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Insights into the influence of additives on the thermal gradient induced migration of lubricant

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ABSTRACT

Thermo-capillary migration is a phenomenon that the thermal gradients will drive a liquid to flow from warm to cold regions. It is of great importance to prevent the lubricant migration on rubbing surfaces in the cases where the amount of lubricant is limited. In this paper, four different lubricant additives are incorporated into one base oil, and the effects of additives on the migration behaviour and surface tension coefficient are investigated. The functional mechanisms of additives are discussed. The experimental results demonstrate that the additives have remarkable influences on the migration performance of lubricant. The migration behaviour shows the relation to not only the surface tension coefficient, but also the actions between the additive and substrate. This should be considered in the designing process of an anti-migration lubricant. Copyright © 2016 John Wiley & Sons, Ltd.

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KEY WORDS: thermal capillary migration; lubricant additives; surface tension coefficient

INTRODUCTION

In tribo-system, whenever two surfaces slide against each other, the spontaneous heat generated by friction will create a thermal gradient on the rubbing area, inducing a movement of liquid lubricant from a high temperature region to a low temperature region.[1–3] This kind of thermal gradient induced movement of liquid is referred as the thermal capillary migration. In essence, the thermal gradient produces a spatial variation in the interfacial tension of the liquid, exerting a tangential stress on the liquid interface, and eventually, results in the migration.[4–6]

The migration would seriously affect the lubrication condition and cause potential lubrication failure of moving mechanisms, especially in the cases where the amount of lubricant is limited.[7] For example, the changeable temperatures (−100 – 200°C) are of the most prominent features of a spacecraft; due to this extremely high thermal gradient, liquid lubricants employed in the mechanical devices in the vacuum and gravity free environment will be driven to escape from the rubbing area, resulting in a

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reduction of service time.[8] It is firmly believed that preventing the migration of lubricant is necessary to ensure the lubrication life and the reliability of those devices.[9]

Over the past decade, abundant correlational researches are carried out to investigate the migration mechanism and effective means to prevent the migration.[10–12] By painting low surface energy fluorocarbon compounds around the rubbing area, chemical gradients will be generated on the surface and the lubricant films can be confined at the desired location.[13] Surface roughness and surface topography also strongly influence on the liquid migration.[14–16] Recently, the surface texture such as the patterns of micro-grooves or micro-dimples is not only used to modify the tribological behaviour of sliding surfaces,[17–19] but also proposed to obstruct the thermal driven migration of lubricants.[20,21]

Moreover, the anti-migration lubricants must be taken into consideration. Rational choice and design of lubricants, for instance, increasing the viscosities of lubricants, have been proven to be effective ways to prevent the migration.[22]

Magnetic fluids are functional fluids used for many applications. These fluids show unusual properties, e.g. they can be confined, positioned, and controlled at desired places by applying an external magnetic field.[23–25] Recently, the authors investigated the migration performance of the magnetic fluids. As shown in Figure 1, under the identical experimental conditions with no external magnetic field, it is found that the droplet of the magnetic fluid migrated much faster and longer than the droplet of carrier liquid.

This is an interesting phenomenon and worth pondering the deeper implications of why the different migration behaviours exist. It is known that magnetic fluid is a colloid suspension of single-domain magnetic particles coated with surfactants and dispersed in a carrier liquid. For the magnetic fluid used in this experiment, the content of surfactant is about 6 wt%, and the nanoparticles is about 4 wt%. Therefore, it is certain that the small amount of additives has changed the migration behaviour of the liquid greatly.

