

The effect of friction on micropitting

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ABSTRACT

Micropitting is a type of surface fatigue damage that occurs in rolling-sliding contacts operating under thin oil film conditions. It is caused by stress fluctuations, brought about by surface asperity interactions, which lead to initiation and propagation of numerous surface fatigue cracks and subsequent loss of material. Despite its increasing importance to gear and bearing reliability, the mechanisms of micropitting are poorly understood. This is particularly the case concerning the effects of friction on micropitting which are difficult to study under controlled conditions. This is because it is difficult to isolate the friction effects from other influential factors, in particular from the build-up of any anti-wear tribofilm and its subsequent effect on the running-in of counterface roughness that is known to strongly affect micropitting through its influence on severity of asperity stresses. This paper presents new data on the impact of friction on micropitting obtained using a new test methodology. Micropitting tests were conducted using a ball-on-disc MTM rig with the additional functionality to continuously monitor the growth of tribofilm during the test. Friction was varied by using custom-made oils containing different concentrations of MoDTC. Crucially, the effect of friction was isolated from the effect of counterface roughness running-in by introducing the MoDTC blend only after the running-in period was completed with a ZDDP solution alone. This approach eliminates the influence of MoDTC on ZDDP anti-wear tribofilm growth in early stages and hence ensures the same running-in takes place in each test. This gives similar asperity pressure history, regardless of the amount of MoDTC present.

Results show that friction has a very significant impact on micropitting; for example, the extent of micropitting was reduced by a factor of 10 when friction coefficient was reduced from about 0.1 to 0.04. Lower friction results in fewer surface cracks which grow at a shallower angle to the surface than those at higher friction. Numerical analysis of contact stresses present under tested conditions indicates that the primary mechanism by which friction affects micropitting is through its effects on near surface stress fields: reducing friction reduces the local tensile and shear stresses in the near surface, asperity-influenced region, which may in turn be expected to mitigate crack initiation and propagation. The results presented may help in designing oil formulations that can extend component lifetimes with respect to both wear and micropitting damage through controlling tribofilm growth and friction.

1. Introduction

Micropitting is a type of surface fatigue damage that occurs in lubricated rolling sliding contacts operating under thin lubricant film conditions. In such conditions, surface asperity interactions lead to asperity stress fluctuations and hence accumulation of fatigue damage at asperity level. This results in initiation of numerous surface cracks which then propagate and interact to cause fragments of material to detach from the surface. Micropitting therefore manifests itself as numerous small pits, tens of microns in size, that give the surface a frosted appearance. If unchecked, micropitting can result in a substantial loss of

material and eventual component failure such as, for example, through a significant loss of a gear tooth profile which may in turn lead to tooth pitting and breakage. Micropitting is increasingly becoming one of the most important failure modes in gears and rolling bearings because of increases in the power density of mechanical systems and the introduction of low viscosity lubricants, with consequently thinner lubricant films and increased asperity interactions [1]. Despite this, the underlying mechanisms of micropitting are poorly understood and there are currently no universally accepted design criteria. Some of the factors recognized to effect micropitting include lambda ratio [2,3], contact pressure [2,4], surface roughness [5–9] particularly of the harder

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counterface [7,9], sliding magnitude and direction [2,4], hardness [8,10,11] and oil anti-wear [7,9] and friction behaviour [6]. While the exact effects of many of these factors are still under investigation, the influence of friction on micropitting is particularly difficult to elucidate because of the difficulties in isolating friction from other influential factors during micropitting experiments.

Because micropitting is a fatigue process it is strongly dependent on the asperity stresses that promote crack development. In a rubbing, rough surface contact, these stresses arise from applied normal load and friction. Precise mechanisms leading to initiation of contact fatigue cracks are not entirely established, but it is accepted that accumulation of plastic strain plays an important role [12]. Since it contributes to the overall contact stress levels, friction promotes occurrence of plasticity within the contact, and hence increases the risk of crack initiation. Once initiated, surface cracks can propagate under the action of tensile (mode I) and/or shear stress (mode II) [13]. On a macro contact scale, higher contact friction is recognized to generate higher tensile stress at the rear of the contact region [14–17]. Hamilton et al. [15] have shown that the magnitude of this tensile stress in a point contact depends on a factor $(1 + A\mu)$, where $A = 3\pi(4 + \nu)/8(1 - 2\nu)$, ν is Poisson's ratio and μ is friction coefficient. When ν is 0.3, as is the case for steels, A is about 12, so that according to this relationship, the magnitude of tensile stress at the rear of the point contact increases by about 65% when friction coefficient increases from $\mu = 0.04$, which is typical of a well lubricated, elastohydrodynamic contact, to $\mu = 0.12$, typical of a contact operating in a boundary regime. The higher tensile stress increases the mode I stress intensity factor (SIF) which increases the crack propagation rate in a rolling-sliding contact [18–20]. As well as increasing tensile stresses, higher friction increases contact shear stresses that are widely recognized to promote fatigue damage [12,21–23], with the orthogonal shear stress amplitude being commonly used as the critical stress value in rolling bearing fatigue life predictions [24]. Growth of surface cracks under rolling-sliding contact is believed to be further promoted by the hydraulic action of oil that is able to enter a surface breaking crack and is then pressurized during subsequent over-rolling [4,25–27]. Friction probably plays a crucial role in this mechanism also, by opening the mouth of the surface breaking crack ahead of the contact which aids oil entry into the crack.

The discussion above considers the potential effects of friction on stresses and propagation of surface cracks on the macro contact scale. In terms of micropitting, it is the stresses in asperity micro-contacts that are of relevance, but it is expected that the same general trends described above exist in these micro contacts. However, the random nature of surface roughness generally introduces additional complexities that make it difficult to determine and track the precise asperity stress history during rolling-sliding. Kim and Olver [28] use a numerical model of dry rough contact to evaluate asperity stress history in a rough, rolling-sliding contact to illustrate how the counterface roughness and sliding speed can contribute to increased surface fatigue damage. Morales-Espejel et al. [29] go a step further and illustrate the effect of various contact conditions on micropitting damage itself, using a numerical model which accounts for lubrication conditions and the evolution of roughness due to running-in. Kadiric et al. [30] show that the magnitudes of the local maximum shear stresses within individual micro-contact regions correlate well with asperity slopes, while their depth is related to the size of micro-contacts and hence the wavelength content of the roughness. Rycerz et al. [23] present experimental data to show that the propagation rates for short surface cracks (tens of microns in length) are different from those of long cracks; they attribute this to the fact that the growth of short surface cracks is governed by the asperity stress fields, since they are located in the near surface region, whereas the growth of longer cracks is governed by the macro stress field. More recently, the same authors [31] have shown that increasing the contact friction coefficient significantly increases the propagation rates of such short cracks, but that the effect of friction on crack propagation rates diminishes as the crack length increases, *i.e.* as the crack

grows deeper into the material and so further away from the asperity influenced region near the surface.

Given its effects on initiation and growth of surface cracks as described above, reduction of friction is clearly one potential measure that may help to reduce micropitting. However, there appear to be very few studies that directly investigate the effect of friction on micropitting. Morales-Espejel et al. [5] used a numerical model to show that increasing the boundary friction coefficient from 0.12 to 0.15 almost doubled the extent of micropitted area for a given set of contact conditions. Perhaps the most notable study on the effects of friction on micropitting is that of Lainé et al. [6] using triple-disc machine (MPR) and lubricant blends with and without molybdenum dialkylthiocarbamate (MoDTC) friction modifier to produce different friction coefficients in their micropitting tests. They showed that a reduction of friction from 0.08 to 0.04, achieved via addition of MoDTC (0.12 wt% of Mo) to their test oil, significantly reduced micropitting damage compared to the original oil which contained zinc dialkylthiophosphate (ZDDP, 0.10 wt% of P) only. The authors suggested that friction was decreased by the formation of an MoS_2 tribofilm from MoDTC on the rubbing surface of the counterface MPR disc. More recently, Soltanahmadi et al. [32] showed that addition of ashless amine friction modifier can also reduce micropitting in a similar manner to that of MoDTC.

These previous studies varied boundary and mixed friction coefficient by using oils with different amounts of friction modifier additive in a test oil containing an anti-wear additive. However, the addition of friction modifiers such as MoDTC and ashless amine compounds in this manner has been shown to slow the growth of antiwear tribofilms because of competitive adsorption with antiwear additives [32,33]. Such slower growth of anti-wear tribofilms with a friction modifier and ZDDP blend will result in a higher reduction of counterface roughness during the running-in phase than is the case with a ZDDP solution alone and this early reduction in roughness should in itself reduce micropitting. Thus a previous study by the authors using a mini traction machine with spacer layer imaging (MTM-SLIM) to monitor tribofilm growth and micropitting in parallel, showed that the amount of micropitting produced was strongly affected by the rate of ZDDP tribofilm growth during the running-in phase [7]. It was shown that early tribofilm growth rate that inhibits running-in strongly promotes micropitting.

This dual effect of friction modifiers, of both reducing friction and of influencing surface roughness, has so far made it impossible to isolate and thereby quantify the effect of friction on micropitting. To address this problem, the current paper describes a test methodology that disentangles the effect on micropitting of boundary friction from that of counterface roughness evolution. The approach involves conducting micropitting tests in which an oil containing only a ZDDP antiwear additive is used during the early stage until the running-in of the counterface roughness is complete. Only then is a chosen concentration of MoDTC introduced into the test oil in order to influence friction. Thus, by varying the concentration of MoDTC in the test oil, it is possible to conduct micropitting tests with different boundary friction coefficients but negligible differences in counterface roughness evolution, hence isolating the effect of friction on micropitting.

2. Test methodology

2.1. Experimental equipment

All micropitting tests were conducted using the same experimental set-up as described in a previous study by the authors [7]. A mini traction machine (MTM) ball-on disc tribometer with a spacer layer imaging attachment (SLIM) was employed to generate micropitting on the ball specimen and to monitor tribofilm formation within the same test. The test rig is shown schematically in Fig. 1. A steel ball is loaded against a steel disc immersed in a lubricant. The ball and the disc are driven by

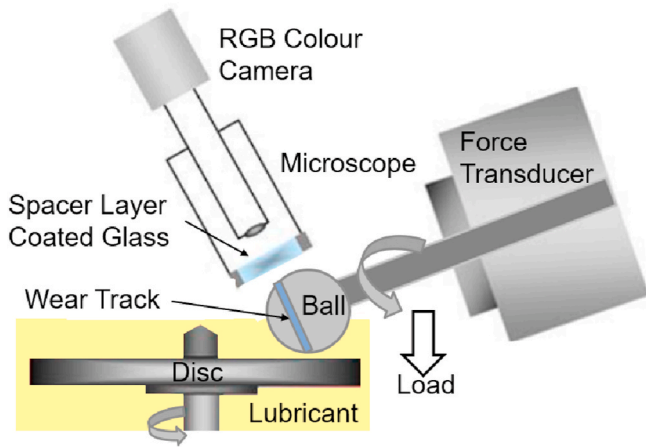


Fig. 1. A schematic illustration of the MTM-SLIM rig.

separate electric motors at user-specified rolling/sliding conditions. To capture a SLIM image, the rubbing test is paused and the ball is raised and loaded against a glass flat. The glass flat surface is coated with a semi-reflective chromium layer on top of which is a transparent silica space layer. Light reflected from the chromium layer and the ball surface, undergo optical interference upon recombination, generating an interference image of the contact between the ball and the glass surface which is then captured by an RGB camera. Tribofilm thickness is calculated from this interferometry image using calibrated RGB values.

