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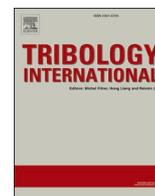
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Dynamics of particle entrainment for glass particles suspended in various fluids

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ABSTRACT

The frictional properties of two sliding surfaces are often influenced by a fluid lubricant's ability to separate and facilitate movement. When particles are present, their entrainment between surfaces can alter friction, either increasing or decreasing it compared to fluid lubrication alone. Understanding the frictional regimes and the dynamics driving particle entrainment in suspensions is thus critical. This study investigates the tribological properties of glass particles suspended in various fluid matrices. We examined suspensions with varying particle concentrations, fluid viscosities, fluid hydrophobicity, and particle hydrophobicity. Our findings reveal that particle lubrication dominates under the following conditions: (I) high particle concentrations, (II) high fluid viscosities, (III) strong particle – surface interactions, and (IV) weak fluid – particle interactions. These insights are crucial for applications in food science, biomedical industries, and pharmaceuticals, where controlling particle friction is essential for optimizing consumer satisfaction and ensuring the performance and safety of lubricants in medical devices.

1. Introduction

The friction and lubrication properties of particle suspensions are of interest because of their applications in industries such as coatings [1,2], pharmaceuticals [3,4], and foods [5,6]. These frictional properties strongly influence how the end-user perceives the product. A skin cream, for example, requires specific lubrication properties to be applied to the body and remain on the skin while forming a thin film without causing abrasions to the skin [7,8]. Similarly, a cream-based dessert is expected to have specific in-mouth lubrication properties, which contribute to the liking of the product without causing excessive grittiness [9,10]. The frictional behavior of Newtonian fluids are commonly described by the Stribeck curve (boundary, mixed and EHL regime) [11–13]. For lubricants that consist of multiple components, such as particles dispersed in a fluid the tribological properties are known to deviate from the regular Stribeck behavior [14–16]. Currently, the tribological properties of such multi-component systems in combination with soft surfaces are still poorly understood. One of the reasons is that particles and fluids in the contact zone are complex to image or visualise which makes it

challenging to uncover the exact mechanism behind particle entrainment. Additionally, the deformability of the surfaces further complicates the manner in which the fluid and particles are entrained. There is thus a lack of fundamental mechanistic understanding of the regimes beyond the Stribeck regimes that apply to particulate lubricants. Understanding the main drivers behind particle entrainment facilitates the design of particulate lubricants with specific frictional properties based on the particle properties and the properties of the fluid matrix.

The lubrication behavior of particles suspensions is affected by many parameters such as the size of the particles, the hardness of the particles, the roughness of the surfaces, etc, which provide different contact areas and interlocking events [15,17–19]. Additionally, the fluid matrix surrounding the particles can also be expected to play an important role as this may facilitate particle entrainment or compete with the particles for entrainment between the surfaces. In recent literature (model) particulate systems such as emulsions and suspensions have been investigated tribologically and many of them displayed behavior that deviates from the traditional Stribeck curve which commonly describes the tribological behavior of Newtonian fluids. Particle-based frictional systems may

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include hydrogel particles [15,16], model dairy products [14,20,21] Al/SiCp composites [22] and nanocellulose dispersions [12]. While the aforementioned systems differ strongly from one another, all of the systems include solid particles present in a liquid phase. The frictional properties of gelatin hydrogel particles and glass particles were also investigated and it was demonstrated that the deformation of particles [15] and surfaces [16] are important drivers for particle entrainment. The effect of hydrophobicity on particle entrainment was also studied and it was found that whey-protein microgel particles were well able to enter the contact of hydrophilic surfaces at low particle volume fractions while particle entrainment appeared to be limited in the case of hydrophobic surfaces [23,24].

Various particle systems have thus been studied to create an understanding of the interactions leading to surface or particle deformation and subsequent particle entrainment. However, knowledge remains limited when it comes to the effects of fluid viscosity, matrix polarity, and particle hydrophobicity on particle entrainment and the overall friction coefficient. The main drivers leading to particle entrainment in the presence of a fluid remain unknown. The aim of this study was therefore to investigate the effect of interactions between particles, surfaces and the fluid matrix on particle entrainment and particle lubrication. We hypothesize that particle entrainment is facilitated by an enhanced gap size which may be caused by particle concentrations, fluid viscosity and sliding speed as well as the relative hydrophobicity of the interacting fluids, particles and surfaces.

Here, we systematically vary the polarity of the fluids (water, glycerol, oil) and the hydrophobicity of the particles (hydrophilic and hydrophobic), which has not been done previously. As such, this study uniquely focuses on how the interactions between fluid, surface, and particles dictate particle entrainment under different combinations of hydrophobic media and particles. This distinct approach allows us to provide new insights into the frictional behavior of suspensions, particularly in terms of the role of hydrophobicity.

To investigate the effect of various parameters on particle entrainment we use glass particles (diameter 100 μm) suspended in Newtonian fluids varying in concentration, viscosity and hydrophobicity. The advantage of using glass particle as a model system is that the particles are spherical, consistent and are not expected to deform under the forces used here. Additionally, the hydrophobicity of the particles can be altered using a coating making glass particles an ideal choice as a model system to understand the effect of particle – fluid – surface interactions during entrainment. This work thus provides a mechanistic understanding of the entrainment of glass particles suspended in various fluids between soft hydrophilic surfaces leading to a broader understanding of the drivers behind particle entrainment in various real-life scenarios. These findings have implications for the design and engineering of soft surfaces and suspensions commonly used in pharmaceuticals, cosmetics, foods as well as biomedical implants and devices. More specifically, cases where (semi-)solid particles in suspension lubricate soft surfaces. This may include medical implants, nutritional beverages or protective skin creams.

2. Experimental section

2.1. PDMS preparation

Hydrophobic polydimethylsiloxane (PDMS) surfaces are used to measure the friction coefficients. These relatively soft surfaces (≈ 3 MPa) surfaces are made using Sylgard 184 in a 10:1 ratio following a commonly used protocol [25]. The fluid PDMS is poured into the respective molds and cured for 2 h at 60 $^{\circ}\text{C}$ to obtain spherical probes as well as substrates used for rheology and tribology.

2.2. Particle suspensions

The particle suspensions used here were made of glass particles (solid

spherical glass particles, $< 106 \mu\text{m}$, Sigma-Aldrich, used as provided unless stated otherwise) suspended in Newtonian fluids. We used 10 % or 50 % particles w/w, combined with the matrix fluid, and the particles were dispersed into the fluid using a spatula and continuous stirring of the suspension. When sedimentation occurred during storage, the suspension was mixed again prior to tribological measurements, to homogeneity of the lubricant. The fluid matrices used consisted of ultra pure water (Milli-Q), glycerol (ChemSupply), mineral oil (light, Sigma-Aldrich).