Additives are the essential parts of modern lubricants. Adding additives to the base oil is an effective approach to accomplish a variety of functions such as control friction and reduce wear.[26–28] However,

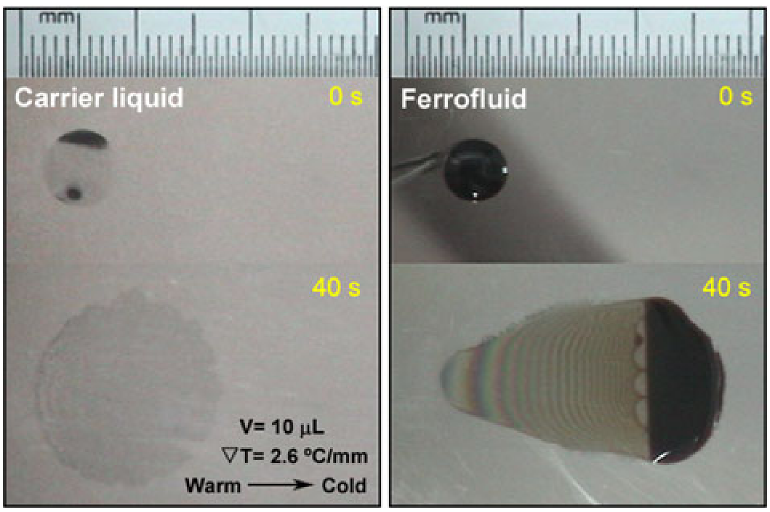

Figure 1. The migration of the carrier liquid and the magnetic fluid on a smooth stainless steel surface with a time interval of 40 s.

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it is still unclear what influences do the additives have on the thermo-capillary migration. Is it possible to modify the migration performance of lubricants by adding some additives? This seems to be an attractive method and further research is quite necessary.

Therefore, this paper is organised as follows: first, three different base oils are employed and the migration properties are tested. Then, the effects of four typical additives on the thermo-capillary migration under various experimental conditions are studied. The mechanisms of the lubricant additives on the migration behaviour are discussed. Special attention is paid to the relationship between the migration velocity and the surface tension coefficient. It is proposed that for the preparation of an anti-migration lubricant, the additives induced modification of migration property should be taken into consideration.

EXPERIMENTAL SECTION

Lubricant preparation

The following three different oils are chosen for the preliminary tests. They are paraffin oil, olive oil and sebacic acid diester (i.e. diester), which belong to mineral oil, vegetable oil and synthetic oil respectively. Paraffin oil is a mixture of liquid hydrocarbons obtained from fractionation of the atmospheric distillation of petroleum. Olive oil is extracted from olive, and it is known for its high oleic acid content. Diester is obtained from monohydric alcohol through esterification reaction. It has a good anti-oxidation ability, often used as the base oil of performance-critical engine oil, gear oil, and grease.

Four typical additives, i.e. anti-wear agent (zinc dialkyldithiophosphates, ZDDP), ashless dispersant agent (polymer succinimide, T161), anti-foam agent (polydimethylsiloxane), and oiliness agent (oleic acid) are used in this study. ZDDP is one of the most common anti-wear additives used in engines; the power of this compound is its ability to function as an excellent anti-wear agent, a mild extreme-pressure agent, and an effective oxidation and corrosion inhibitor simultaneously. T161 is a high performance ashless dispersant; it is a necessary additive for helping prevent contaminants from precipitating out of the oil and depositing on engine surfaces, which enhances the dispersion performance of lubricants. Serious foaming will break the good lubrication state and result in air entrainment, cavitation damage, and lubricant starvation; polydimethylsiloxane is always employed in liquid lubricants for the anti-foam function for its low surface tension. Oleic acid is an original oiliness additive added into lubricants to improve tribological performances. It enhances the adsorption ability of the lubricants, forming a dense and stable film on the metal surface to prevent the direct contact between rubbing surfaces.

Additives are always incorporated into lubricants with a relatively low content, usually between 0.5 and 4 wt%. In order to simplify the research, diester is chosen as the base oil, and mixed with these four different additives with the content of 1 and 3 wt% via the ultrasonic method respectively. Migration experiments are first performed on the base oils, and then on those mixed with additives.