2.2. Test conditions

Table 1 shows the materials and conditions employed in the MTM micropitting tests. Disc roughness and ball hardness are controlled to promote micropitting on the ball. Standard MTM tests employ very smooth discs with centre-line average roughness, R_a of ~ 10 nm.

Table 1
MTM specimen materials and test conditions.

Ball specimen	Material: AISI 52100 steel; diameter 19.05 mm; measured hardness: 730–780 HV; measured roughness $R_a \sim 0.01 \mu\text{m}$
Disc specimen	Material: AISI 52100 steel; measured hardness: 800–850 HV; measured roughness $R_a = 0.42 \pm 0.02 \mu\text{m}$
Test temperature ($^{\circ}\text{C}$)	65
Test Load (N)	75 (Maximum Hertzian pressure: 1.25 GPa, Hertzian contact radius: $170 \mu\text{m}$)
Entrainment speed (m/s)	3
Slide-roll-ratio (SRR)	-0.05
	$2 \cdot (U_{\text{ball}} - U_{\text{disc}}) / (U_{\text{ball}} + U_{\text{disc}})$
Test cycles	8 million cycles: Test paused for specimen inspection after 0.1 million cycles, 1 million cycles, 4 million cycles, 8 million cycles

Table 2
Test oil formulations.

Lubricant name	Base oil	ZDDP (secondary-primary mixed), P concentration, ppm	MoDTC, Mo concentration, ppm
ZDDP	PAO 10	800	0
ZDDP + Mo(25)	PAO 10	800	25
ZDDP + Mo(50)	PAO 10	800	50
ZDDP + Mo(100)	PAO 10	800	100
ZDDP + Mo(200)	PAO 10	800	200
ZDDP + Mo(500)	PAO 10	800	500

However, in this study, disc specimens with $R_a \sim 0.4 \mu\text{m}$ were used in order to subject the ball to severe asperity contact stress cycles and hence promote micropitting on the ball. This higher roughness is also more representative of that present in gears and bearing raceways. As is common in controlled contact fatigue tests [4,6,32,34], the specimens on which micropitting damage is to be studied, in this case the MTM balls, were made softer than the rough disc counterfaces; this ensures that disc roughness persists during the tests, thereby promoting micropitting on the ball. This was achieved by softening the standard MTM steel balls (martensitic through-hardened AISI 52100 steel) from their normal value of 845 ± 25 HV down to a measured range of 755 ± 25 HV, by holding the balls at 250°C for 1 h in pure polyalphaolefin (PAO 100) base oil in an air atmosphere followed by slow cooling. Mean speed, slide roll ratio (SRR), load and test temperature were held the same in all tests. It should be noted that, in common with other controlled micropitting studies [4,6,32,34], the surface speed of the ball specimen where the micropitting is being generated is slower than that of the disc. This means that the SRR is negative using the SRR definition specified in Table 1. MTM-SLIM tests were conducted for a fixed duration that subjected each position on the ball track to 8 million Hertzian contact cycles. These test conditions were chosen to reliably generate micropitting while suppressing other failure modes such as wear, as well as being representative of bearing and gear operations. The authors discuss the reasoning behind the chosen conditions in Ref. [7] but in brief; the maximum pressure achievable on the MTM (1.25 GPa) is used here as higher pressure promotes micropitting as long as pitting is avoided [4], the entrainment speed of 3 m/s is chosen to maximise the rate of accumulation of contact cycles, hence accelerating micropitting, while still keeping the lambda ratio close to 0.5 which is known to increase risk of micropitting, and finally, SRR of 5% is representative of bearings and gears while being low enough not to produce significant adhesive wear which competes with micropitting as the dominant failure mode [1,4,7]. The test temperature of 65°C is representative of many operating gears and rolling bearings and is high enough for the relevant tribofilms to form in a reasonable time while minimising oil ageing during the 40 h test.

2.3. Test lubricants

50 ml of oil was used for each test. Test oils were formulated as shown in Table 2. Varying concentrations of MoDTC (Adeka Sakura Lube 165 at 0, 25, 50, 100, 200 and 500 ppm Mo) and ZDDP (secondary-primary) at a fixed concentration of 800 ppm P were mixed with PAO 10 ($62.8 \text{ mm}^2/\text{s}$ and $9.9 \text{ mm}^2/\text{s}$ kinematic viscosity at 40°C and 100°C , respectively). In all experiments the initial lambda ratio was around 0.3 (determined from a calculated minimum oil film thickness of 150 nm), where lambda is defined as the ratio of the calculated EHD film thickness to the initial composite root mean square roughness of the two surfaces. Following the running-in, lambda ratio was 0.5 in all tests.

2.4. Test procedure

Prior to each test, the ball and disc specimen were cleaned in toluene. The specimens were then mounted in the MTM rig, the oil bath filled with the chosen lubricant and the rolling-sliding test started. Tests were

interrupted after 0.1, 1, and 4 million cycles to observe and measure wear and micropitting on the ball and to measure the roughness of the disc. Prior to making these surface measurements, ball and disc were rinsed in toluene in an ultrasonic bath for 10 min to remove supernatant oil. Rinsing the ball and disc with toluene did not influence the thickness of the ZDDP tribofilm which, after the first hour of rubbing, can generally only be removed by chemically-active solutions such as EDTA [35]. Micrographs of surface cracks and micropitting on the ball were obtained using an optical microscope. The extent of micropitting was quantified as the percentage micropitted area on the ball rubbing track. This was obtained by taking a series of optical images to cover the entire ball wear track and then manually measuring the size of the area affected by micropitting in each image as a percentage of the whole wear track. The reported value is the average of all values obtained and therefore represents an estimate of the total extent of micropitting around the whole circumference of the ball. As well as determining the micropitted area on the ball, the amount of any mild wear on the ball was assessed by measuring the loss of material in the rubbing track away from any micropitted areas using a stylus profilometer. Negligible amounts of wear were measured in all tests used to assess micropitting, regardless of the level of MoDTC. The Ra surface roughness of the discs was also measured with the stylus profilometer at four different locations spaced at 90° around the disc track; the average of these four measurements is reported here in all cases. The maximum and minimum measured Ra roughness values were within 0.03 μm of this average in all cases. After these surface measurements, the specimens were returned to the MTM and the rolling-sliding test continued.

To isolate friction from the effect of counterface roughness, tests were run using two methods as shown schematically in Fig. 2. In Method 1, the entire test was conducted with a single test oil (ZDDP and ZDDP + Mo at concentrations of 25, 50 or 500 ppm). Method 2 was designed to isolate friction effects by achieving the same running-in, and hence the same disc roughness during the micropitting test, regardless of the amount of MoDTC used. In Method 2, the test was first run for 0.1 million cycles, corresponding to duration of the running-in phase, with the ZDDP only oil; the oil was then replaced with one containing the chosen concentration of MoDTC, i.e. one of ZDDP + Mo at 50, 100, 200 or 500 ppm solutions in Table 2. This is because our previous study showed the reduction of disc roughness due to running-in ceased after 0.1 million cycles, after which the disc roughness remained more or less the same to the end of the 8 million cycle test [7]. Before oil replacement at 0.1 million cycles, the specimens were removed for inspection as described above and the MTM test chamber was cleaned with toluene.

During tests, the rubbing was periodically paused to measure the ZDDP tribofilm thickness using SLIM; this is done automatically by the control software and does not necessitate specimen removal. At the end of the micropitting tests, in addition to inspecting the surfaces of the specimen as described above, a polished cross-section of the ball rubbing track in the rolling-direction was prepared and inspected with an optical microscope. This is to investigate the morphology of the generated surface cracks and micropits, and establish any correlation of these with friction. All tests shown in this paper were repeated twice.

2.5. Calculation of contact pressure and stress

In order to better interpret the observed trends, contact pressure distribution and subsurface stresses in the ball-disc contacts at the applied normal load and measured friction were calculated using an existing numerical model for contacts of rough surfaces [16,36,37]. The model predicts asperity pressures and the corresponding sub-surface stresses for the given normal load and friction, and uses real measured rough surfaces with no assumptions on asperity shapes or distribution. The numerical analysis included the effect of ZDDP tribofilm on contact pressure distribution by using a version of the above model applicable to multi-layered surfaces [38,39] and representing the tribofilm with a layer of uniform thickness (set equal to 175 nm, which was the maximum tribofilm thickness measured in this study) on top of the rough steel substrate. In this analysis, Poisson's ratio of the ZDDP tribofilm was set to 0.26 and the Young's modulus to 37.7 GPa. The latter was measured using nano-indentation as described below. The corresponding values for the steel substrate were 0.3 and 207 GPa.

2.6. Tribofilm elastic modulus

To calculate the contact pressure distribution in the presence of tribofilm, the elastic modulus of ZDDP tribofilm formed on MTM balls was measured using a nanoindentation method. A Nano Indenter G 200 with a Berkovich tip manufactured by Keysight Technologies was employed. The mechanical properties of tribofilm vary with depth, so they cannot be properly measured at the point of the initial unload [40]. Therefore, ZDDP tribofilm was measured in the Continuous Stiffness Measurement (CSM) mode, which allows measurement of the contact stiffness at any point along the loading curve [41]. To mount a ball on a flat sample stage, the ball was cut to obtain a flat surface with the wear track at the top of the curved ball segment. Several locations were indented on the wear track of the ball to obtain representative results. The elastic modulus at each indentation depth was determined by analyzing the amplitude and phase response. The frequency and displacement amplitude values were 45 Hz and 2 nm, respectively and the surface approach velocity was 10 nm/s. Under a load range of 50 mN, a target depth of 500 nm and a strain rate of 0.01 s^{-1} , 25 points at 25 μm intervals inside and outside of the wear track were measured. All tests were measured at room temperature. Elastic modulus was estimated using 0.26 for Poisson's ratio of ZDDP tribofilm [42], and the standard deviation was then calculated.

3. Results

3.1. Effect of MoDTC addition at the beginning of the test (method 1 in Fig. 2)

This section presents results of friction, surface damage and tribofilm formation when MoDTC was present from the beginning of the tests. The lubricants used are a ZDDP-only solution and three solutions consisting of ZDDP and MoDTC at concentrations of 25, 50 and 500 ppm. The same lubricants were used from the beginning to the end of the tests with no

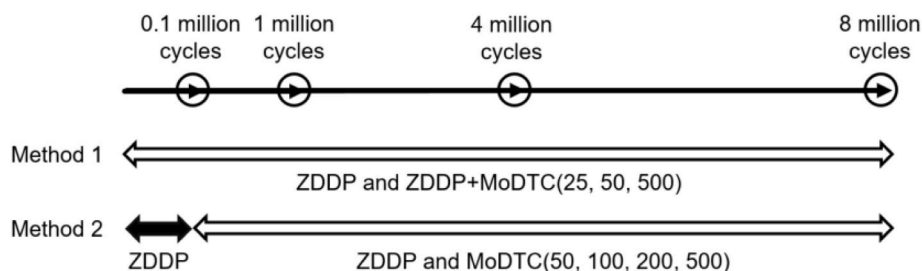


Fig. 2. Micropitting test procedures.