2.2.1. Washing particles with Decon 90

Decon 90 is a commonly used cleaning agent which is described as a “A mixture of anionic and non-ionic surface active agents, stabilising agents, non-phosphate detergent builders, alkalis and sequestering agents, in an aqueous base” (<http://www.decon.co.uk/>). Here, Decon 90 is used to render the glass particles more hydrophilic by the removal of potential contamination. A protocol similar to that presented by Khoshnaw and co-workers is used [26]. Glass particles are fully submerged in a 10 % Decon 90/water solution and placed on a mechanical shaker over night. Then, the Decon 90 solution is removed using a funnel and filtration paper and washed thoroughly using water. The washing step is repeated until no visible foam/bubbles are being formed on the air-water interface (5 - 10 times). The glass particles are then left to dry by air until particles are fully dry and no longer adhere to one another.

2.3. Rheological measurements

A Haake MARS III stress-controlled rheometer was used to measure the viscosity of Newtonian fluids and suspensions. This rheometer was equipped with a parallel plate geometry. The parallel plates with a diameter of 35 mm were made of steel and the bottom plate was equipped with a sandpaper disk with a diameter of 35 mm, and the upper plate was equipped with a PDMS disk with a diameter of 35 mm. Measurements were performed with a 1 mm gap size and a temperature of 20 $^{\circ}\text{C}$. The viscosity of the suspensions was measured between the smooth PDMS plate and the rough sandpaper plate to simulate the wetting properties of the tribometer while using a rough surface to keep poorly wetting fluids and particles from being expelled from the gap. The measured viscosities for the Newtonian fluids were similar to those obtained using smooth, solid surfaces.

2.4. Tribological measurements

The tribometer used here is the PCS Instruments Mini Traction Machine (MTM). This setup consists of a rotating PDMS ball as the probe (diameter: 19 mm) and PDMS disk as the substrate (diameter: 46 mm). The PDMS surfaces were made using DOW SylgardTM 184 in a 1:10 (curing agent:base) ratio and cured in an oven at 60 $^{\circ}\text{C}$. A pot filler insertion (a metal cylinder that ensures fluid volumes smaller than 10 ml can be used while remaining between the surfaces) avoid lubricant flowing away from the sliding surfaces. The tribometer was operated at speeds between 1 and 1000 mm/s and a load of 1 N was applied. We fix the slide-to-roll ratio (SRR) at 0.5 and which can be calculated as $SRR = U_{ball} - U_{disk}/U_{total}$. The measurements were performed at 20 $^{\circ}\text{C}$ in bidirectional mode in which the probe and the substrate rotate in opposite directions. This provides a sliding friction coefficient and a rolling friction coefficient [27,28]. Here, we only report the sliding friction coefficient, similar to previous work on glass particle suspensions using the MTM [29]. In the current study, the rolling friction coefficients were relatively low (0.001 - 0.01), noisy and the results showed limited reproducibility, likely due to the particulate nature of the lubricants. During measurements we obtain an average friction coefficient from five data points per sliding velocity. Our measurements were performed in triplicate. Measurements were performed at decreasing speed from 1000 to 1 mm/s followed by increasing speed from 1 to 1000 mm/s for fluid lubricants. For the particle suspensions, however, a limited speed range

from 100 to 1 mm/s was used as particles were excluded from the gap at higher velocities, leading to data that was variable and not reproducible. An overview of all the samples measured are listed in Table 1.

3. Results

The frictional behavior of glass particles between two PDMS surfaces are assessed in this work. The most important interactions in the case of two surfaces lubricated by particles in a fluid are displayed schematically in Fig. 1. In this figure, we show the various types of contact and accompanying interactions that may occur and that can directly influence the friction coefficient which we relate to particle (P), surface (S) and fluid (F) interactions: particle – particle (P – P) interactions, surface – particle (S – P) interactions, surface – surface (S – S) interactions, fluid – surface (F – S) interactions and fluid – particle (F – P) interactions. P – P and S – P interactions (e.g. deformation, adhesion which may affect lubrication) are primarily influenced by the number of particles present, as the size and shape of the particles are kept constant.

3.1. Dry glass particles

Dry glass particles were used as a reference for particle lubrication in the absence of fluids (Fig. 2). The case where the complete substrate surface is (visibly) covered with a single layer of dry particles is referred to as 100 % dry surface coverage. The frictional values measured here using the MTM tribometer are very similar to our findings presented previously, when measuring dry glass particles lubricating PDMS surfaces measured on a Bruker TriboLab tribometer [16]. Dry glass particles show a rather constant friction coefficient of around 0.15 over the entire velocity range (Fig. 2). This constant frictional value indicates that no changes occur in terms of particle entrainment or surface deformation. The particles are thus able to become entrained between the surfaces by deforming the PDMS surface, even at low velocities. Once the particles

Table 1

An overview of the lubricants used in this study. The fluid matrix and particle concentrations as well as the parameters investigated are listed here.

Sample	Matrix type	Glass particle percentage	Purpose
Dry particles	Dry	100 %	Control
P50 in Water	Water	50 %	Effect of particle concentration
P10 in Water	Water	10 %	Effect of particle concentration
Water	Water	0 %	Effect of particle concentration
G25	Glycerol/water (25:75)	50 %	Effect of matrix viscosity
G50	Glycerol/water (50:50)	50 %	Effect of matrix viscosity
G75	Glycerol/water (75:25)	50 %	Effect of matrix viscosity
G95	Glycerol/water (95:5)	50 %	Effect of matrix viscosity
Oil	Oil	100 %	Effect of particle concentration
P50 in Oil	Oil	50 %	Effect of particle concentration
P10 in Oil	Oil	10 %	Effect of particle concentration
P50 in Oil (hydrophilic)	Oil	50 % (hydrophilic)	Effect of hydrophobicity
P10 in Oil (hydrophilic)	Oil	10 % (hydrophilic)	Effect of hydrophobicity
P50 in Water (hydrophilic)	Water	50 % (hydrophilic)	Effect of hydrophobicity
P10 in Water (hydrophilic)	Water	10 % (hydrophilic)	Effect of hydrophobicity
D90	Decon 90	50 % (hydrophilic)	Effect of hydrophobicity

are entrained, the friction coefficient remains relatively low as particles avoid direct S – S contact between the PDMS surfaces and have the ability to roll between surfaces [16].

