Speed-dependent mean force and fluctuations between fluid-immersed PDMS surfaces upon sliding

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ABSTRACT
In our previous work on shear flows of densely packed PDMS (polydimethylsiloxane) particles [arXiv: 2002.07329], we have found that the system exhibits an intriguing transition zone of driving rates, and interpreted the behaviors as consequences of the speed-dependent friction between particles. To provide solid supports for the interpretation, we set a force balance for time-resolving the contact force between two curved PDMS surfaces that are sliding against each other. Two types of experiments include the passing of two semi-spheres, and the sliding with a sustained contact between two orthogonal semi-cylinders of which one is moving at a controlled speed $U$ with a fixed depth of compression $D_p$ into the other. For sphere-passing experiments, the tangential force reveals a characteristic speed $V_c$, beyond which the time-averaged value decays substantially with the increase of $U$, whereas the normal force shows no detectable change. The characteristic speed $V_c$ appears insensitive to the change of $D_p$ and subsequently to the normal force. Data from experiments with a sustained contact follow the similar trend. We have also verified that the normal force shows a dependence on $D_p$ with a Hertzian scaling. Varying the viscosity $\eta$ by two decades lead to shifts of the speed dependence and we find that $\kappa \equiv \eta U$ provides a good collapse of data, in consistence with prior studies. In addition, we study the fluctuation of tangential force over time. We have found that prominent stick-slip patterns occur only at sufficiently high driving speeds. The change of pattern suggests a change from a regime with sustained solid-to-solid contact, to that of mixed lubrication where the direct contact is intermittent. Detailed analyses on the fluctuations reveal a high sensitivity to surface condition, presumably due to factors such as long-time absorption of molecules, the effects of wearing, or a combination of both.
In our previous work on shear flows of densely packed PDMS(polydimethylsiloxane) particles, we have found that the system exhibits an intriguing dependence on driving rates\(^1\). With the packing density kept fixed, the system goes from a state with a plateau of a high mean stress, via series of transitional states exhibiting prominent stick-slip fluctuations with its mean stress decreasing with the increase of driving rate, eventually to a regime where grains appear to be well lubricated and flow smoothly. We have interpreted such transitions as the consequence of the speed-dependent tribology between particle surfaces. To test such hypothesis, we design a system of force balance to resolve the contact force over the course of sliding, either between two spheres or, alternatively, with an enduring contact at a fixed depth of overlap. Our goal is to understand how the dynamics between contacting PDMS surfaces depends on the speed of sliding \( U \).

There has been a long tradition in tribology on studying the friction between two solid surfaces sliding against each other. Effects from material factors, such as elasticity, roughness, and the interplay with fluid in-between surfaces, have also been studied extensively\(^2,3\). These studies have been done in various combinations of geometry of contacts, also known as *tribopairs*, including but not limited to sphere-to-plane, pin-to-plane, and cylinder-to-cylinder\(^2,8\). For instance, the sphere-on-plane geometry is advantageous in providing a long distance of steady sliding. On the other hand, the cylinder-to-cylinder retains a symmetry that can be advantageous from the theoretical point of view. To the best of authors’ knowledge, studies with a tribopair using a sphere-to-sphere setup have not been reported, despite its relevance to flows of granular particles.

In tribology, the notion of Stribeck diagram can be traced back to 1902\(^9,10\). Generally speaking, the diagram describes how the friction coefficient varies non-monotonically on the sliding speed in the presence of fluid lubrication. The non-monotonic relationship, usually with a plateau at the low-speed limit, is used to separate the dynamics into different regimes: *solid-to-solid contact* (often referred to as *boundary lubrication*) for the plateau, *mixed lubrication* for its decline with driving speed, and *hydrodynamic lubrication* as curve for friction coefficient goes past a minimum and increase with the driving speed. In specific contexts, the curve can be described as a function of a dimensionless number \( S \), known as the Sommerfeld number. Traditionally, a Sommerfeld number is usually in the form of \( S = \eta U / \left( \frac{\text{normal force}}{\text{length}} \right) \), depending on the geometry of the tribopair being discussed\(^3,11,12\). Specifically for sphere-to-sphere contact, the definition \( S = \frac{R \eta U}{f_N} \), where \( R \) represents the radius of particle, \( \eta \) the fluid viscosity, \( U \) the tangential speed along contact, and \( f_N \) the normal force, has been derived from low-Reynolds-number hydro-dynamics between hard spheres\(^13\).