Test procedures

The migration of the liquid lubricants was studied using the apparatus as shown in Figure 2. A digital video camera was adopted to monitor the complete dynamic lubricant migration process. All specimens were made of SUS 316 stainless steel with the dimensions of 76 mm × 30 mm × 3 mm. The testing surface was processed by grinding and polishing treatment sequentially to obtain the final

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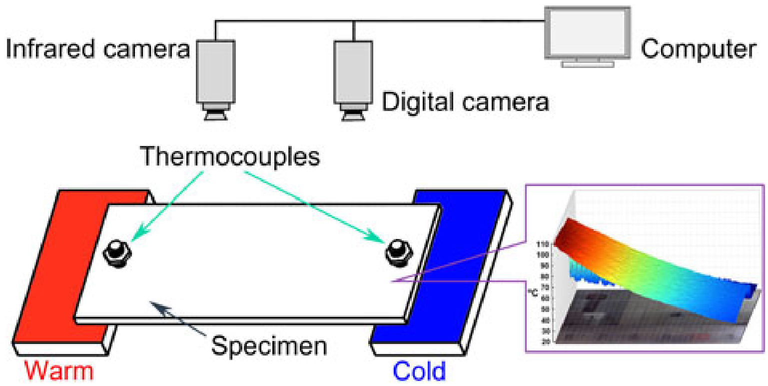

Figure 2. Schematic diagrams of the experimental apparatus.

surface roughness Ra in the range of 10–20 nm, accompanied with high quality of flatness. Our previous research found that the migration velocity decreases with decreasing surface roughness under certain conditions, and the differences tend to disappear with the further decreasing of surface roughness (Ra 1.2 μm).[15] In this study, the surfaces for migration testing were enough smooth to ignore the influence of surface roughness. So in the following sections, all the surfaces were regarded as smooth surfaces. The cold side was maintained at a constant temperature of 0°C. By changing the temperature of the warm side, a thermal gradient could be generated along the length of the surface. An infrared camera and thermocouples were used to obtain the thermography on the surface of the specimen, and then to calculate the thermal gradient. The details of the experimental apparatus are available in Ref.[15].

Prior to the experiments, specimens were ultrasonically cleaned in acetone and ethanol, rinsed with deionised water, and dried by nitrogen. The lubricant with a volume of 5 μL was precisely placed on the same position of the substrate surface using a microlitre syringe. As the liquid lubricant touches the solid surface, spreading occurs, and the initial process is somewhat complex.[29–31] In order to simplify the analysis, the initial measurement started at 1 s after the droplet was placed on the surface. The droplet was permitted to migrate for 30 s, so that the differences between the migration distances could be significant. The migration distance was measured at the front edge of the droplet, and the mean migration velocity within that time was used in this study.

The surface tension of the lubricants was measured via the Wilhelmy plate method. By measuring the surface tensions of the lubricants at variation temperatures, the surface tension coefficient γ_T can be obtained by the following equation:

$$\gamma_T = \frac{\gamma(T) - \gamma_0}{T - T_0} \tag{1}$$

where $\gamma(T)$ and γ_0 are the surface tension of the lubricant at the temperature T and T_0 respectively. The detailed data of the surface tensions of diester and diester mixed with different additives are shown in Table I. Basing on the above equation (eq. 1), the numerical fitting value of the surface tension coefficients of these lubricants was calculated and employed in the following study.

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Table I. Surface tensions of diester and diester mixed with different additives at different temperatures.

	Surface tension at different temperatures (mN/m)			
Lubricants	20°C	40°C	60°C	75°C
Diester	27.823	26.104	24.405	23.617
+1% ZDDP	30.916	28.834	27.074	26.239
+3% ZDDP	31.118	29.137	27.358	26.043
+1% T161	30.532	27.297	25.052	24.376
+3% T161	31.806	28.914	26.913	24.902
+1% Polydimethylsiloxane	21.474	19.867	18.946	17.683
+3% Polydimethylsiloxane	22.323	21.025	19.593	18.582
+1% Oleic acid	28.348	25.315	23.516	22.04
+3% Oleic acid	29.589	25.78	24.163	22.727