3.2. The effect of particle concentration on particle entrainment

The fraction of particles present in a fluid matrix can be expected to affect the degree of particle entrainment and the ability of particles to separate the surfaces and reducing the friction coefficient. Pure water as well as a suspension that consists of 90 % water and 10 % glass particles were measured (Fig. 2). The 10 % particle suspensions show different frictional behavior than observed for the dry particles. For 10 % particles, we first observe a plateau (Fig. 2, grey symbols) around a friction coefficient of $\mu \approx 0.8$. This is lower than the friction coefficient of $\mu \approx 1.5$ for water in the boundary regime (Fig. 2, dashed line), similar to previous findings by Yakubov and co-workers [29]. Even at such low concentrations of particles, particles are entrained and decrease the lubrication behavior of water by almost a factor 2. However, the friction coefficient is much higher than that of 50 % particles (Fig. 2). At lower particle concentrations the load per particle is higher leading to larger indentation depths between the particle and the PDMS surface and a larger overall contact area and higher friction coefficient. The relatively high friction coefficient for 10 % particles in suspension indicates that fluid dominates the lubrication here as similar frictional regimes are found for water and for 10 % particles suspended in water. After the boundary regime, both water and the 10 % suspension show a steep decrease in the friction coefficient, which resembles the Stribeck mixed regime. Using graphene nanoparticles, it was shown that friction reduction may occur when 0.5 % particles are present in suspension [30]. Additionally, molecular dynamic simulations showed that introducing nanodiamonds as particles, strongly improved the lubrication properties of water with a friction reduction of up to 70 % observed [31]. These experimental and theoretical studies confirm our findings that particles are effective at decreasing friction coefficients.

For pure fluids lubricating (semi-)solid surfaces the mixed regime represents a transition from a S – S contact dominated boundary regime, to a mixed regime where both asperity contact and F – S interactions play an important role, as the fluid starts to form a lubrication layer. For the glass particle suspensions, a different mechanism may be at play; In the first regime of the 10 % particle suspension, some particles are likely present between the surfaces, which is hypothesized based on the lower friction coefficient found for particles compared to that of water alone. This observation indicates that both F – S and S – P interactions are driving the frictional behavior. The friction coefficient of 50 % glass particles suspended in water strongly resembles that of dry particles (Fig. 2). Water is poorly able to wet both the hydrophobic tribopairs and the particles [26,32], hence the lubrication behavior of the suspension is similar to the case where water is absent (dry particles). These glass particles are thus be entrained between the surfaces and generate a strong decrease in the friction coefficient compared to PDMS lubricated by water alone. At high particle percentages and high velocities particle lubrication dominates as the gap size increases due to particles deforming the surfaces and becoming entrained between the surfaces. This increase in gap size is caused by the particles rather than the water as a low viscosity fluid would be unable to generate a large enough (hydrodynamic) gap.

3.3. Matrix viscosity effects on particle entrainment

To better understand the effect of fluid viscosity on particle entrainment, glycerol solutions of 0 %, 1 %, 25 %, 50 %, 75 %, and 95 % in water are combined with 50 % glass particles. The rheology and tribology of the fluid matrix as well as the suspensions are discussed here.

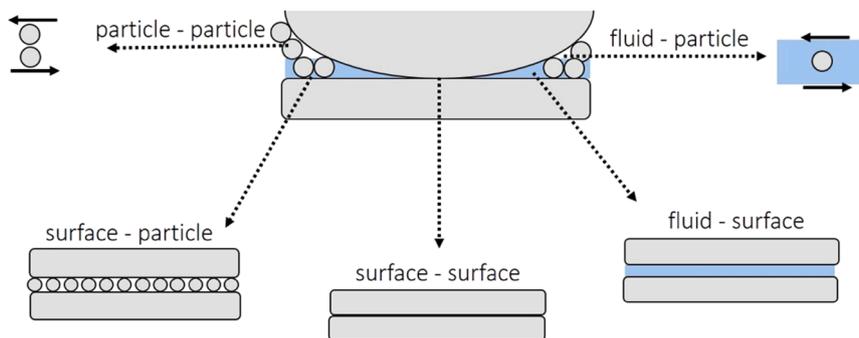


Fig. 1. A schematic representation of the different types of contact that influence the friction coefficient of PDMS surfaces lubricated by glass particles suspended in a fluid. Specifically, P – P interactions, S – P interactions, S – S interactions, F – S interactions and F – P interactions are displayed here.

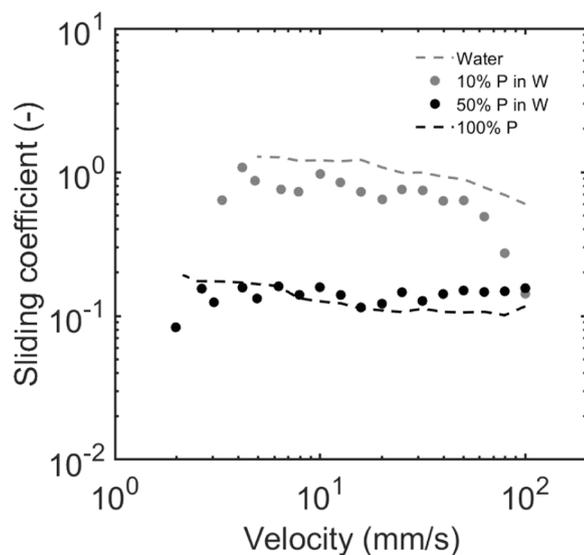


Fig. 2. Tribological properties of water (grey dashed line), 10 % (10 % P in W) and 50 % (50 % P in W) particles suspended in water (black circles) and dry particles (100 % P, black dashed line). In the case of dry particles, the substrate surface is completely covered with particles.

3.3.1. Rheology of particles suspended in a glycerol/water matrix

The low-shear viscosity of glycerol solutions are shown in Fig. 3. The viscosity increases with glycerol content (open symbols) and our results are consistent with literature values [33]. Comparing the glycerol solutions to the particle suspensions with glycerol reveals that the addition of glass particles increases the viscosity (Fig. 3, closed symbols), as expected. The particle sedimentation in the low viscosity matrix (containing 25 % or less glycerol) prevented accurate rheological measurements in the parallel plate geometry. The viscosity of the particle suspension of 25 % glycerol (Fig. 3, dashed line) is therefore estimated by using the measured values of the other glycerol solutions and the Maron-Pierce-Quemada (MPQ) model [34,35]. The MPQ model gives the viscosity of the suspension (η_s) based on the viscosity of the matrix (η_M) and the particle volume fraction (ϕ) as $\eta_s = \eta_M / (1 - \phi / \phi_{max})^{-2}$. The value for ϕ_{max} used here is 64 % assuming randomly packed hard spheres [36,37]. It is important to note that we keep the particle weight percentage constant at 50 %. For the particles in glycerol, this means that the particle volume fraction is between 28 % and 32 % depending on the density of the glycerol/water mixtures. The MPQ expression accurately describes the behavior of our non-interacting particles in suspension as the theoretical values are in line with our experimental results. The theoretical values are thus used as an indication of the suspension viscosity. The suspension viscosity will be used to assess the lubrication behavior in the following sections.