The work by Israelachvili and coworkers\(^4\) has pointed out that the tribology between soft surfaces is sensitive to the rheology of lubricants between solid surfaces, the different lubricant may result in the different Stribeck curve and stick-slip patterns. Here, we focus on the simplest case with Newtonian fluid, i.e., glycerol-water mixture between PDMS surfaces that create by molding ---see Supplemental Information. The main goal is to understand how the contact force, both qualitatively and quantitatively, reflects the transition in tribology as we change the driving speed \( U \), as well as the viscosity \( \eta \) of the interstitial fluid. In the first section, we introduce our setup and the two modes of operation. Results on time-averaged force, their dependence on driving speed and fluid viscosity are described in the second section. In addition, we systematically study the fluctuations and the possible indication on the gradual change of surface condition, as we describe in the third section followed by our discussion and summary.
SECTION I: Setup of a vectorial force balance and two types of experiments

Shown as Fig.1(a), four load cells are installed on a fixed block, barely touching a central piece (CP) with steel balls to measure the vectorial force exerted on CP. Omitted from the schematics are (a) a rubber band that keeps the CP from leaving the stationary block but with only a negligible bias on S4; and (b) two extra force gauges that prevent the undesired lateral (y-) movements of the CP, especially in SP mode. They also provide a sensitive check on the mirror symmetry in y. This mechanism also allows the height of CP to be fine-tuned. There are two types of experiment, using different geometries in our tribopair: (1) **Fixed-depth Dragging** (FD) --- see Fig.1(b): A 5cm-long PDMS sample with a radius of curvature \( r = 4.5 \text{mm} \) in direct x is installed on a holder attached to CP. The lower sample is 10cm-long with the same radius of curvature, and is attached to the bottom of fluid container driven by a motorized stage at a speed \( U \) along direction x. The fluid is filled to a level that is just slightly above the contact zone, to ensure a full immersion while the drag from the fluid is minimized. By fine-tuning the height of CP, the distance \( S \) between CP and the fluid container is adjustable and consequently for the pressing depth \( D_p \). (2) **Sphere Passing** (SP) --- see Fig.1(c): One spherical PDMS sample is installed on a holder that is attached firmly on CP. The counterpart is installed on a specially designed container (not shown) that is driven by a rotating motor with a finite amplitude at a specified relative speed \( U \) -- shown schematically on the graph. Following the same rationale in FD experiments, the fluid is filled to a level that is just slightly above the contact zone. The pressing depth \( D_p \) is defined by the anticipated maximal overlap of two PDMS particles during the rotation, as shown by two dashed lines on the graph. The value of \( D_p \) is adjustable by fine-tuning the height of CP.
Fig. 1(a) Schematics for measuring the forces on a light-weighted central piece (CP), which can be adapted to the upper sample in one of the two modes of operation (described below). Force gauges S1, S2, S3, and S4 are installed on a stationary block (represented by a dotted rectangular). (b) Fixed-depth Dragging: The depth of pressing $D_p$ is preset by adjusting the distance between the two semi-cylindrical samples, with the lower one moving at a speed $U$ along $x$. Inset: a photograph of the actual cross-section of the sample holder (in aluminum), with an acrylic mold that seals the holder in creating the sample. (c) Sphere Passing experiments, with the $D_p$ controlled by the distance of CP from the shaft of the motor. The motor creates a circular movement for the lower sample to slide against the upper one at speed $U$. 

