundary lubrication applications through surface energy engineering. Although the present work focused primarily on trend analysis, future studies should incorporate full statistical treatment and expanded repeatability analysis to further strengthen quantitative interpretation.

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