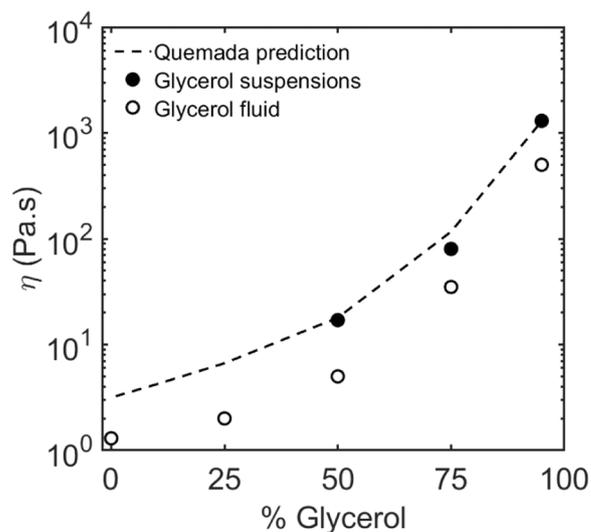


Fig. 3. Measured viscosities of 50 %, 75 % and 95 % glycerol/water solutions (open symbols) and of water/glycerol solutions containing 50 % glass particles (closed symbols). The dashed line represents the theoretical values obtained using the MPQ equation.

3.3.2. Tribology of particles suspended in a glycerol/water matrix

The friction coefficients for different glycerol/water mixtures (without particles) are shown in Fig. 4 (dashed lines). The frictional behavior of 50 % particles suspended in 25 %, 50 % and 75 % glycerol solutions show a strong resemblance to that of the glycerol fluids without particles, especially at low velocities and viscosities Fig. 4 (filled symbols). This indicates that particles do not contribute to the lubrication behavior at this stage and that the frictional behavior is dominated by the viscosity of the fluid matrix. At higher sliding velocities, the curves of the suspensions start to deviate from those of the glycerol fluid and shows a plateau. This plateau indicates that particles begin to contribute to the lubrication at high velocities. Particles in a low viscosity matrix appear to get entrained at higher sliding velocities than particles in a high viscosity matrix Fig. 4.

A higher matrix viscosity thus enables particle entrainment at lower speeds likely due to the larger gap sized caused by the viscosity. For 95 % glycerol suspensions the friction coefficient is dominated by the glass particles over the entire velocity range and is higher than the friction coefficient of the glycerol solution alone. In this case the viscosity is high enough to continuously entrain the particles.

To obtain a Stribeck master curve and correct for the viscosity differences between the sample the velocity (U) is multiplied by the viscosity of the glycerol/water matrix (η_{matrix}) in Fig. 5 [32,38,39]. A reasonable collapse onto a single curve is obtained and three Stribeck

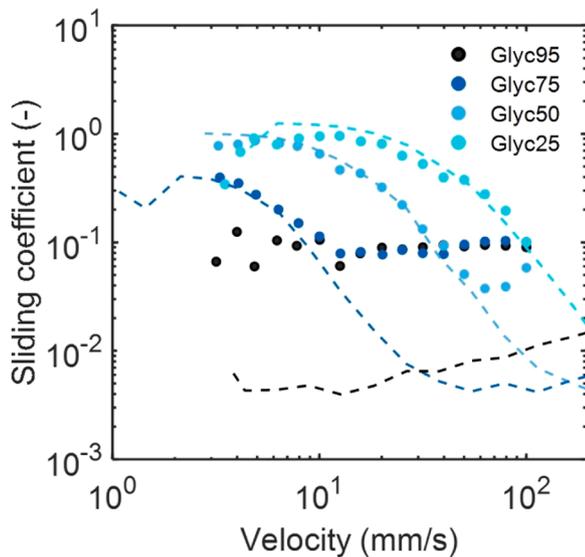


Fig. 4. Sliding friction coefficients of 25 %, 50 %, 75 % and 95 % glycerol/water solutions (dashed lines) and of 50 % glass particles suspended in 25 %, 50 %, 75 % and 95 % glycerol solutions (filled symbols).

regimes are found as expected. For the particle suspensions (filled symbols), however, we observe different regimes than the regular Stribeck regimes. For the lubrication curves of the particle suspensions, using both the matrix viscosity and the suspension viscosity results in a collapsed curve. When correcting for the matrix (glycerol/water solutions) viscosity, the curves of the suspensions and the fluids overlap in the boundary and the mixed regime in Fig. 5a. The viscosity-corrected lubrication curves for the suspension and the solutions overlap until approximately $400 \mu\text{N/m}$. After this, the friction coefficient plateaus indicating particle-mediated lubrication.

These results show that the fluid matrix lubricates the surfaces and determines the frictional behavior at low $U\eta$ values and particle-mediated lubrication dominates at high values and large gap sizes. Previous work using $10 \mu\text{m}$ glass particles confirms the enhanced effect of particulate lubricants at high viscosities; the frictional behavior of (45 %) glass particles in high viscosity fluids (glycerol, corn syrup) deviate strongly from the fluids alone at high velocities (EHL regime). In the previously mentioned work, however, particle lubrication is also observed at lower velocities and viscosities, likely due to the smaller diameter of the particles which allows particles to be easily entrained

[29]. When we construct a Stribeck curve using the viscosity of the suspension instead of the matrix fluid, we find two distinct collapsed curves for the fluids and the particle suspensions with very limited overlap (Fig. 5b). For particle suspensions in glycerol it thus appears that at low velocities (and fluid viscosities),

the fluid matrix provides lubrication. At higher velocities and matrix viscosities, the gap size increases, particles enter the gap and particle-mediated lubrication occurs after $U\eta$ values of $400 \mu\text{N/m}$.