RESULTS

Migration of base oils

First, experiments were performed to investigate the migration property of the three base oils. Figure 3(a) provides the key frames with a time interval of 30 s, which shows the migration process of olive oil, diester, and paraffin oil on the smooth surface. Under a thermal gradient of 1.8°C/mm, the olive oil droplet migrated in a short distance, just about 1.4 mm, the diester droplet migrated much longer, nearly 10 mm, and the paraffin oil droplet migrated in a longest distance of 11.6 mm. Figure 3(b) shows the migration velocities of these three lubricants under two different thermal gradients. It can be seen that a higher thermal gradient increased the migration velocity significantly. When the thermal gradient increased to 3.0°C/mm, the migration velocity of paraffin oil was about 0.88 mm/s, which was nearly 144% higher than the 0.61 mm/s observed for the diester. Meanwhile, for the olive oil, it always migrated with a very slow rate, the velocity was just about 0.13 mm/s, which was much lower compared with the other two. One possible reason is that the content of oleic acid in the olive oil used in this study is quite high.

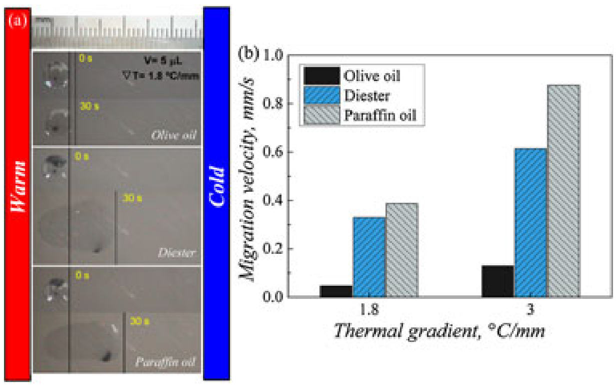

Figure 3. Thermal capillary migration of the three base oils on smooth surface: (a) Detailed macroscopic process under a thermal gradient of 1.8°C/mm. (b) Migration velocity under different thermal gradients.

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The migration result shows that both diester and paraffin oil are suitable to be chosen as base oil. However, compared to paraffin oil, diester has a good anti-oxidation ability (under a high thermal gradient, the temperature is relatively high at the start point), and more importantly, diester can dissolve most kinds of lubricant additives.[32] Therefore, to study the effect of additives on the migration performance, in the following sections, additives are incorporated into diester and migration experiments are performed.

Effect of lubricant additives

Figure 4 presents the effect of ZDDP on the migration velocity and the surface tension coefficient of diester. The histogram shows the relationship between migration velocity and ZDDP content. A rapid increasing in a thermal gradient led to an increasing migration velocity. The ZDDP content of 3% yielded a higher migration velocity than that of 1%. At the thermal gradient of 3.0°C/mm, the velocity of content of 3% was about 0.89 mm/s, which was 130% higher than the 0.69 mm/s observed for the content of 1%, and 146% higher than the 0.61 mm/s observed for the pure diester. The point-line exhibited the influence of ZDDP on the surface tension coefficient of diester. The surface tension coefficient was increased with increasing content of ZDDP. It indicates that when adding ZDDP into the lubricant, the migration velocity will be increased obviously.

Figure 5 shows the effect of T161 on the migration velocity and the surface tension coefficient of diester. As the histogram exhibited, at a thermal gradient of 3.0°C/mm, T161 content of 1% yielded a higher migration velocity than the pure diester. When the content increased to 3%, the migration velocity was about 1.15 mm/s, which was nearly 150% higher than the 0.77 mm/s observed for the content of 1%. It demonstrated that the migration velocity was increased with increasing content of T161. The point-line exhibited the influence of T161 on the surface tension coefficient of diester. With the increasing content, the surface tension coefficient of diester was also increased.

As indicated by the similar trend in the migration rate under the two thermal gradients, it can be found that by adding T161 into lubricant, the migration velocity will be increased greatly.