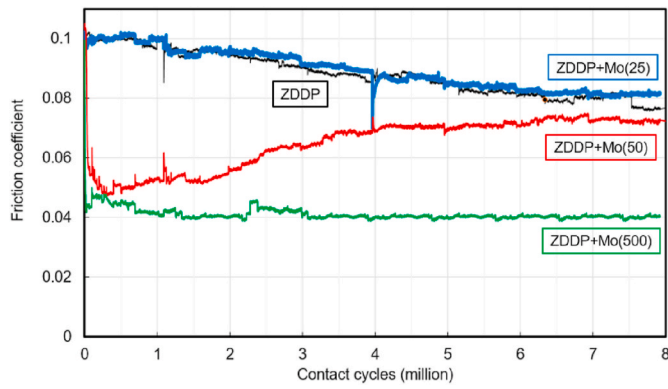


Fig. 3. Friction behaviour in tests conducted with oils having different concentrations of MoDTC from the outset.

replacement.

3.1.1. Friction behaviour

Fig. 3 shows the evolution of friction coefficient in tests conducted with oils having different Mo concentrations. The friction coefficient of the Mo-free ZDDP oil was initially around 0.1 and gradually decreased to reach 0.08 at the end of the test (8 million contact cycles). The addition of 25 ppm of Mo did not reduce friction, resulting in the same friction coefficient as the ZDDP-only oil. 50 ppm and 500 ppm of MoDTC reduced friction coefficient from 0.1 to about 0.04–0.05 almost immediately after starting the tests. While the friction coefficient of ZDDP + Mo(500) remained at 0.04 throughout the test, the friction coefficient of ZDDP + Mo(50) gradually increased again to reach 0.07 at 4 million cycles, and then remained stable at this value to the end of the test. It should be noted that the reduction in friction with increasing MoDTC is not due to any extra smoothing of the specimen surface roughness since it occurs very rapidly, and before any measurable change in

surface roughness due to running-in takes place. Instead friction reduction is due to the well-established mechanism of MoDTC action originating from the formation of nanocrystals of low shear strength MoS_2 on rubbing asperities [43–45]. Since MoS_2 forms on contacting asperities that support load, it has a strong effect on reducing friction in the mixed lubrication regime conditions present in this study. MoS_2 formed in this way is not durable, but is continually being formed and rubbed off [46,47]. It is likely that 50 ppm of Mo was not sufficient to reduce friction throughout the test because of the diminished ability for this renewal of the low friction film and because of the oil aging; this is discussed in section 3.3. It should also be noted that the momentary blips in the recorded friction traces apparent in the figure are artefacts of the measurement method on the MTM-SLIM rig and occur just after motion was paused to capture a SLIM image.

3.1.2. Evolution of surface damage on balls

Fig. 4 shows representative optical micrographs of the rubbed track on the balls after 0.1, 1, 4 and 8 million cycles with different MoDTC concentrations, as well as measured percentage of micropitted area on the ball wear track after 8 million cycles. The severity of micropitting on the wear tracks varied with MoDTC concentration (Fig. 4). ZDDP and ZDDP + Mo(25) showed similar densities of surface cracks. After 0.1 million cycles these cracks were tens of microns in length and appeared to grow across the rolling track in a fanned, shallow ‘V’ shape, typical of surface-initiated fatigue. The appearance of these cracks is the first visible sign of micropitting taking place. As the test progressed, the number of cracks increased considerably. Small pits (tens of microns in size) associated with these cracks became apparent at 4 million cycles. By the end of the test, these micropits covered 25–27% of areas of the rubbing track. By contrast, with ZDDP + Mo(50 and 500), far fewer cracks and micropits were observed. With ZDDP + Mo(50) a few isolated cracks are present after 1 million cycles. The density of these cracks increased after 4 and 8 million cycles and covered 8.9% of area of the rubbing track after 8 million cycles, but was clearly significantly less

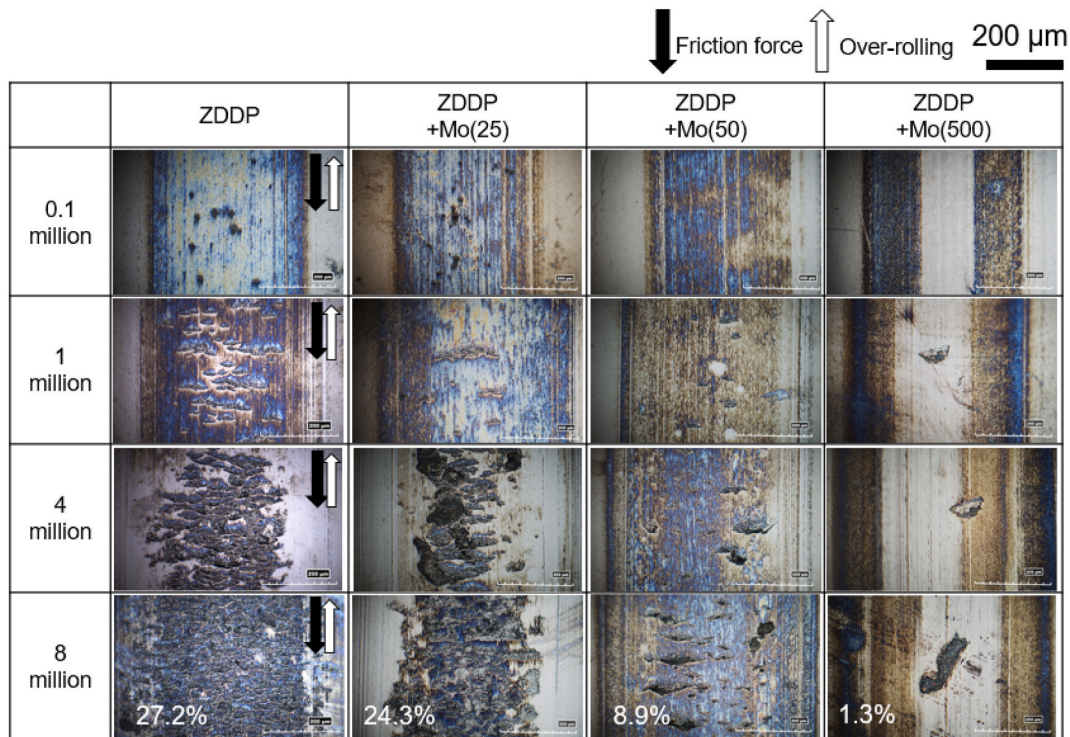


Fig. 4. Typical examples of optical micrographs of wear tracks on balls from tests conducted with different MoDTC concentrations from the outset as well as measured percentage of micropitted area on ball wear track after 8 million cycles (Note: the scale bar included in the top right corner of the figure applies to all images shown).

than in the case of ZDDP only and ZDDP + Mo(25) oils. In the case of ZDDP + Mo at 500 ppm, the highest concentration used here, only a very few very isolated micropits are apparent and the surface damage was minimal, covered 1.3% of areas of the rubbing track even after 8 million cycles.

The bluish colour evident in the wear tracks in Fig. 4 is caused by ZDDP tribofilms, and hence allows a qualitative comparison of the effect of Mo concentration on ZDDP tribofilm growth, which is an important factor to consider in isolating the influence of friction on micropitting as explained earlier. In the tests with ZDDP and ZDDP + Mo(25 and 50) a tribofilm is formed over most of the wear track width after 0.1 million cycles. In contrast, in the tests with ZDDP + Mo(500), tribofilm is only apparent in the regions towards both edges of the wear track, but not in the central region where the contact pressure is highest (Fig. 4). Similar results have not been previously reported and may be a consequence of the unusual MTM test conditions used here to produce micropitting. However, considering that friction was low from the start of the test it is likely that a large amount of MoS₂ tribofilm formed in the central, high pressure region of the wear track relatively quickly after the start of the test and this may compete with ZDDP adsorption to reduce ZDDP tribofilm formation during the test [6]. Indeed, the low friction resulting from MoS₂ formation may lower the shear stress at asperities to the extent that the mechanochemically-driven reaction of ZDDP is no longer possible or is very slow [48,49]. Any MoS₂ tribofilm formed would be much thinner than ZDDP tribofilm [50] so that it would not necessarily be apparent in the optical micrographs.

3.1.3. Evolution of surface roughness of counterface discs

Fig. 5 shows the evolution of the disc roughness during tests. In all cases the disc roughness reduced in the initial stages of the test and then remained relatively stable, a behaviour indicative of running-in. However, considerable differences are evident in the level of roughness reduction with different MoDTC concentrations. ZDDP and ZDDP + Mo(25) oils reduced disc roughness less than the higher MoDTC concentrations ZDDP + Mo(50 and 500). In the former cases, the roughness was reduced from the initial Ra value of $\sim 0.45 \mu\text{m}$ to $0.30 \mu\text{m}$ after 0.1 million cycles and stayed constant at this level for the rest of the test. By contrast, oils with 50 and 500 ppm of MoDTC reduced disc roughness from the initial $\sim 0.45 \mu\text{m}$ Ra to around $0.25 \mu\text{m}$ Ra after the first 0.1 million cycles and to $\sim 0.22 \mu\text{m}$ Ra after 1 million cycles, after which roughness remained at this value for the rest of the tests.

3.1.4. Evolution of tribofilm

Fig. 6 shows tribofilm thickness in the middle region of the ball rubbed tracks obtained from SLIM images. ZDDP and ZDDP + Mo(25) formed tribofilms rapidly during the first 0.1 million cycles, to reach

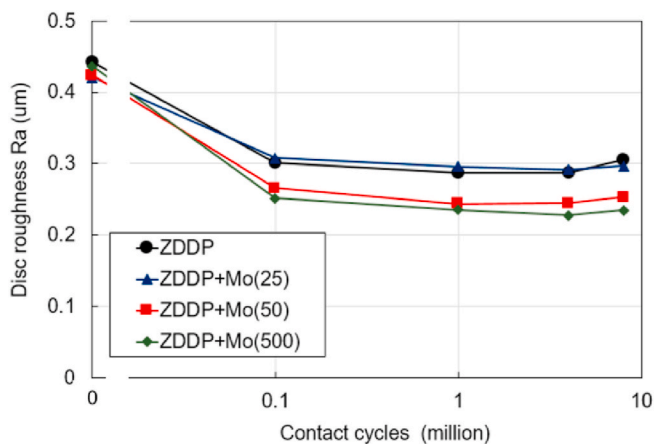


Fig. 5. Disc roughness evolution with oils containing different MoDTC concentrations when MoDTC is present from the start of the test.

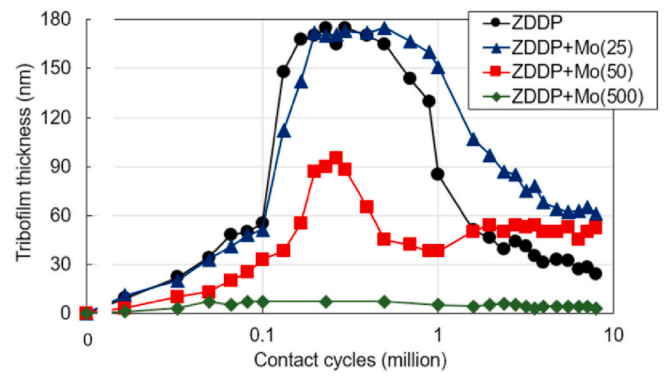


Fig. 6. Tribofilm evolution in tests with oils containing different MoDTC concentrations present from the start of the test.