At a value of $400 \mu\text{N/m}$, we estimated the film thickness (gap size) created by the hydrodynamic pressure to be below $5 \mu\text{m}$ using a method as commonly used in similar tribological studies [27,29,40]. With this method, the fluid film thickness is calculated using U (sliding velocity), η (viscosity) W (load), E (elastic modulus of the surfaces) and R (radius of the contact area). Particle entrainment at these $U\eta$ values are not trivial as the gap size is far smaller than the size of the particles ($100 \mu\text{m}$). In this case, particles are likely being entrained due to the deformation of the substrate [38]. Using a finite element model, Xu et al., recently suggested that asymmetric deformation of the substrate occurs due to both shear and normal contact forces. This causes the formation of a cavity in front of the contact with a length scale that is significantly larger than what is to be expected from hydrodynamics alone [38]. This cavity is sufficiently large to facilitate the entrainment of the particles used here.

At high sliding speeds, the contribution of the particles becomes far more apparent, as we find two different sliding regimes: a regime that represents glycerol solutions without particles (boundary and mixed regime) and a curve that represents particle friction (plateau). The effect of the particles is especially noticeable for the suspension with 95 % glycerol and large changes in the frictional behavior are observed. The hydrodynamic regime is absent for the 95 % glycerol suspensions indicating that the contribution of the matrix fluid becomes less relevant. The friction coefficient is determined by the particles and not by the hydrodynamics of the fluid. To obtain hydrodynamic friction the fluid would have to be able to generate a gap larger than the size of the particles. Additionally, at high viscosities, drag forces on the particles are high enough to drag the particles into the gap. The drag force is quantified as $F_d = 6\pi UR\eta$ with U the sliding velocity and R the particle radius and η the matrix viscosity. Hence, at high viscosities (high drag forces) particles remain suspended in the fluid as migrating through the fluid is energetically unfavorable [41]. For our particles suspended in glycerol, we thus find three frictional regimes: (I) boundary regime where the lubricant determines the frictional behavior, (II) a mixed fluid lubricated and asperity contact regime, (III) particle lubrication regime.

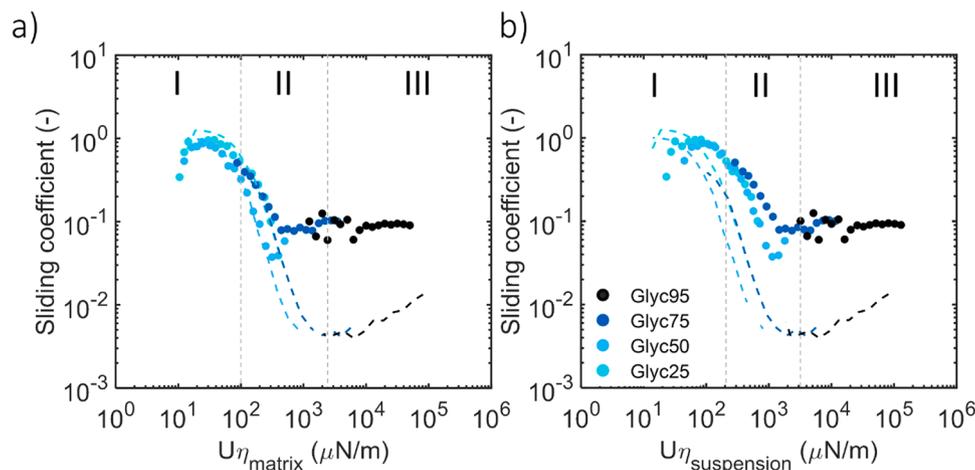


Fig. 5. The sliding friction coefficients of glycerol/water solutions (25, 50 %, 75 % and 95 % dashed lines) and 50 % particles suspended in the different glycerol/water solutions (filled circles). The viscosity was corrected for a) the matrix viscosity and b) the suspension viscosity. In both curves, we find three frictional regimes: (I) boundary, (II) mixed, and (III) particle lubricated.

3.4. Glass particles suspended in oil

In the previous sections, hydrophobic glass particles suspended in (hydrophilic) water and glycerol were discussed. To further investigate the role of F – S interactions, we suspended hydrophobic glass particles in (hydrophobic) mineral oil and the frictional behaviour measured between hydrophobic PDMS surfaces are assessed.

3.4.1. Lubrication and wetting properties of water and oil

We first compare the lubrication behavior of the fluid matrix, mineral oil (Fig. 6, yellow dashed line), to that of water (Fig. 2, grey dashed line). It is immediately clear that mineral oil is much better able to lubricate the PDMS surfaces, resulting in significantly lower friction coefficients for oil ($\mu_{max} = 0.1$) than for water ($\mu_{max} = 1.5$). This difference can be attributed to the superior wetting ability of oil on the hydrophobic PDMS surface, which is influenced by the fluid's surface tension.

Mineral oil, with a lower surface tension (26 mN/m), spreads more easily over the hydrophobic PDMS (surface energy 19–21 mJ m⁻²) [42], creating a continuous lubricating film that effectively reduces friction. In contrast, water has a much higher surface tension (73 mN/m) resulting in worse wetting and a less effective lubricating layer. This leads to the observed high friction coefficient in the case of water, as the fluid is less able to maintain separation between the surfaces. The viscosity, surface tension and hydrophobicity of water, oil and glycerol are found in Table 2. The effect of surface wettability was recently discussed by Gamonpilas et al., and the authors found a strong decrease in the friction coefficient with enhanced surface wettability [39]. Lubricants with poor wettability are easily removed from the contact zone leading poor lubrication and high friction coefficients [32,39].

Moreover, the differences in viscosity (η) between the two fluids further contribute to their lubrication behavior. Mineral oil, with a viscosity of 17 mPa·s, is more capable of separating the surfaces at lower velocities and gap sizes than water, which has a viscosity of 1 mPa·s. The combination of lower surface tension and higher viscosity allows mineral oil to provide more effective lubrication, limiting the boundary regime and resulting in lower friction.

For these fluid lubricants, the F – S interactions dominate the friction

Table 2

An overview viscosity, surface tension and hydrophobicity of the fluids used here. The viscosity reported is as measured. The surface tensions of water and glycerol are obtained from Lide et al., [43] and mineral oil from Mozes et al. [44].

Sample	Viscosity (mPa·s)	Surface Tension (mN/M)	Hydrophobicity/Polarity
Water	1	63	Hydrophilic/polar
Glycerol	340	73	Hydrophilic/polar
Mineral oil	17	26	Hydrophilic/polar

coefficient. It should be noted that the viscosity of mineral oil is similar to that of 50 % glycerol in water. However, limited similarities exist in the frictional behavior between these two lubricants indicating that the effect of viscosity is less important than the effect of hydrophobicity and the F – S.

interactions.