Figure 6 shows the influence of the polydimethylsiloxane on the migration behaviour and the surface tension coefficient. As the histogram exhibited, when the thermal gradient is 1.8°C/mm, the pure diester achieved a migration velocity of 0.33 mm/s; once the polydimethylsiloxane with content of 1%

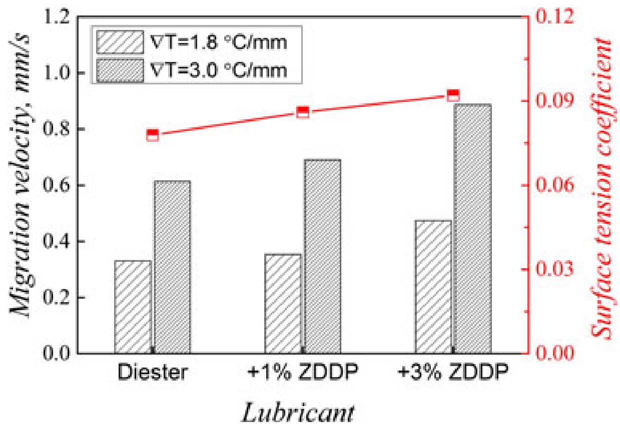

Figure 4. The effects of ZDDP on the migration behaviour and the surface tension coefficient of diester.

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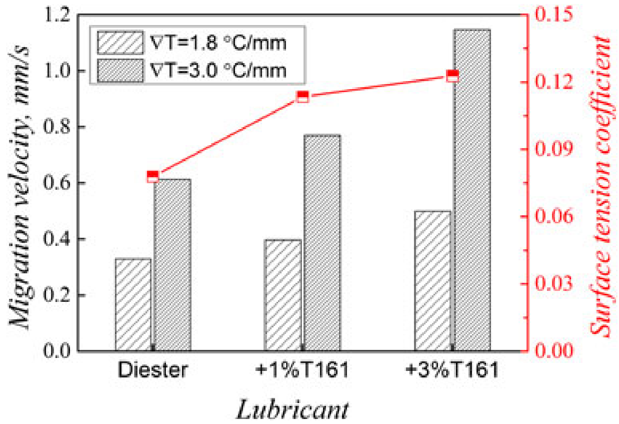

Figure 5. The effects of T161 on the migration behaviour and the surface tension coefficient of diester.

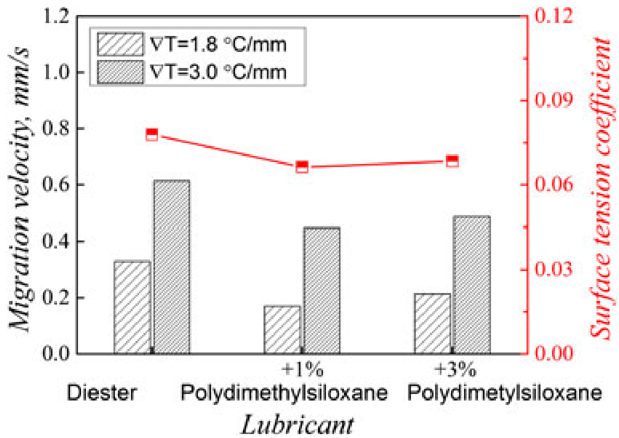

Figure 6. The effects of polydimethylsiloxane on the migration behaviour and the surface tension coefficient of diester.

was added into diseter, the migration velocity rapidly declined by 50%, just about 0.17 mm/s. As the content increases to 3%, the migration velocity increased a little compared with that of 1%, but it is consistently lower than the pure one. And a similar change rule of migration velocity was observed at the thermal gradient of 3°C/mm. For the surface tension coefficient, as the point-line presented, it was decreased when adding 1% polydimethylsiloxane, while as the content increased to 3%, the surface tension coefficient increased a little, but still lower than the pure one. The migration velocity and surface tension coefficient trends were quite similar. It is noted that adding polydimethylsiloxane into the lubricant will decrease the migration velocity and surface tension coefficient.