175 nm after 0.2 million cycles; after this, tribofilms thickness decreased slowly to stabilize at around 30 nm and 60 nm, respectively. ZDDP + Mo(50) formed tribofilm at a much slower rate in the first 0.1 million cycles, to reach around 90 nm after 0.3 million cycles before decreasing to 60 nm by 1 million cycles. By contrast, with ZDDP + Mo(500) oil negligible film thickness was observed using SLIM in the central area of the rubbed wear track. This result suggests that the addition of over 50 ppm of MoDTC hinders ZDDP tribofilm formation. Interestingly, after about 1 million cycles, the presence of low concentrations of MoDTC in the ZDDP solution resulted in thicker tribofilm thicknesses than ZDDP alone. MoDTC is probably functioning as an antioxidant, reducing the rate of depletion of tribofilm-forming ZDDP [51].

The results in this section show that although addition of more than 50 ppm of MoDTC to the ZDDP oil from the beginning of the test reduces friction coefficient and visibly reduces micropitting damage, the counterface roughness is also reduced because MoDTC slows ZDDP tribofilm growth during running-in. This makes it impossible to establish the effect of friction on micropitting from these results alone since the observed reduction of counterface roughness itself is known to strongly reduce micropitting.

3.2. Effect of MoDTC addition after running-in (method 2 in Fig. 2)

This section presents the results of tests designed to investigate the influence of concentration of MoDTC when the latter is introduced after running-in for an initial 0.1 million cycles with an oil containing ZDDP only. Four different concentrations of Mo were tested, i.e. 50, 100, 200 and 500 ppm, blended with a ZDDP containing oil. ZDDP + Mo(25) was not tested here because no friction reduction was seen with this blend using method 1 as described above.

3.2.1. Friction behaviour

Fig. 7a shows the evolution of friction coefficient in tests with different MoDTC concentrations over the 8 million cycles. Fig. 7b shows the same data, but zoomed in to 0–0.5 million cycles range. It is evident that for all four Mo concentrations, friction coefficient decreased from the initial value of 0.1 obtained with ZDDP oil to 0.04–0.05 after the addition of MoDTC at 0.1 million cycles. The friction coefficient at Mo concentrations of 50 and 100 ppm then increased slowly after 1 and 3 million cycles and stabilised at 0.06 and 0.07, respectively. This slow increase in friction coefficient is probably caused by oil aging. By contrast, for ZDDP + Mo(200 and 500) the friction coefficient remained low, at between 0.04 and 0.05, throughout the test duration.

3.2.2. Evolution of surface damage on balls

Fig. 8 shows representative optical micrographs of rubbed tracks on balls from tests lubricated with ZDDP for the first 0.1 million cycles and then ZDDP + MoDTC(50, 100, 200 and 500) for the rest of each test. Fig. 9 shows optical micrograph and SEM images of polished sections of

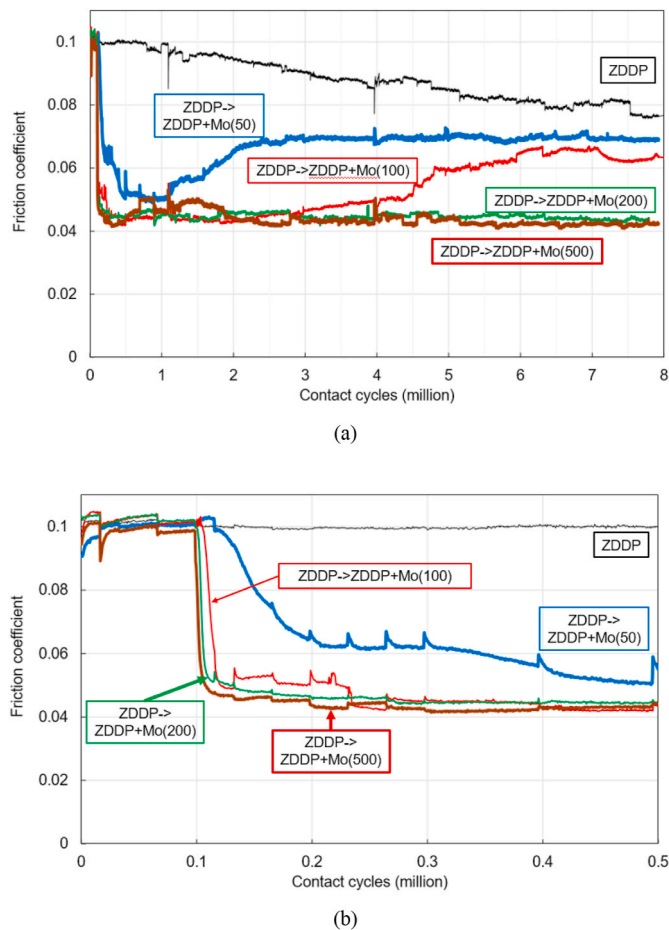


Fig. 7. Friction behaviour in tests conducted where oils with different MoDTC concentrations were added after 0.1 million cycles. (a) 0–8 million cycles and (b) the same data by zoomed in to highlight the 0–0.5 million cycles range.

the ball rubbed track along the rolling-direction after 8 million cycles. Fig. 10 shows the evolution of the micropitting damage on the balls expressed as a percentage micropitted area over total area of the ball rubbing track. This was quantified from optical micrographs of entire wear tracks.

Fig. 9 clearly shows multiple surface-breaking cracks, growing at a shallow angle of approximately 20° to 30° to the surface and against the direction of the applied friction force. Small micropits, approximately $30\ \mu\text{m}$ across and about $10\ \mu\text{m}$ deep, can be seen to be formed by detachment of surface material due to interaction of these cracks. These observations confirm that the generated damage is micropitting. As is evident in the images of Fig. 8 and the quantitative data in Fig. 10, the amount of micropitting varied significantly with MoDTC concentration. ZDDP without any MoDTC produced considerable micropitting after 4 and 8 million cycles as shown in Fig. 8. By contrast, all MoDTC-containing blends produced very little micropitting damage with only isolated cracks and pits present.

Fig. 10 shows that the percentage of micropitted area on the rubbed track decreased with increasing concentration of Mo. For the blend containing ZDDP only, the percentage of micropitted area increased slowly between 0.1 million (where very little damage was seen) and 1 million cycles (4.2% micropitted area), and then rose sharply to reach 14.7% at 4 million cycles and 27.2% after 8 million cycles. This micropitted area was markedly reduced by the addition of MoDTC after 0.1 million cycles, producing 11.5%, 6.4%, 3.8% and 1.7% micropitted area after 8 million cycles with 50, 100, 200, and 500 ppm MoDTC respectively.

Fig. 8 shows that the ZDDP + Mo(500) blend retards the formation of

tribofilm in the central region of the rubbing track in a manner similar to that observed in Fig. 4 and as discussed above.

3.2.3. Evolution of surface roughness of counterface discs

Fig. 11 shows how the disc roughness changed during the tests with different MoDTC concentrations added after 0.1 million cycles. It is evident that in all tests, the disc roughness reduces from the initial $0.45\ \mu\text{m Ra}$ to $0.3\ \mu\text{m Ra}$ during the initial 0.1 million cycles of running in with lubricant containing only ZDDP. There is then no further change in roughness once the initial ZDDP blend is replaced with the MoDTC-containing blends at Mo concentrations of 50, 100 and 200 ppm. This shows that the running-in of the counterface discs was complete when MoDTC oil was added, and that therefore the addition of MoDTC at 0.1 million cycles did not interfere with this running-in process. The addition of MoDTC at Mo concentration of 500 ppm at 0.1 million cycles did decrease disc roughness further, to $\sim 0.23\ \mu\text{m Ra}$ after 4 million cycles, after which the roughness remained relatively unchanged to the end of the test.

3.2.4. Evolution of the tribofilm on the ball

Fig. 12 shows the evolution of tribofilm thickness measured in the central region of the rubbed tracks using SLIM. As expected, tribofilms developed at the same rate (reaching around $50\ \text{nm}$ at 0.1 million cycles) in all tests during the first 0.1 million cycles conducted with a blend containing ZDDP only. After replacement with MoDTC-containing lubricants at 0.1 million cycles, the evolution of tribofilm thickness varies with MoDTC concentration. Lubricants containing ZDDP and MoDTC grew tribofilms more slowly than the ZDDP-only oil. ZDDP + Mo at 50, 100 and 200 ppm blends reached a maximum tribofilm thickness after about 0.2–0.3 million cycles with thickness values being $160\ \text{nm}$, $125\ \text{nm}$, $110\ \text{nm}$, respectively. The tribofilm thicknesses then decreased to stabilize at around $70\ \text{nm}$ after around 0.5 million cycles. These stabilised tribofilms were thicker than those from the Mo-free ZDDP solution from about 2 million cycles onwards. By contrast, as is evident in Fig. 12 (as well as Fig. 8) the addition of ZDDP + Mo at 500 ppm at 0.1 million cycles leads to rapid removal of the tribofilm formed during the initial 0.1 million cycles of rubbing in the ZDDP only oil. The tribofilm thickness then remains negligible to the end of the test.

4. Discussion

The results presented in this paper show that decreasing friction by addition of MoDTC to a lubricating oil decreases micropitting damage. Crucially, the methodology employed in the present paper allows for the effect of friction on micropitting to be isolated from other factors that may influence micropitting. In particular, the potential effect of increased running-in of the counterface that can occur due to reduction in antiwear tribofilm effectiveness when MoDTC is added to the oil blend, is eliminated by only adding the MoDTC at 0.1 million cycles, at which point the running-in process is already complete.

4.1. Other factors that may influence micropitting in the present experiments

Before concluding that friction has a direct effect on micropitting rate, two other factors that might potentially accompany a change in friction and also affect the observed micropitting trends must be discounted. These are (i) the effect of friction on specimen bulk temperature and (ii) the magnitude of asperity contact pressures due to the presence of ZDDP tribofilm on a steel substrate whose thickness might vary with concentration of MoDTC.