3.4.2. Suspensions of glass particles in oil

In Fig. 6, the lubrication properties of 10 % and 50 % glass particles in oil (grey and black filled symbols, respectively) are shown. Upon the addition of particles to mineral oil, we find three frictional regimes: (I) a decrease in the friction coefficient with an increase in velocity, followed by (II) an increase in the friction coefficient and (III) a subsequent plateau. The curves for 10 % and 50 % particles in oil overlap in regimes I and II. However, in regime (III) a higher friction coefficient is found for the 10 % suspension than for the 50 % suspension. All the oil-based lubricants do show lower friction coefficients than the water-based lubricants and the dry particles. The first regime in Fig. 6 strongly resembles the first section of the measurement conducted with pure oil. This is an indication that particles are not yet entrained and that the behavior is dominated by the fluid matrix. This particle exclusion might be a result of the good F – S contact between oil and PDMS.

A second regime arises at 25 mm/s. At this point, an increase in the friction coefficient

is observed and the curve strongly deviates from that of oil alone (Fig. 6). This increase in the friction coefficient in regime II is an indication of particle inclusion within the gap. This requires the surfaces to deform, and once the particles are present between the surfaces, they will contribute to the friction coefficient. Here it can be seen that particle inclusion causes an increase in the overall friction coefficient. The second regime begins when particles are entrained which gives way to an increase in the friction coefficient. This is in contrast of what we saw for the water-lubricated surfaces where particle entrainment caused a decrease in the friction coefficient. This increase in friction coefficient appears to arrive at a plateau at 60 mm/s. At this plateau, regime III begins. The friction coefficient remains constant, indicating that no changes are taking place in terms of fluid entrainment, particle entrainment, or surface deformation as particles are fully entrained. However, as expected, the particle lubrication regime is limited for particles suspended in oil, as fluid lubrication is favoured for this non-polar fluid between hydrophobic surfaces. In this particle-dominated regime we find that the friction coefficient is much higher than that of oil alone at the same velocity ($\mu = 0.006$). For 10 % glass particles, we find a plateau value of 0.06 and for 50 % glass particles in oil, this value is around 0.03. The lower value for the suspension with more particles is likely caused by more surface separation and less surface deformation due to the presence of more load-bearing particles. In this regime, the S – P interactions are most important.

Interestingly, the oil-suspended particles ($\mu = 0.06$) showed a much lower friction coefficient than dry particles ($\mu = 0.15$). This reduction in friction is likely due to the excellent lubricating ability of oil, especially when combined with hydrophobic surfaces and particles. The lower surface tension of oil compared to water allows it to wet both the hydrophobic particles and PDMS surfaces more effectively, creating a

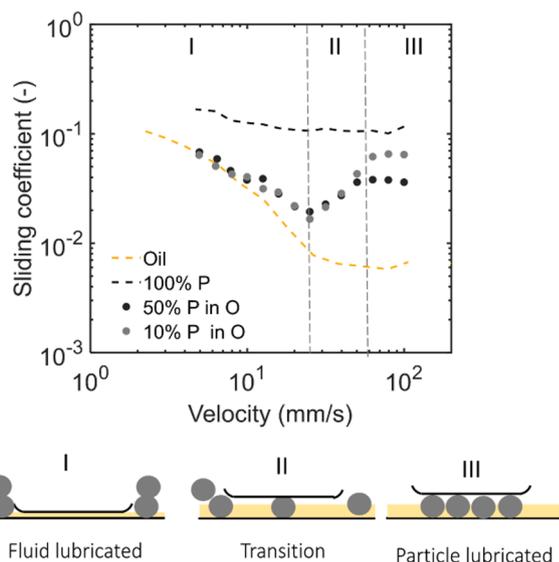


Fig. 6. Lubrication curves of oil (yellow dashed line), dry glass particles (black dashed line), and 10 % and 50 % glass particles (grey and black filled symbols respectively) in oil. The three different sliding regimes are indicated with the grey dashed lines and the mechanisms for each regime are shown below the graph. Regime I is fluid dominated, regime II is a mixed F – P lubricated regime and regime III is the particle lubrication regime.

continuous lubricating film that reduces interparticle friction.

For glass particles suspended in oil, this wetting effect leads to lower friction coefficients than those observed for particles suspended in water or for dry particles. The ability of oil to spread and coat the particles and surfaces results in more efficient lubrication and reduced friction. Additionally, for particles in oil, particle entrainment occurs at relatively high sliding velocities compared to particles in water. This difference is related not only to the fluid's polarity and surface hydrophobicity but also to the influence of surface tension on the interactions between the particles and the fluid. The lower surface tension of oil facilitates better F – P interactions, which likely delays the point at which particles are able to entrain the surfaces.

3.5. Hydrophilic glass particles suspended in water and oil

The results so far show that the interactions between the fluid with the surfaces and the particles lead to diverse frictional behavior. In the previous experiments, both particles and surfaces were hydrophobic. In this section, the hydrophobicity of the particles is adapted. To change the P – F, P – S, and P – P interactions. Particles were washed using a solution of 10 % Decon 90 (a surface active cleaning agent) in water. The adsorption of hydrophobic sides of the surface active agent onto the particles provides a more hydrophilic layer on the outside of the particles, making them more hydrophilic [26]. For glass sheets it was found that the contact angle changed from 47° to 17° after washing with Decon 90 indicating decreased hydrophobicity [26]. We refer to the glass particles that were washed using Decon 90 as 'hydrophilic particles' for simplicity.

3.5.1. Hydrophilic particles in water

The hydrophobic particles are made more hydrophilic by washing the particles using a Decon 90, a surfactant. For hydrophilic particles suspended in water, again we observe multiple frictional regimes (Fig. 7b). A decrease in the friction coefficient is found from $\mu = 0.4$ to $\mu = 0.04$ up until 5 mm/s. Between 5 mm/s and 30 mm/s, an increase in the friction coefficient is seen from $\mu = 0.04$ to $\mu = 0.1$. At this value, the friction coefficient appears to plateau, similar to observations in previous sections.

The first regime for these hydrophilic particles in water strongly resembles the lubrication curve of a 1 % Decon 90 solution indicating that traces of Decon 90 are present (Fig. 7a and b). More importantly, the similarities between the lubrication curves of the suspension and the solution indicates that fluid lubrication is taking place. This is similar to

regime I in Fig. 6 where the first regime overlapped with the lubrication curve of oil at low sliding velocities (and gap sizes). Both oil (hydrophobic) and Decon 90 (surfactant) are considered good lubricants for these surfaces, confirmed by the low friction coefficients measured (Fig. 7). It thus appears that, in this first regime, limited or even no particles are entrained as F – S interactions dominate. Once particles are able to enter the gap, the friction coefficient increases, similar to regime II for particles suspended in oil (Fig. 6). In this case, the lubrication provided by the fluid matrix (water with traces of surfactant), generates a lower friction coefficient than particle-driven lubrication. The regime where sufficient particles are entrained and particle-driven lubrication dominates the friction coefficient (Fig. 7b), displays a constant friction coefficient. This is similar to regime III for the hydrophobic particles in oil (Fig. 6) and dry glass particles.