$$D_p = S - 2r$$
Fig 2-1 Results from Sphere Passing (SP) experiments: (a) Typical timeseries of $f_x$ and $f_z$ for a sphere-passing event, plotted over the displacement $U_t$. (b) Integrated force responses over one event, $W_x = \int f_x U dt$ and $W_z = \int f_z U dt$, as functions of the driving speed $U$ for different pressing depths $D_p$. Each symbol represents an average over five runs with a standard deviation mostly comparable or smaller than the size of the symbol. (c) the ratio $\mu_{SP} = W_x/W_z$ as functions of $U$. 
Fig 2-2 Dependence of the mean forces on sliding speed $U$ --- (a) tangential force $\langle f_x \rangle$, (b) normal force $\langle f_z \rangle$, and (c) their ratio $\mu_{FD} = \langle f_x \rangle / \langle f_z \rangle$, as functions of $U$ in Fixed-depth Dragging (FD) experiments. Forces are averaged over a total distance of $\sim 2.5$ cm for every driving speed in a run sequence. Each symbol represents the mean over the results from multiple run sequences (up to 10), with an error bar showing their standard deviation that in many cases is within the size of the symbol. Dependence of $\langle f_z \rangle$ on the pressing depth $D_p$ is also shown in the inset to panel (b) in log-log scales.
Fig. 2-3 Effects of fluid viscosity $\eta$. The main graph displays the ratio $<f_x>/<f_z>$ plotted against $\kappa = \eta U$ for three different values of $\eta$, at the same $D_p=0.25$mm for multiple run sequences. The concentrations of glycerol in the aqueous solution are 0, 60% and 90%, respectively. The inset shows the unscaled data as functions of the driving speed $U$, but with values averaged over different run sequences. A characteristic value $\kappa^{FD}_c$ is defined for subsequent discussions.
SECTION II : Results on time-averaged force

Experiments on Sphere Passing

Figure 2-1(a) shows the timeseries of $f_x$ and $f_z$ during one experiment in SP experiments. We integrated force responses over the time and the driving speed $U$, and define $W_x=\int f_x U dt$ as well as $W_z=\int f_z U dt$. Fig.2-1(b) shows $W_x$ and $W_z$ as functions of the driving speed $U$, which the range of $U$ is from ~0.02 to ~10 mm/s, with four different $D_p$ from 0.05 to 0.25 mm. The fluid between the two samples is 60% glycerol. At small values of $U$, both $W_x$ and $W_z$ are insensitive to $U$. However, at driving speeds above a certain value, defined as $V_c$, the measured $W_x$ decays substantially while the change in $W_z$ is insignificant. Interestingly, this decay and the characteristic $V_c$ are insensitive to the change of $D_p$ by a factor of nearly five with a rise of $W_z$ by one decade.

Figure 2-1(c) shows the ratio of $W_x/W_z$, and we can regard it as an effective friction coefficient $\mu_{SP}$. At the slow-speed limit, the values of $\mu_{SP}$ for four different $D_p$ are between 0.5 and 1. For driving speeds beyond $V_c$, the value of $\mu_{SP}$ decays and can become as small as 0.1.

Experiments on Fixed-depth Dragging

The time-averaged force $<f_x>$ and $<f_z>$ as functions of the driving speed $U$ are shown in Fig.2-2, for five different values of $D_p$. For $D_p=0.25$mm, we show data with the driving speed $U$ ranging from 0.04 to 42 mm/s. For other values of $D_p$, data are available up to $U=3$ mm/s. We use 60% glycerol-water mixture as the interstitial fluid. Similarly to SP experiments, $<f_x>$ and $<f_z>$ are insensitive to $U$ at the low-speed limit except with a dependence on $D_p$. We show in the inset of Fig.2-2(b) that the dependence of $<f_z>$ on $D_p$ is consistent with the prediction from Hertzian model. Interestingly, we find that (1) the decay of $<f_x>$ over the increase of $U$ resembles that of SP experiments but appears smoother; and that (2) the FD setup allows us to observe the occurrence of stick-slip fluctuations along the decay of $<f_x>$. Both points will be elaborated in subsequent sections. By the same token, we also define $\mu_{FD} \equiv <f_x>/<f_z>$ as the effective friction coefficient. We note that $\mu_{FD}$ are generally at the same values comparing to $\mu_{SP}$.