Higher friction will generate more frictional heating in the contact and in some systems, this heating could result in an increase in the bulk temperature of the specimens. This would reduce the viscosity of the oil in the contact inlet region and hence reduce the fluid film thickness, which would be expected to increase micropitting damage. However,

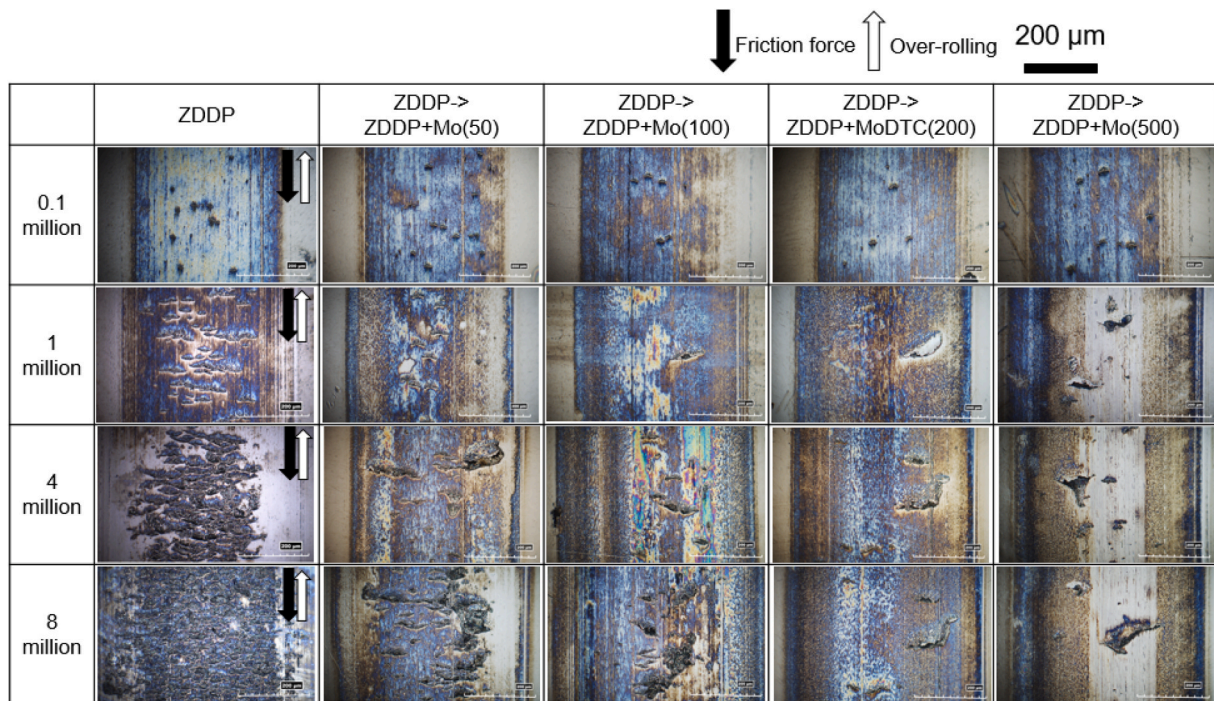


Fig. 8. Typical optical micrographs of wear tracks on balls in tests conducted where oils with different MoDTC concentrations were added after 0.1 million cycles of running-in with ZDDP solution (Note: the scale bar included in the top right corner of the figure applies to all images shown).

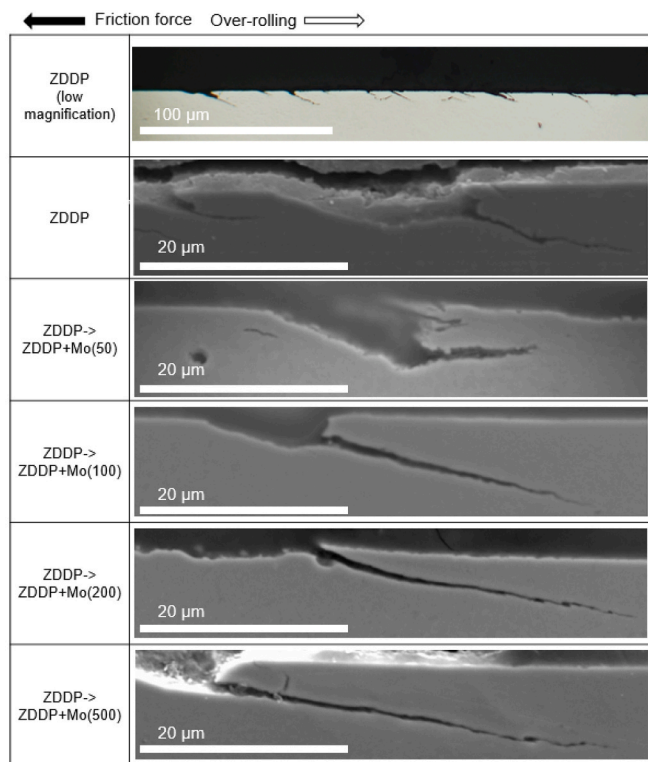


Fig. 9. Typical SEM images of polished cross-sections of the ball rubbing track along the rolling-direction in tests conducted where oils with different MoDTC concentrations were added after 0.1 million cycles. These show the appearance of typical micropits formed by crack propagation (Note: the cross-section obtained in ZDDP with low magnification shown in the upper-most image was captured using an optical microscope.).

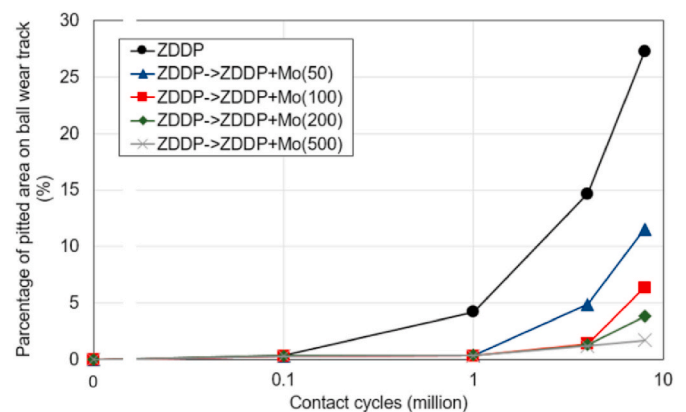


Fig. 10. Measured percentage of micropitted area on ball wear track in tests in which different MoDTC concentrations were added after 0.1 million cycles of running in with ZDDP solution.

this effect is not present in the current experiments because the specimens were immersed in an oil bath at the temperature which is actively controlled by the MTM rig to remain at 65 °C throughout all the tests. The specimen bulk temperature and film thickness are therefore the same in all tests regardless of the level of friction.

It is possible that the presence of a relatively compliant ZDDP-based tribofilm on the steel substrate might reduce the magnitude of the asperity contact pressures and hence affect micropitting. The magnitude of pressure change induced by a surface layer is determined by the elastic properties of the layer and the ratio of its thickness to the contact dimensions [30,52]. Given that tribofilm thicknesses formed by the ZDDP used here are of the order of 100 nm and the Hertz contact radius is 170 µm, the effect of this tribofilm at macro-contact level is certainly negligible. However, at the asperity contact level, where the micro-contact dimensions are of the order of a few microns, even this small tribolayer thickness might have some effect on asperity contact pressure. If these pressure changes were significant, then any differences

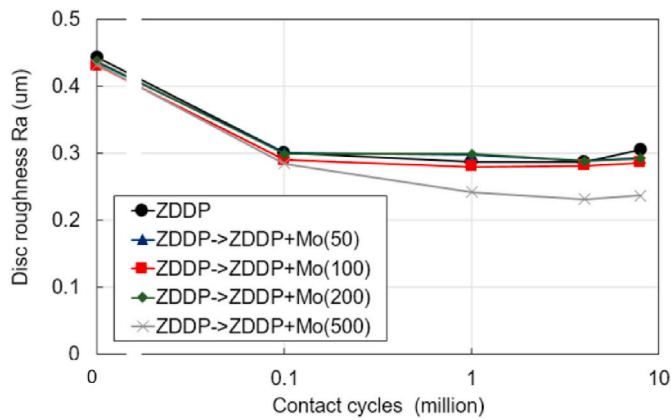


Fig. 11. Evolution of disc roughness in tests conducted where oils with different MoDTC concentrations were added after 0.1 million cycles of running-in with ZDDP solution.

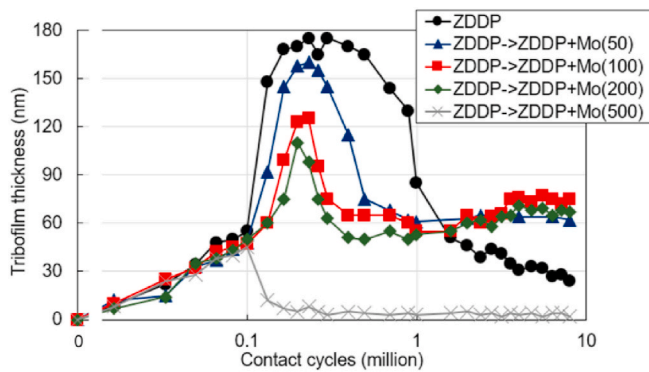


Fig. 12. Evolution of tribofilm thickness in tests conducted where oils with different MoDTC concentrations were added after 0.1 million cycles of running-in with ZDDP solution.

in tribofilm thickness with different MoDTC concentrations might potentially influence the observed trends. To investigate this, the elastic properties of the tribofilm formed in the present experiments were measured using nano-indentation, and then used in an existing numerical model for layered, rough surface contacts [38,39,52] to predict the asperity pressure distribution with and without the tribofilm. The model uses real measured surface roughness profiles and accounts for different

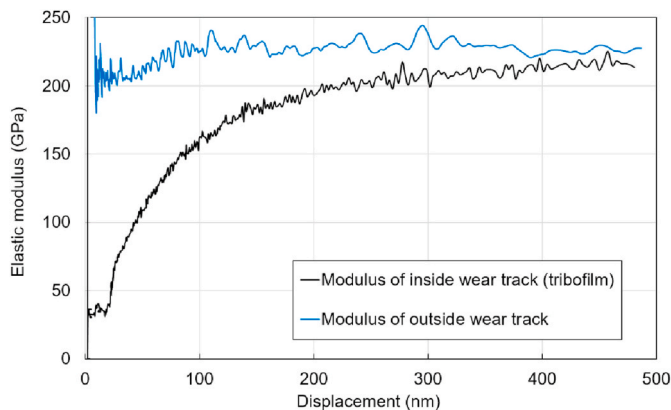


Fig. 13. Elastic modulus of ZDDP tribofilm formed on AISI 52100 steel substrate after 0.3 million cycles of rubbing in ZDDP only solution as measured by nano indentation in CSM mode. The measured ZDDP tribofilm thickness was 175 nm.

elastic properties of the layer and the substrate and the layer thickness to predict the asperity pressure distribution in non-conformal, layered, rough contacts.

The analysis presented here is for the thickest tribofilm measured in the present tests, namely 175 nm (see Figs. 6 and 12), and therefore presents the worst-case scenario in terms of the magnitude of potential changes in asperity pressures due to the presence of the tribofilm. In this analysis, ZDDP tribofilm was assumed to have a Poisson's ratio of 0.26 based on Matori et al. [42] who cite this value for zinc phosphate glasses. Fig. 13 shows representative nanoindentation results for a ZDDP tribofilm, as measured within the rubbed track, and for steel substrate, as measured outside of the wear track. In all tests, elastic modulus showed a high value at an indentation depth of less than 10 nm. This is an artefact due to imperfections in the tip geometry and does not represent the surface properties of tribofilm or steel [53,54]. For steel substrate, elastic modulus was measured to be around 220 GPa as expected. For ZDDP tribofilm the elastic modulus was in the range of 30–40 GPa up to 30 nm of tip displacement. As tip displacement increased further, elastic modulus increased steadily and at a tip displacement of about 400–500 nm it reached a similar value to that of steel. This increase of elastic modulus is caused by the well-known substrate effect [55], so that generally the true mechanical properties of nanofilms on substrates can only be determined from indentation data originating from the outermost 10% of the film thickness. Considering this, the elastic modulus of ZDDP tribofilm was determined to be an average of 37.7 GPa with 5.0% standard deviation based on 25 indentations in different positions on the specimen all within a tip displacement range of 15–25 nm. It should be noted that although this modulus was measured at room temperature while the micropitting tests were carried out at 65 °C, Pereira et al. [56] have shown that the elastic modulus of ZDDP tribofilm does not vary significantly between 25 °C and 100 °C.