We find that for suspensions with matrix fluids that are well able to wet the surface (oil, surfactant solution), fluid dominated friction occurs at low sliding velocities and small gap sizes. At high velocities, the fluids are able to separate the surfaces, and at sufficiently large gap sizes the particles are entrained. Above a certain threshold of particles present between the surfaces, particle lubrication dominates and the friction coefficient reaches a constant value.

3.5.2. Hydrophilic particles in oil

For hydrophilic particles suspended in oil, the measured friction coefficients were lower than when the particles were suspended in water (Fig. 7a and b). As expected, it is found that at low sliding velocities the friction coefficients of hydrophilic particles in oil overlap with the friction coefficient of the fluid matrix (oil) alone. The decrease in the friction coefficient in the fluid-dominated regime is followed by a slight increase in the friction coefficient. The change in the friction coefficient, however, is far less apparent than for hydrophilic particles in water (Fig. 7b). This is likely due to differences in the interactions between the particles and fluid well as with the surfaces. The effect of particle hydrophobicity and the relative hydrophobicity of the remaining components of the tribosystem was studied by Qin et al. and the surface, fluid and particle characteristics all appeared to have a specific effect on friction and wear depending on the exact composition of the system [45]. To better understand the potential mechanisms behind particle entrainment, we show a combination of measurements previously discussed: Hydrophobic particles in water, hydrophobic particles in oil, hydrophilic particles in water and hydrophilic particles in oil (Fig. 8).

From Fig. 8a and b it is clear that particles suspended in water (black circles) give higher friction coefficients than particles suspended in oil,

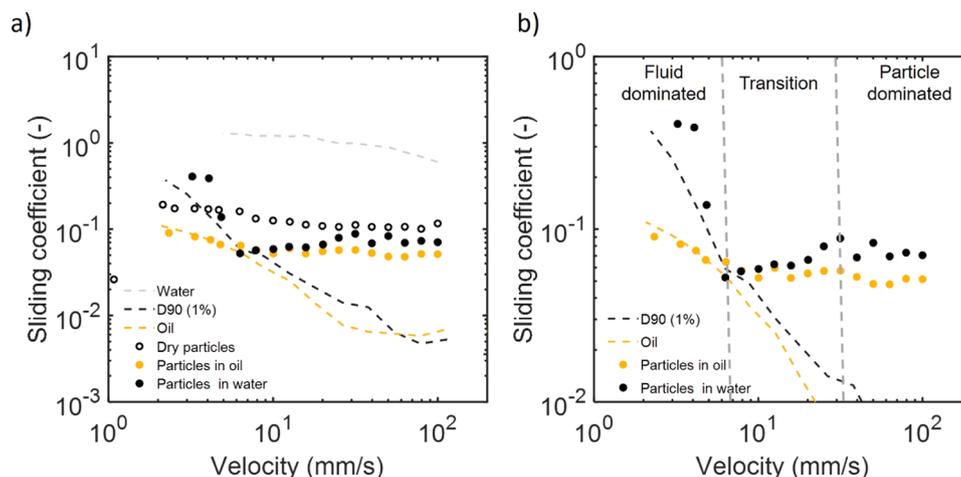


Fig. 7. Sliding friction coefficients of 50 % hydrophilic (washed) particles in oil and in water. a) shows the lubrication curves of water, oil, 1 % Decon 90 in water (to show the effect of (traces of) Decon 90 on the frictional properties of the surfaces) alongside dry particles, hydrophilic particles in water and hydrophilic particles in oil. b) The same data are shown as in a). In this case, the y-axis range is selected to highlight the frictional regimes for the particles in suspension as indicated in the figure.

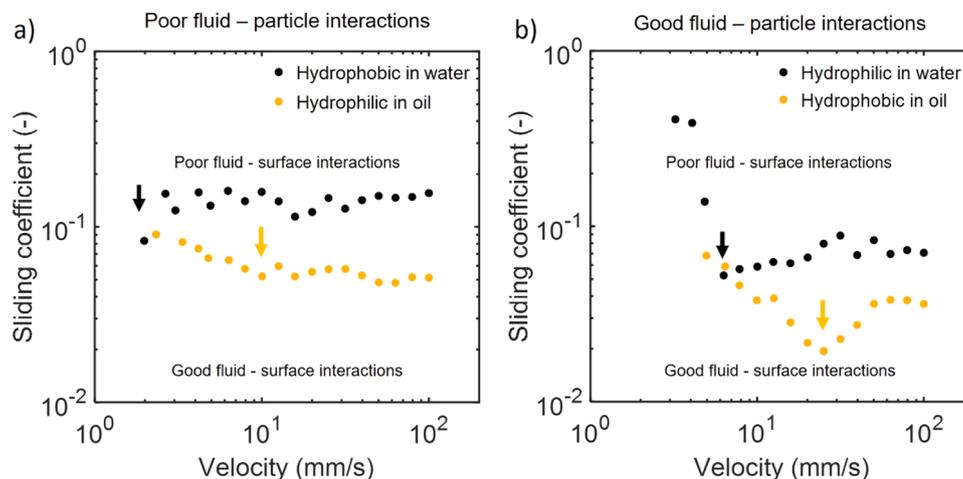


Fig. 8. a) Sliding friction coefficients of hydrophobic particles in water and hydrophilic particles in oil. These particles are expected to show poor interactions (e.g. limited wettability properties) with the matrix fluid. b) Sliding friction coefficients of hydrophilic particles in water and hydrophobic particles in oil. These particles and fluid matrices are expected to have good fluid – particle properties.

for hydrophilic and hydrophobic particles. It thus appears that fluid matrices with good F – S interactions result in lower friction coefficients than poor F – S interactions, regardless of the particle properties. These interactions also affect when particles are entrained as is indicated by arrows in Fig. 8a and b. Particle entrainment seems to occur at relatively low velocities for situations where poor F – P interactions exist (e.g. hydrophobic particles in water and hydrophilic particles in oil (Fig. 8a)). When good F – P interactions exist (Fig. 8b) it can be seen that fluid lubrication dominates over a wider spectrum of the sliding velocity range and particles are entrained at higher sliding speeds for hydrophobic particles in oil. Particle entrainment thus occurs more readily in the case of poor F – S interactions.