Figure 7 shows the influence of oleic acid on the migration velocity and the surface tension coefficient of diester. The histogram presents the relationship between migration velocity and oleic acid content. At a constant thermal gradient of 3.0°C/mm, the velocity of the droplet with content of 1% was nearly 0.45 mm/s, about 27% lower than the 0.61 mm/s observed for the pure diester. As the content

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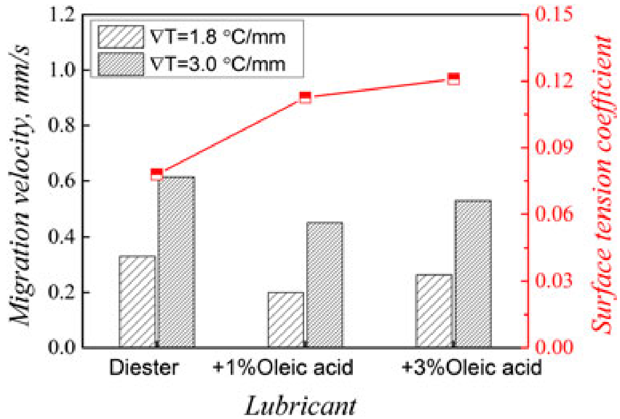

Figure 7. The effects of oleic acid on the migration behaviour and the surface tension coefficient of diester.

increased to 3%, the velocity was increased a little to 0.53 mm/s, but still lower than that of the pure diester. The point-line shows the influence of oleic acid on the surface tension coefficient, the surface tension coefficient of the lubricants was increased with increasing content of oleic acid.

Briefly, adding oleic acid into the lubricant is an effective way to decrease the migration velocity of lubricant.

The overall effects of the additives on the migration process can be summarised in Table II. And a comprehensive comparison on the effect of the four additives at the thermal gradient of 3°C/mm is shown in Figure 8. It is proven that adding additives to the pure diester will modify the migration behaviour significantly. In general, the migration velocity will increase with the increasing surface tension coefficient. Once the surface tension coefficient is lower than the pure one, the migration velocity will drop down correspondingly. However, there exists a special case for the oleic acid, when adding oleic acid into the diester, the surface tension coefficient was increased compared with the pure diester, but the migration velocity descends dramatically. Further analysis was necessary to investigate the mechanism and reasons.

DISCUSSION

Fluid dynamics

To aid in the description of the migration process, a side view of the two-dimensional liquid droplet on a unidirectional heated solid surface is shown in Figure 9. The droplet migrates from the warm side (*A*)

Table II. Overview of the effect of additives on the migration process.

Additives	*Descriptions*	*Surface tension coefficient*	*Migration velocity*
Extreme pressure agent	ZDDP	↑	↑
Ashless dispersant agent	T161	↑	↑
Anti-foam agent	Polydimethylsiloxane	↓	↓
Oiliness additive	Oleic acid	↑	↓

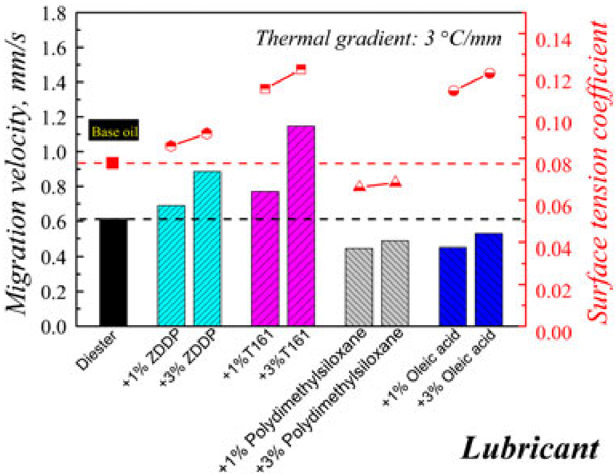

Figure 8. Comprehensive comparison on the effects of these four additives on the migration behaviour and the surface tension coefficient of diester.