Fig. 14 compares the predicted pressure distributions for a steel-on-steel contact with the contact of steel against a 175 nm thick ZDDP tribofilm (elastic modulus 37.7 GPa) on a steel substrate. Both cases use the same measured roughness profile of a fresh MTM disc as employed in the present experiments. To illustrate the maximum possible influence of the tribofilm on contact pressures, the example results shown here are from a pure elastic analysis. This does cause some discrete pressure spikes to be artificially high but in practice these pressures would quickly be reduced to the plastic limit on first contact. The results show that 175 nm thick ZDDP tribofilm can reduce the contact pressures on individual asperities by up to about 10% compared to the equivalent steel-on steel contact, with the reduction being the largest for higher asperity pressures. Given that this elastic analysis was an extreme case

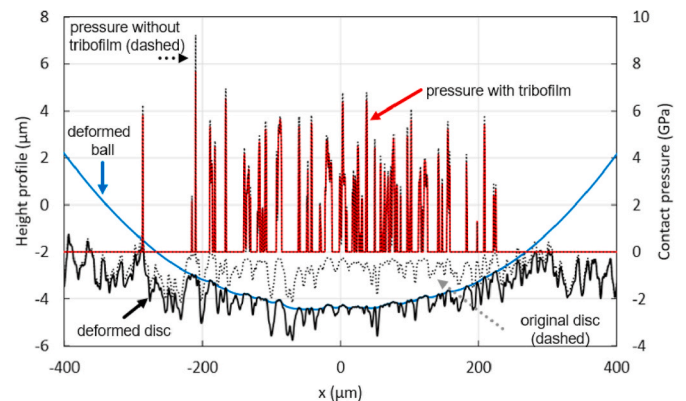


Fig. 14. Predicted contact pressure distributions for a steel-on-steel contact and an equivalent steel on 175 nm thick ZDDP tribofilm on steel substrate contact. Analysis uses measured roughness profile from a fresh MTM disc with $R_a \sim 0.43 \mu\text{m}$ and measured elastic modulus of ZDDP film ($E = 37.7 \text{ GPa}$). Note that the results shown are from a pure elastic analysis to illustrate the extreme case (this causes some pressures peaks to be unrealistically high).

and in practice (i) the tribofilm thickness of 175 nm used here is much larger than any differences in tribofilm thickness between different tests, and (ii) the virgin disc roughness used here would quickly be modified during the running-in process, it can be concluded that any differences in asperity pressures caused by small differences in tribofilm thickness are very small and inconsequential in practice. In addition, the tribofilm thicknesses from oils including MoDTC were lower than that from the oil containing only ZDDP, and this could only increase the asperity pressures. This would act to increase micropitting, but the opposite trend of lower damage with MoDTC blends was observed here. This confirms the conclusion that reducing friction reduces micropitting damage and this remains true even if there are small increases in asperity pressures due to reduced thickness of the relatively compliant ZDDP tribofilm when MoDTC is added.

4.2. Mechanisms by which friction influences micropitting

This study shows that friction has a direct effect on micropitting, with low friction reducing the tendency to micropit and vice-versa. Because MoDTC reduces asperity-asperity friction by forming MoS₂ on asperities, the primary mechanism by which reduction in friction alleviates micropitting is by reducing the mechanical stresses experienced by asperities during rubbing. For an ideally smooth contact, it is well known that frictional force moves the location of the maximum subsurface shear stress nearer to the surface, as well as generating tensile stress at the rear of the contact region [1–3,5–7]. In real, rough surface contacts such as those used here, the presence of roughness and friction will combine to generate high shear stresses at or very near the surface [16,30]. These stresses are associated with micro-asperity contacts and are ultimately responsible for the formation of surface fatigue cracks which can lead to micropitting. Using a pure elastic analysis of rough contacts as a worst case scenario, Bailey and Sayles [16] showed that the magnitude of the near surface shear stresses associated with local asperity contacts for a virgin ground surface with friction coefficient of 0.1 could be twice as high as the maximum subsurface shear stress for an equivalent ‘smooth’ contact. The magnitude of these local shear stresses due to asperity contacts may be expected to increase with increasing friction. The higher shear stresses will then result in higher accumulation of the plastic strain on the local level which is ultimately believed to be responsible for the formation of fatigue cracks [12,18].

In addition to these near surface shear stress effects, since friction is known to generate tensile stresses at the trailing edge of macro-scale contacts it is reasonable to assume that the same effect will occur at the trailing edge of asperity micro-contacts. These tensile stresses enhance crack propagation rates by increasing mode I stress intensity factors. Although, under rolling contact, mode II stress intensity factors are commonly dominant, mode I stress intensity is significant and increases the most with increasing friction. Furthermore, higher friction also increases mode I intensity due to pressurisation of any fluid that may have entered the crack [25,26] as it acts to open up the crack mouth ahead of the approaching contact thus allowing the fluid to enter it.

In the case of micropitting, multiple cracks are generated due to asperity stress fields, but, unlike in macro-pitting, these only propagate to a relatively short length, in the region of tens of microns, before creating a micropit. The reasons for this are twofold. Firstly, the closely-spaced, multiple surface cracks interact with each other, and this leads to a formation of a micropit before cracks are able to propagate to longer lengths. Secondly, for a crack to grow longer, it needs to cross the low-stress ‘quiescent’ zone that exists between the highly-stressed near-surface regions and the subsurface stress region associated with macro-level Hertzian stresses [58,59]. The size of this ‘quiescent’ zone is determined by the surface roughness properties and the macro stresses resulting from the applied loading and macro geometry so that the exact combination of these will favour micropitting or macro-pitting.

To test these arguments further, (i) the effect of friction on near surface stresses in the present experiments was investigated and (ii) the

morphology of surface cracks obtained at different friction levels was analysed.

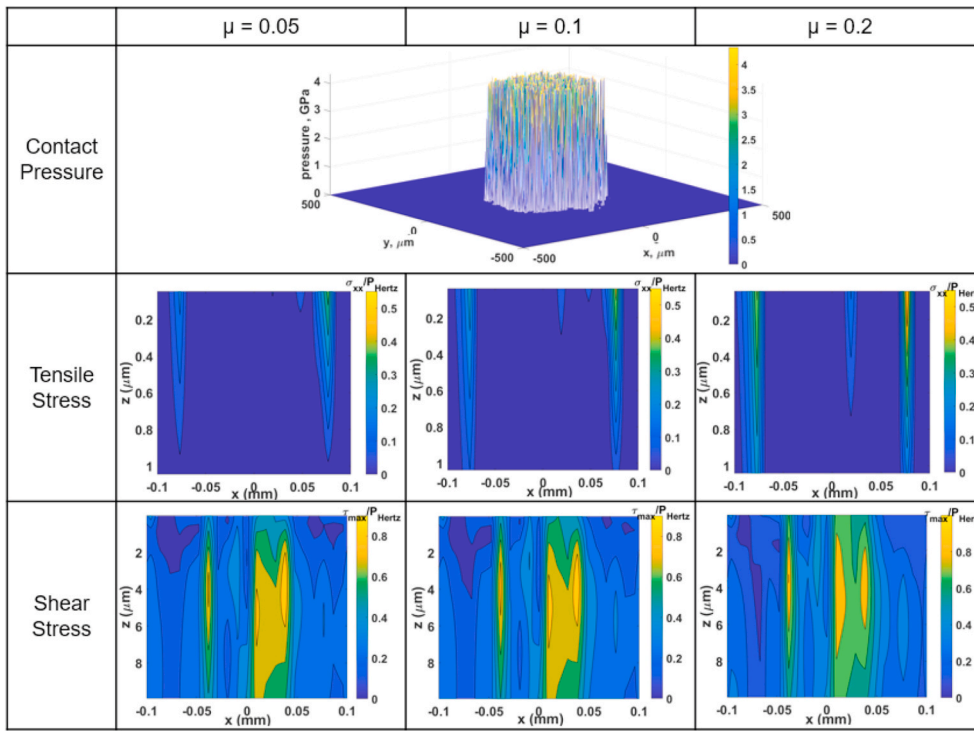
4.2.1. Influence of friction on near-surface stresses

A well-established numerical model for contact of rough surfaces [16,38,39,52] was employed to analyse the subsurface stress fields under the conditions of the present experiments. The numerical results presented in this section use an existing model and are not novel in themselves but serve to illustrate the mechanisms at play. The simulations make a number of simplifying assumptions including ignoring any effect of lubricant on contact stresses and assuming elastic-perfectly plastic material behaviour with an imposed plastic limit for asperity pressures of 4 GPa. The former is justified on the grounds of very low lambda ratios employed in the tests. The latter is a very common way to deal with elastic-plastic rough contacts although it ignores important phenomena such as work hardening and asperity persistence [60,61]. ZDDP tribofilm is not included in the model since its effects were already shown to be small. The simulation considers the conditions used in the MTM tests *i.e.* a 19.05 mm diameter steel ball pressed against a steel disc with a load of 75 N, and the real measured roughness profile of the MTM disc. The ball is treated as ideally smooth given that its Ra is less than 20 nm. Results for an equivalent smooth ball on disc contact are shown for comparison and to illustrate the validity of the model predictions. Simulations are performed at friction coefficients of 0.05 and 0.1, *i.e.* similar to those measured in the present tests with and without MoDTC. Further results are also obtained at friction coefficient of 0.2 to emphasise how contact stress varies with increasing friction.

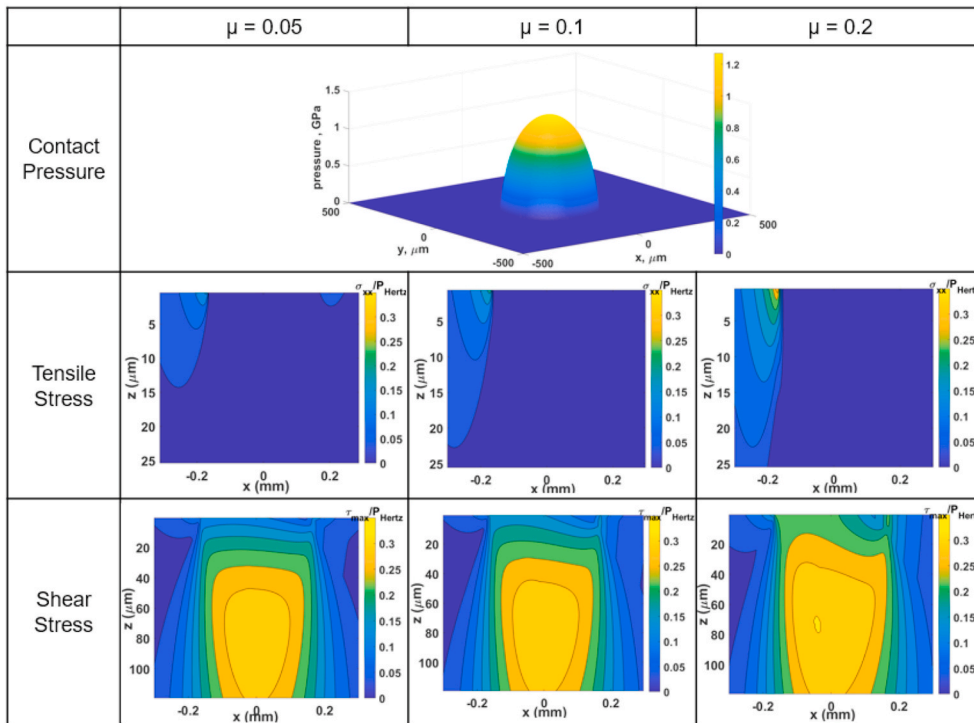
Fig. 15a shows the predicted contact pressure distribution and the contour plots of maximum shear stress and positive (tensile) σ_{xx} stress in the central plane section in rolling direction ($y = 0$). For comparison, Fig. 15b shows the equivalent results for a smooth contact. Tensile and shear stresses, P_{hertz} , are shown relative to the maximum Hertzian pressure for a smooth contact. It should be noted that only the tensile σ_{xx} stresses are shown, with compressive stresses set to zero so that tensile stress contours are clearly visible. The sliding direction (direction of friction) is from left to right in all cases. The results for the smooth surface show that friction produces tensile stress at the trailing edge of the contact at all three friction coefficients studied. For the rough contact, there are several local regions of tensile σ_{xx} stress near the surface associated with individual asperity contacts. These tensile stresses are very shallow, reaching only about 1 μm below the surface. It is evident that for both smooth and rough contacts, higher friction increases the size of the tensile σ_{xx} regions and increases their maximum magnitude. For a smooth surface, these magnitudes are 0.20 GPa, 0.28 GPa and 0.44 GPa for friction coefficients of 0.05, 0.1 and 0.2, respectively. The equivalent magnitudes for the rough disc are about twice as high at 0.50 GPa, 0.56 GPa and 0.70 GPa, respectively.