4. Main factors leading to particle entrainment

Using hydrophobic and hydrophilic glass particles suspended in various Newtonian fluids, we find that different frictional regimes arise depending on the fluid properties and the particle properties. We have found fluid lubricated regimes, particle lubricated regimes, and transition/mixed regimes. Using these glass particle suspensions it is clear that the interactions between the fluid, particles and surfaces strongly affect the point at which particles are entrained. Depending on the fluid matrix, the friction coefficient either increases or decreases upon particle entrainment.

For hydrophobic particles in a hydrophilic medium, particle lubrication occurs over the entire frictional regime. This is likely the case as the hydrophilic medium is poorly able to wet both the particles and the surface while the hydrophobic particles are readily entrained between the deformable hydrophobic surfaces.

Once particles are entrained hydrophilic particles in a hydrophilic medium, show a lower friction coefficient than hydrophobic particles in a hydrophilic medium. In this case, the fluid is well able to wet the particles which may enhance the lubrication properties as both water and particles are lubricating the surfaces due to good F – P interactions. The same is found hydrophobic and hydrophilic particles in oil. When good F – P interactions exist the friction coefficient is lower (hydrophobic particles in oil) than for poor F – P interactions. It was also observed that particles suspended in oil all display a lower friction coefficient than particles suspended in water. This is expected as oil is hydrophobic and good F – S interactions exist. The magnitude of the friction coefficient thus seems to be driven by the suspension matrix and the F – S interactions. The moment of entrainment is an effect of the F – P interactions. It appears that poor F – P interactions are favourable for particle entrainment. This is seen for hydrophobic particles in an

hydrophilic medium (water) which are entrained at a lower sliding velocity than hydrophilic particles in a hydrophilic matrix. Additionally, hydrophilic particles in a hydrophobic matrix (oil) are entrained at a lower sliding speed than hydrophobic particles in a hydrophobic matrix. Particles suspended in highly viscous glycerol are also entrained at lower sliding speeds than particles suspended in a glycerol/water solution with lower viscosities. The entrainment in this case is attributed to the high viscosity which drags particles into the gap and make it difficult for particles to migrate out of the gap. Based on our findings enhanced particle entrainment is thus driven by increased particle concentration, poor F – S wettability, poor F – P wettability as well as a high viscosity of the surrounding fluid. The mechanism are displayed schematically in Fig. 9.

The results shown here were obtained using solid glass particles that are 100 μm in size. When we compare our results to findings in literature obtained using suspensions made of hollow 10 μm glass particles [29], deviating frictional regimes are found. In case of smaller particles, particle lubrication dominates at low gap sizes, whereas an EHL regime is found due to the fluid formation of high viscosity fluids [29]. Such an EHL regime is found as the fluid film thickness exceeds the particle diameter. For our larger particles, the viscous fluid is unable to increase the gap size to values larger than the particle size, and an EHL regime is not obtained. These differences highlight the importance of the particle size on the frictional properties of suspensions. The friction coefficient is thus strongly dependent on the characteristics of the system, in particular the type of particles and fluids used.

5. Conclusions

The frictional behavior of hard (glass) particles in suspension between two soft (PDMS) surfaces was investigated to create a better understanding of particle entrainment in relation to interactions among the fluid, particles, and surface. It was shown that particles can enter the gap in different situations, even at gap sizes that are theoretically far smaller than the size of the particles. This likely occurs due to the deformability of the soft surfaces. Particles and fluids enter the gap separately or simultaneously depending on the suspension properties.

The properties of the suspension components (e.g. viscosity, polarity, hydrophobicity) lead to specific interactions between the fluid matrix, glass particles and PDMS surfaces which ultimately lead to fluid or particles being entrained between the surfaces. Two main lubrication regimes were observed: fluid lubrication and particle lubrication. Fluid lubrication dominates at low particle volume fractions at low sliding velocities and low matrix viscosities. Particle lubrication dominates at

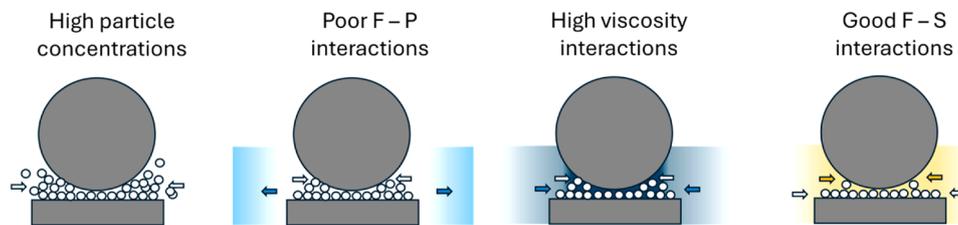


Fig. 9. Mechanisms behind particle entrainment shown schematically. Surfaces are shown in grey, particles in white and fluids is light blue, dark blue or orange. Arrows indicate the direction of the fluids and particles.

high particle percentages, high matrix viscosities, and poor fluid – particle wetting. Overall, we have shown that the frictional regimes for hard particles lubricating soft surfaces are a direct consequence of the ability of particles to entrain the surfaces. The entrainment is driven by the particle – fluid interactions as well as the fluid – surface interactions. At sliding velocities and fluid viscosities at which particles are able to become and remain entrained, particle lubrication becomes more dominant over fluid lubrication. Future investigations into similar systems could study the effect of different particle sizes, surface hydrophobicity and surface roughness on the friction coefficients of soft surfaces and particulate lubricants. The findings presented here contribute to an enhanced understanding of frictional properties of suspensions. This may lead to improved lubrication in the context of food, medical devices and skin lotions. Future research on the subject of particle entrainment is required to develop more comprehensive predictive physical models or numerical simulations describing the interactions at play. Novel methods to visualise the contact zone and particle entrainment in situ would further enhance the understanding of the lubrication behaviour observed.

Statement of originality

The authors hereby declare that the results and conclusions presented in this manuscript are original and written by the authors. The words/works of others are referenced in an appropriate manner.

CRediT authorship contribution statement

Raisa Rudge: Writing – original draft, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Joshua Dijkman:** Writing – review & editing, Validation, Supervision, Project administration, Methodology, Conceptualization. **Elke Scholten:** Writing – review & editing, Validation, Supervision, Project administration, Methodology, Funding acquisition, Conceptualization. **Heather Shewan:** Writing – review & editing, Supervision, Methodology, Investigation, Formal analysis. **Jason Stokes:** Writing – review & editing, Validation, Supervision, Resources, Project administration, Funding acquisition, Conceptualization.

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.

Data Availability

Data will be made available on request.

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