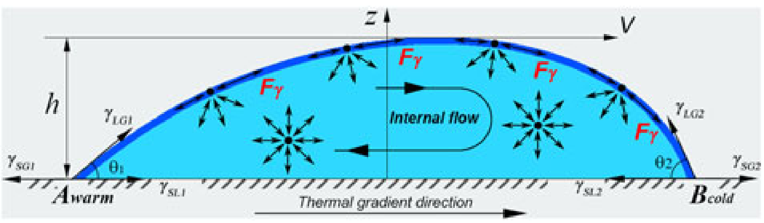

Figure 9. Side view of a two-dimensional droplet on a solid surface and the unbalance interfacial tensions at lubricant surface inducing the migration.

to the cold side (B). Young's equation defines the force balance between the interfacial tensions existing at the solid–liquid (γ_{SL}), solid–gas (γ_{SG}), or liquid–gas (γ_{LG}) interface and contact angle (θ). Under the effect of the thermal gradient, the solid–liquid interfacial tension γ_{SL2} at the front of the droplet is greater than γ_{SL1} at the rear surface; moreover, the substantial variations in the interfacial tension of lubricant provide a force (F_γ) acting on the lubricant surface. As the simplified graphic representation exhibits, these unbalance forces provide a tractive force on the droplet, generating an internal flow in the droplet and eventually driving the droplet migrating from the warm to the cold side.

Basing on the simplified model, the lubrication approximation theory is employed to determine the migration velocity, the thermal capillary migration can be specified by the balance between the tractive forces generated by interfacial tensions and the viscous forces, the migration velocity (V) can be governed by the formula:[33,34]

$$V = \frac{h}{2\mu}\frac{d\gamma}{dT}\frac{dT}{dx} \tag{2}$$

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where h is the thickness of the spreading droplet, μ is the viscosity of the liquid, $d\gamma$ is the surface tension change with the temperature dT, i.e. the surface tension coefficient γ_T, and dT/dx is the thermal gradient ∇T in x direction. It can be noticed that the migration velocity increases with decreasing viscosity. Since the viscosity decreases with increasing temperature, the migration velocity will be faster under higher temperature.

This formula indicates that the migration velocity has a proportional relationship with the surface tension coefficient. Therefore, it can be understood the effects of additives on the migration performance. When adding ZDDP and T161 into the base oil, the surface tension coefficient was increased, so the migration velocity V was increased correspondingly. When adding polydimethylsiloxane into the base oil, the surface tension coefficient was decreased, so the migration velocity V was dropped down. However, for the oleic acid, when it was added into the base oil, the surface tension coefficient was increased, the migration velocity was decreased on the contrary.

It is known that migration always occurs on the metal surface, to understand the functional mechanisms of additives, not only the variations of surface tension coefficient induced by additives, but also the interaction effect between the additives and the metal surface should be taken into consideration.

Molecular interaction effect

To gain insights into the mechanism of these four additives on the migration behaviour, sketches of the interaction effect between additives and the metal surface are shown in Figure 10. Figure 10a shows a simple representation of the structural formula of ZDDP and the formation process of the metal polyphosphate layer. As the ZDDP incorporated into the lubricants, it is adsorbed on the metal surface, and only under extreme working conditions, decomposition and surface reaction will take place, and a metal polyphosphate layer will be generated. This reaction film acts as a protective barrier to prevent direct metal contact and adhesion.[35] However, the migration experimental conditions did not meet the