For the smooth contact, the maximum shear stress is located in the subsurface at a depth that is about half the contact radius, but moves closer to the surface as friction increases from 0.05 to 0.2, as is in line with contact mechanics theory [62]. For the rough disc case, there are several local regions of high shear stresses, all occurring very near, generally within 5 μm , of the surface. These local shear stress maxima are due to local stress fields caused by asperity contacts and the general trends shown here are in line with previous studies [16,18,19,25,30,57]. The magnitudes of the local high shear stress regions increase with friction (See Fig. 15b). In addition, the existence of multiple regions of high shear stress means that the number of fluctuations in shear stress experienced by points in the material within this near surface zone during the passage of one surface over another in rolling-sliding contact is increased.

These characteristics of the local shear stresses and direct tensile stresses associated with higher friction will lead to increased accumulation of plastic strains with increasing friction, and hence increased crack initiation in the affected near surface regions. The increase in magnitude of tensile and shear stresses with increasing friction also



(a)



(b)

Fig. 15. Surface pressure distribution, contours of positive (tensile) σ_{xx} stress (compressive σ_{xx} stresses set to zero for clarity) and contours of maximum shear stress in the central plane ($y = 0$) in the rolling direction in the contact of an MTM ball and disc under normal load of 75 N and friction as stated: (a) smooth MTM ball on the rough disc (disc roughness as used in present tests and measured using a stylus profilometer); (b) smooth ball on smooth disc under same loading conditions. All stresses have been normalised by the maximum Hertz pressure. Note that the top of the figures of stress distribution ($z = 0$) are the contact surfaces.

means that such cracks are more likely to propagate. This in turn will lead to more micropitting damage. Therefore, the primary mechanism by which higher friction increases micropitting is via its effect on near-surface stresses at the asperity conjunctions that occur in rough surface lubricated contact.

4.3. Effect of friction on observed crack morphology

To investigate the effect of friction on surface crack morphology, all cracks in each prepared ball cross-section in the rolling direction after 8 million cycles were analysed to measure (i) crack length in the subsurface and (ii) the angle at which the cracks grow in relation to the surface. (Typical cross-section were illustrated in Fig. 9). The analysed cross-sections were all from tests run using Method 2 (*i.e.* MoDTC added

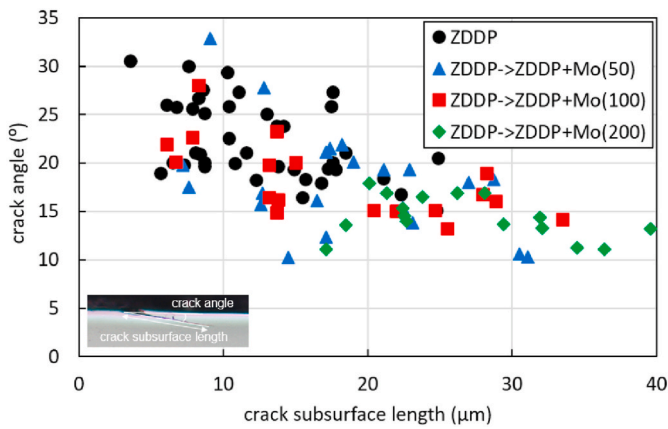


Fig. 16. Relationship between crack subsurface length and crack angle relative to the surface as measured for all cracks observed in the prepared cross sections of the balls in the rolling direction at 8 million cycles for the lubricant blends containing different amounts of MoDTC as stated. Parts of the relevant cross sections are shown in Fig. 9.

after the initial running-in was complete) at different concentrations of the MoDTC, and hence different friction. Fig. 16 shows the relationship between the measured crack subsurface length and the crack angle. The results of ZDDP + Mo(500) were excluded because of its lower counterface roughness after 1 million cycles compared to other blends, which in itself obscures the influence of friction on micropitting as explained above. It should be noted that the number of cracks observed increased with increasing friction coefficient, so that the tests with ZDDP alone (black circles) generated the most cracks. Two features are immediately apparent in Fig. 16, (i) higher friction resulted in shorter cracks on average, and (ii) at higher friction cracks appear to grow at a steeper angle to the surface although this may be affected by the crack length itself (as evident on the graph) rather than only friction. For example, in the test with ZDDP-only, (friction coefficient of around 0.1), cracks were between 5 μm and 25 μm long and their angle to the surface was between 15° and 30°. By contrast, for tests where 200 ppm Mo was added after 0.1 million cycles (friction coefficient around 0.04), the crack lengths were between 15 μm and 40 μm long and the crack angle to the surface was between 10° and 20°.

The exact reason for the apparent change of crack angle with friction is difficult to establish with certainty as both friction and crack length are changing as one moves from left to right along the x axis in Fig. 16. However, this must be related to the changes in the stress fields experienced by the crack for different friction coefficients and observed crack lengths (longer at lower friction). Further understanding and determination of the change in crack angle with friction is likely to be the subject of future investigations.

The trend for shorter cracks at higher friction may seem counterintuitive given that high friction gives higher shear and tensile stresses as shown above. However, there are plausible reasons for these observations. Firstly, the fact that the number of cracks is higher for higher friction coefficients is in accord with the higher magnitude of the near surface stresses discussed above. However, if there is a higher density of cracks, these are more likely to interact to form a micropit, thus truncating any further growth of the affected cracks. Consequently, any remaining cracks that may be observed in the cross-sections from high friction tests are likely to be shorter than in the case of low friction, which must grow longer before interacting.

The entrainment of oil into the surface-breaking crack may also play a part in determining their relevant lengths at high and low friction. Several authors [25–27,63] have shown numerically that once entrained into the crack, the lubricant can be pressurized during over-rolling, which increases mode I stress intensity factor and hence promotes crack propagation. Olver et al [64], have provided remarkable direct

observations of such lubricant interaction with a micropit passing across the contact. Another consequence of lubricant entering a crack is the reduction in crack face friction. This is a less-studied effect, but it has been shown to be important in relation to crack propagation under rolling contact [7,26,65]. If high friction exists between crack faces during the passage of the crack through the contact, the crack faces will lock and be less able to slide past each other. This will reduce the mode II stress intensity factor, and hence reduce crack propagation rate [26,65]. Bower [26] predicts that mode II crack propagation is unlikely if crack face friction exceeds 0.2. If lubricant does enter the crack, one with low friction properties such as the MoDTC blends used in the present tests, will decrease crack face friction more than one that gives higher friction (such as that containing only ZDDP in the present tests). Consequently, the propagation of surface breaking cracks under mode II may well be enhanced by the presence of low friction oil. This may be another reason why the cracks observed here in the tests with MoDTC blends were longer than those observed with the oil containing ZDDP only. This does not imply that more micropitting would be caused by a lower friction oil, indeed the opposite is true as shown here, because fewer cracks are initiated with the low friction oil than the high friction oil in the first place, so that crack interaction that precedes micropit formation is less likely to occur.

5. Conclusions

This study used MTM-SLIM ball-on-disc tests with custom oil blends containing ZDDP and different concentrations of MoDTC friction modifier to investigate the effect of friction on micropitting. In order to generate micropitting on the balls, non-standard MTM discs with roughness of $0.42 \pm 0.02 \mu\text{m Ra}$ and softer balls were used. The main findings can be summarised as follows:

- It was shown that in micropitting tests in which the initial 0.1 million cycles were conducted with an oil containing ZDDP alone, after which the oil was replaced by one containing ZDDP and MoDTC until the end of the test, the same amount of roughness reduction of counterface discs due to running-in was observed regardless of whether or not the MoDTC was present in the second stage of the test, i.e. regardless of the level of friction in the second stage. This approach makes it possible to isolate the effect of friction on micropitting from that of running-in which would otherwise be affected by addition of MoDTC.
- Reduction of friction coefficient from around 0.10 (ZDDP alone) to around 0.04 (ZDDP + MoDTC at Mo concentration of 200 ppm) resulted in an order of magnitude reduction in the extent of micropitted area within the ball wear track. This shows that friction has a very significant impact on micropitting.
- Lower friction resulted in fewer surface cracks that were longer and grew at a shallower angle to the surface compared to those produced at higher friction.
- Analysis of the contact stresses in the MTM ball-on-disc contact at different friction coefficients suggests that the primary mechanism by which friction affects micropitting is through its effects on the near-surface shear stresses and direct tensile stresses associated with asperity contacts.

Credit author statement

Mao Ueda: Conceptualization, Methodology, Software, Validation, Formal analysis, Investigation, Resources, Data Curation, Writing - Original Draft, Visualization, Project administration.

Benjamin Wainwright: Software, Formal analysis, Investigation.

Hugh Spikes and Amir Kadiric: Supervision, Writing - Review & Editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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References

- [1] G.E. Morales-Espejel, P. Rycerz, A. Kadiric, Prediction of micropitting damage in gear teeth contacts considering the concurrent effects of surface fatigue and mild wear, *Wear* 398–399 (2018) 99–115.
- [2] M.N. Webster, C.J.J. Norbart, An experimental investigation of micropitting using a roller disk machine, *Tribol. Trans.* 38 (1995) 883–893.
- [3] J.J. Wagner, S. Sundararajan, Failure mode mapping for rolling/sliding contacts under low lambda conditions, *Wear* (2021) 203855.
- [4] P. Rycerz, A. Kadiric, The influence of slide-roll ratio on the extent of micropitting damage in rolling-sliding contacts pertinent to gear applications, *Tribol. Lett.* 67 (2019) 63.
- [5] G.E. Morales-Espejel, V. Brizmer, Micropitting modelling in rolling-sliding contacts: application to rolling bearings, *Tribol. Trans.* 54 (2011) 625–643.
- [6] E. Lainé, A.V. Olver, M.F. Lekstrom, B.A. Shollock, T.A. Beveridge, D.Y. Hua, The effect of a friction modifier additive on micropitting, *Tribol. Trans.* 52 (2009) 526–533.
- [7] M. Ueda, H. Spikes, A. Kadiric, In-situ observations of the effect of the ZDDP tribofilm growth on micropitting, *Tribol. Int.* 138 (2019) 342–352.
- [8] A. Vrček, T. Hultqvist, Y. Baubet, P. Marklund, R. Larsson, Micro-pitting damage of bearing steel surfaces under mixed lubrication conditions: effects of roughness, hardness and ZDDP additive, *Tribol. Int.* 138 (2019) 239–249.
- [9] C. Benyajati, A.V. Olver, C.J. Hamer, An experimental study of micropitting, using a new miniature test-rig, *Tribol.* 43 (2003) 601–610.
- [10] A.V. Olver, *Wear of Hard Steel in Lubricated, Rolling Contact*, PhD Thesis, Imp. Coll. London, 1986.
- [11] M. Ueda, H. Spikes, A. Kadiric, Influence of black oxide coating on micropitting and ZDDP tribofilm formation, *Tribol. Trans.* (2021) 1–21.
- [12] A.V. Olver, The mechanism of rolling contact fatigue: an update, *Proc. Inst. Mech. Eng. Part J J. Eng. Tribol.* 219 (2005) 313–330.
- [13] M. Guagliano, L. Vergani, M. Vimerati, Sub-surface crack propagation analysis in hypoid gears, *Eng. Fract. Mech.* 75 (2008) 417–426.
- [14] Y. Enomoto, D. Tabor, The frictional anisotropy of diamond, *Proc. R. Soc. London. A. Math. Phys. Sci.* 373 (1981) 405–417.
- [15] G.M. Hamilton, L.E. Goodman, The stress field created by a circular sliding contact, *J. Appl. Mech. Trans. ASME* 33 (1964) 371–376.
- [16] D.M. Bailey, R.S. Sayles, Effect of roughness and sliding friction on contact stresses, *J. Tribol.* 113 (1991) 729–738.
- [17] D.F. Diao, K. Kato, K. Hayashi, The maximum tensile stress on a hard coating under sliding friction, *Tribol. Int.* 27 (1994) 267–272.
- [18] A.P. Voskamp, Material response to rolling contact loading, *J. Tribol.* 107 (1984) 359–364.
- [19] A.V. Olver, Micropitting and asperity deformation, *Dev. Numer. Exp. Methods Appl. to Tribol.* (1984) 319–323.
- [20] I.I. Kudish, K.W. Burris, Modern state of experimentation and modeling in contact fatigue phenomenon: Part II—analysis of the existing statistical mathematical models of bearing and gear fatigue life. New statistical model of contact fatigue, *Tribol. Trans.* 43 (2000) 293–301.
- [21] A. Otsuka, H. Sugawara, M. Shomura, A test method for mode II fatigue crack growth relating to a model for rolling contact fatigue, *Fatig. Fract. Eng. Mater. Struct.* 19 (1996) 1265–1275.
- [22] T.E. Tallian, Simplified contact fatigue life prediction model-part I: review of published models, *J. Tribol.* 114 (1992) 207–213.
- [23] P. Rycerz, A. Olver, A. Kadiric, Propagation of surface initiated rolling contact fatigue cracks in bearing steel, *Int. J. Fatig.* 97 (2017).
- [24] E. Ioannides, T.A. Harris, A new fatigue life model for rolling bearings, *J. Tribol.* 107 (1985) 367–377.
- [25] M. Kaneta, M. Suetsugu, Y. Murakami, Mechanism of surface crack growth in lubricated rolling/sliding spherical contact, *J. Appl. Mech. Trans. ASME* 53 (1986) 354–360.
- [26] A.F. Bower, The influence of crack face friction and trapped fluid on surface initiated rolling contact fatigue cracks, *J. Tribol.* 110 (1988) 704.
- [27] R. Balcombe, M.T. Fowell, A.V. Olver, S. Ioannides, D. Dini, A coupled approach for rolling contact fatigue cracks in the hydrodynamic lubrication regime: the importance of fluid/solid interactions, *Wear* 271 (2011) 720–733.
- [28] T.H. Kim, A.V. Olver, Stress history in rolling-sliding contact of rough surfaces, *Tribol. Int.* 31 (1998) 727–736.
- [29] G.E. Morales-espejel, P. Rycerz, A. Kadiric, Prediction of micropitting damage in gear teeth contacts considering the concurrent effects of surface fatigue and mild wear, *Wear* 398 (2018) 99–115.
- [30] A. Kadiric, R.S. Sayles, X.B. Zhou, E. Ioannides, A numerical study of the contact mechanics and sub-surface stress effects experienced over a range of machined surface coatings in rough surface, *J. Tribol.* 125 (2003) 720–730.
- [31] P. Rycerz, A. Kadiric, Propagation of surface initiated rolling contact fatigue cracks in bearing steels, 73rd STLE Annu. Meet. Exhib. (2018) 40.
- [32] S. Soltanahmadi, A. Morina, M.C.P. Van Eijk, I. Nedelcu, A. Neville, Investigation of the effect of a diamine-based friction modifier on micropitting and the properties of tribofilms in rolling-sliding contacts, *J. Phys. D Appl. Phys.* 49 (2016) 505302.
- [33] K.T. Miklozic, T.R. Forbus, H.A. Spikes, Performance of friction modifiers on zddp-generated surfaces, *Tribol. Trans.* 50 (2007) 328–335.
- [34] V. Brizmer, C. Matta, I. Nedelcu, G.E. Morales-Espejel, The influence of tribolayer formation on tribological performance of rolling/sliding contacts, *Tribol. Lett.* 65 (2017) 1–18.
- [35] K. Topolovec-Miklozic, T.R. Forbus, H.A. Spikes, Film thickness and roughness of ZDDP antiwear films, *Tribol. Lett.* 26 (2007) 161–171.
- [36] M.N. Webster, R.S. Sayles, A numerical model for the elastic frictionless contact of real rough surfaces, *J. Tribol.* 114 (1986) 334–340.
- [37] M.A. West, R.S. Sayles, Paper VIII (i) A 3-dimensional method of studying 3-body contact geometry and stress on real rough surfaces, *Tribol.* 12 (1987) 195–200.
- [38] A. Kadiric, R.S. Sayles, E. Ioannides, Thermo-mechanical model for moving layered rough surface contacts, *J. Tribol.* 130 (2008).
- [39] J. Nyqvist, A. Kadiric, S. Ioannides, R. Sayles, Semi-analytical model for rough multilayered contacts, *Tribol. Int.* 87 (2015) 98–112.
- [40] M. Aktary, M.T. McDermott, G.A. McAlpine, Morphology and nanomechanical properties of ZDDP antiwear films as a function of tribological contact time, *Tribol. Lett.* 12 (2002) 155–162.
- [41] X. Li, B. Bhushan, A review of nanoindentation continuous stiffness measurement technique and its applications, *Mater. Char.* 48 (2002) 11–36.
- [42] K.A. Matori, M.I. Sayyed, H.A.A. Sidek, M.H.M. Zaid, V.P. Singh, Comprehensive study on physical, elastic and shielding properties of lead zinc phosphate glasses, *J. Non-Cryst. Solids* 457 (2017) 97–103.
- [43] I.M. Feng, W.L. Perilstein, M.R. Adams, Solid film deposition and non-sacrificial boundary lubrication, *ASLE Trans* 6 (1963) 60–66.
- [44] C. Grossiord, K. Varlot, J. Martin, T. Le Mogne, C. Esnouf, MoS₂ single sheet lubrication by molybdenum, *Tribol. Int.* 31 (1999) 737–743.
- [45] M. Ueda, A. Kadiric, H. Spikes, Wear of hydrogenated DLC in MoDTC-containing oils, *Wear* 474–475 (2021) 203869.
- [46] J. Graham, H. Spikes, R. Jensen, The friction reducing properties of molybdenum dialkylthiocarbamate additives: part ii - durability of friction reducing capability, *Tribol. Trans.* 44 (2001) 637–647.
- [47] A. Morina, A. Neville, Understanding the composition and low friction tribofilm formation/removal in boundary lubrication, *Tribol. Bar Int.* 40 (2007) 1696–1704.
- [48] J. Zhang, H. Spikes, On the mechanism of ZDDP antiwear film formation, *Tribol. Lett.* 63 (2016) 1–15.
- [49] J. Zhang, J.P. Ewen, M. Ueda, J.S. Wong, H.A. Spikes, Mechanochemistry of zinc dialkylthiophosphate on steel surfaces under elastohydrodynamic lubrication conditions, *ACS Appl. Mater. Interfaces* 12 (2020) 6662–6676.
- [50] K. Hoshino, Film Forming and Friction Properties of Sulphur-Free Antiwear Additives Zinc Dialkylphosphates (ZDPs), PhD Thesis, Imp. Coll. London, 2011.
- [51] V.J. Gatto, Y.L. Bezjak, The antioxidant properties of organomolybdenum compounds in engine oils, *Tribol. Lubric. Technol.* 62 (2006) 32–39.
- [52] S.J. Cole, R.S. Sayles, A numerical model for the contact of layered elastic bodies with real rough surfaces, *J. Tribol.* 114 (1992) 334–340.
- [53] G.Z. Voyiadjis, R. Peters, Size effects in nanoindentation: an experimental, *Acta Mech.* 211 (2010) 131–153.
- [54] W.D. Nix, H. Gao, Indentation size effects in crystalline materials: a law for strain gradient plasticity, *J. Mech. Phys. Solid.* 46 (1998) 411–425.
- [55] R. Saha, W.D. Nix, Effects of the substrate on the determination of thin film mechanical properties by nanoindentation, *Acta Mater.* 50 (2002) 23–38.
- [56] G. Pereira, D. Munoz-Paniagua, A. Lachenwitzer, M. Kasrai, P.R. Norton, T. W. Capehart, T.A. Perry, Y.T. Cheng, A variable temperature mechanical analysis of ZDDP-derived antiwear films formed on 52100 steel, *Wear* 262 (2007) 461–470.
- [57] A.F. Bower, N.A. Fleck, Brittle fracture under a sliding line contact, *J. Mech. Phys. Solid.* 42 (1994) 1375–1396.
- [58] T.E. Tallian, Y.P. Chiu, E. Van Amerongen, Prediction of traction and microgeometry effects on rolling contact fatigue life, *J. Tribol.* 100 (1978) 156–165.
- [59] G.R. Miller, L.M. Keer, H.S. Cheng, The role of near-surface inclusions in the pitting of gears, *ASLE Trans* 28 (1985) 111–116.
- [60] I. Lee-Prudhoe, R.S. Sayles, A. Kadiric, Investigations into asperity persistence in heavily loaded contacts, *J. Tribol.* 121 (1999) 441–448.
- [61] T.H.C. Childs, Persistence of roughness between surfaces in static contact, *Proc R Soc London Ser A* 353 (1977) 35–53.
- [62] K.L. Johnson, *Contact Mechanics*, 1987.
- [63] M. Kaneta, Y. Murakami, Effects of oil hydraulic pressure on surface crack growth in rolling/sliding contact, *Tribol. Int.* 20 (1987) 201–217.
- [64] A.V. Olver, L.K. Tiew, S. Medina, J.W. Choo, Direct observations of a micropit in an elastohydrodynamic contact, *Wear* 256 (2004) 168–175.
- [65] W.J. Wang, R. Lewis, M.D. Evans, Q.Y. Liu, Influence of different application of lubricants on wear and pre-existing rolling contact fatigue cracks of rail materials, *Tribol. Lett.* 65 (2017) 1–15.