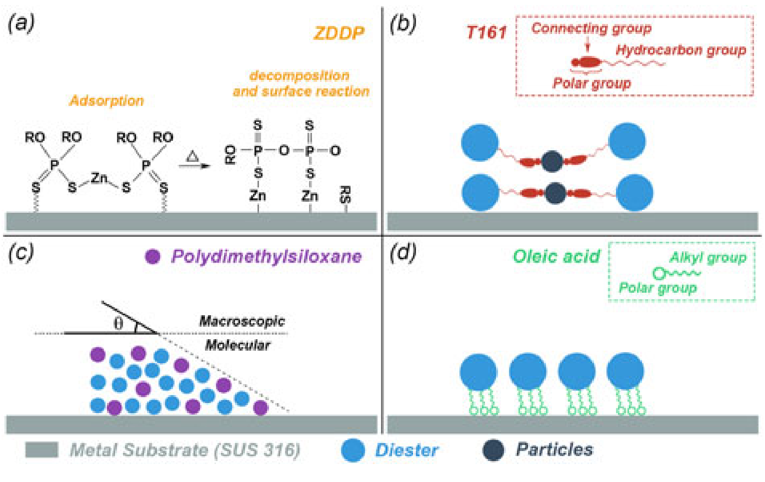

Figure 10. Mechanisms of the four additives on the metal surface: (a) Schematic of anti-wear film formation by ZDDP on the metal surface. (b) Graphic representation of the dispersant molecule and mechanism of particle–additive interaction. (c) Schematic of the functional mechanism of polydimethylsiloxane molecular on diester. (d) Graphic representation of oleic acid and functional mechanism with diester and metal surface.

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requirement of reaction (high temperature and pressure) in this study; herein, ZDDP is regarded as a surfactant, which increases the surface tension coefficient of the lubricant.

Figure 10b shows the mechanism of dispersant of T161 on the lubricant. Chemically, a dispersant consists of three distinct structural features: a hydrocarbon group, a connecting group, and a polar group.[36] The polar head group of the dispersant is attracted to the polar particles from contamination. The hydrocarbon tail prevents the contaminant particles from coming close enough to interact, keeping such particles suspended in the base lubricant. Actually, during the migration behaviour, no particles or soot was involved in the lubricant. It means that the dispersant (T161) can also be recognised as a surfactant in the lubricant, which increases the surface tension coefficient of the lubricant.

Polydimethylsiloxane is characterised by its low surface tension. As shown in Figure 10c, when it was incorporated into the diester, the contact angle of the lubricant would be decreased at the macroscopic. Moreover, it is important to realise that the lubricant migration is driven by a surface tension gradient between the front and the rear of the spreading droplet. The existence of this polydimethylsiloxane is thought to act as an inducement force eliminating the surface tension gradient generated by the thermal gradient; consequently, the migration velocity will drop down.

Figure 10d presents the mechanism of oleic acid on the lubricant. It is known that oleic acid is usually polar in nature; it consists of two structural features: a polar group and an alkyl group. The polar molecules in oleic acid have strong affinity to the metal surface, as well as the alkyl group has a good oil solubility. When the oleic acid was incorporated into the diester, the adsorption ability of the lubricant was extremely enhanced. The lubricant would be adsorbed on the metal surface, forming dense and stable film. As a result, it became difficult for the lubricant to migrate. Therefore, when the oleic acid incorporated into the lubricant at first, the migration velocity was decreased. With the increasing content, the adsorption ability would not be enhanced further. However, the surface tension coefficient was increased all the time, and the migration velocity was increased with increasing surface tension coefficient. That is the reason why the velocity first decreases and then increases with the increasing content of oleic acid.

CONCLUSIONS

In this study, experiments were carried out to investigate the effects of additives on the thermal gradients induced migration of lubricant. Four different additives were employed and the influence of content was studied. Particularly, the mechanisms of the lubricant additives on the migration behaviour were investigated. It is proven that additives can modify the migration property of lubricant significantly. The conclusions drawn from this study are as follows:

1. Even only with a little amount, the additives could have remarkable influences on the migration performance of lubricant. The reasonable use of additives can modify the thermal capillary migration behaviour effectively.
2. The oleic acid and polydimethylsiloxane can slow down the migration of lubricant.
3. For the additives of ZDDP and T161, adding them into the lubricant will increase the surface tension coefficient, which will accelerate the migration of lubricant. If these additives are essential, low content should be better.
4. Not only variation of surface tension coefficient induced by the additives, but also the interaction effect between additives and the metal surface should be considered in the designing process of anti-migration lubricants.

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