Dependence on fluid viscosity---

We conduct experiments with fluids at different viscosities, by adjusting the concentration of glycerol, all at a fixed pressing depth $D_p=0.25$ mm. Here in Fig.2-3, we show data for each run sequence of different driving speeds separately. The non-overlapping symbols reveal a gradual change over the course of experiments. The implication will be revisited in Discussion. In addition, the driving speed $U$ is rescaled by viscosity $\eta$ with the definition $\kappa = \eta U$, and data show a reasonable collapse over experiments of different viscosities spanned over two decades. The inset of Fig.2-2(c) shows the unscaled data as functions of $U$. Our results are consistent with prior findings in the literature.
Fig.3.0 Time series of tangential force $f_x$ at with nine different sliding speeds $U$, displayed over a total distance of 10mm, for a run sequence with $\eta = 212\text{mPa-s}$ and pressing depth $D_p = 0.25\text{mm}$. Circles on the projection onto the side plane represent the time-averaged value. A vertical dashed line indicates $V_c \equiv \frac{\kappa_c^{FD}}{\eta}$.
Fig. 3-1 Time series of $f_x$ plotted against $x \equiv U(t - t_n)$, in which $t_n$ indicates the start time for a sequence of runs at the same speed $U=1.82\text{mm}$. Pressing depth $D_p = 0.25\text{mm}$, and $\eta = 10.5 \text{ mPa-s}$. For visual clarity, the vertical axis is inverted, to emphasize the occurrence of stick-slip pattern and its propagation.
Fig. 3.2 Results for repeating the same sequence of speeds \( \{U_n\} \), as shown on the graph. Pressing depth \( D_p = 0.25 \text{mm} \), and \( \eta = 10.5 \text{ mPa-s} \). The sample maintains a speed \( U_i \) for a fixed distance \( \sim 7.8 \text{mm} \) before it is reset to the starting position for the next speed \( U_{i+1} \). Upon the repetition of a speed sequence, the coordinate \( x \) is shifted incrementally by \( x_m = mS_0 \) for the \( m \)-th sequence. Similarly to Fig. 3-1, the vertical axis is inverted to emphasize the occurrence of stick-slip behaviors, for visualizing the gradual shift of \( V_c \) over the repetition.
SECTION III: Fluctuations --- observations on stick-slip patterns

FD experiments are advantageous for observing the pattern of fluctuations at a constant speed and pressing depth. Fig.3-0 shows timeseries of $f_x$ in a FD mode experiment with a sequence of nine different speeds, in 90% glycerol-water mixture at the pressing depth of $D_p = 0.25mm$. As $U$ goes above $V_c = \kappa_{FD}^p / \eta$ in which is defined in Fig.2-3, the stick-slip pattern occurs. Stick-slip behaviors have been observed in tribopairs of both relatively hard material, like metal$^5$ or mica$^8$, or soft ones such as PDMS$^4$. In our case with PDMS surfaces, we find similar stick-slip patterns persist for an increase of $U$ by a factor of about 40. In this range, the graph shows that the amplitude of stick-slips relative to the mean also stays similar. This serves as a vivid demonstration of the dynamics of mixed lubrication referred in literatures of tribology$^{12}$: The solid-to-solid friction is lost as the fluid invades the spot of contact and builds up a film for lubrication, but only intermittently. The film could be drained, followed by build-up of solid-to-solid contact with the tangential force increasing until the next invasion. The processes repeat cyclically.

We also find that the spatial extent for the stick-slip patterns to persist can spread over the repetition of experiments. Fig.3-1 demonstrates one such example, for which we show timeseries of $f_x$ from a sequence of runs with the same speed, pressing depth, and fluid, up to 40 times. The total distance of sliding is about 25mm for each run in the sequence.

Figure 3-2 shows that not only the spatial extent, but also the $V_c$ can shift over the repetition of experiments. Data from a total of multiple sequences of experiment of the same combination of driving speeds $\{U_n\}$ are displayed. For the purpose of demonstration, we shift the coordinate by

$$x \equiv U_n(t - t_{m,n}) + mS_0, \ m = 1, 2, 3 \ldots$$

in which $t_{m,n}$ stands for the starting time of the n-th driving speed (out of 16) in the m-th sequence (out of 18). The data shows a decrease of $V_c$ by roughly a factor of two, in the course of experiments with the accumulated distance adding up to ~4.5 meters (with the backward movements to the starting point included). We also have carefully monitored and compensated the viscosity of fluid to make sure that the factor of change in the viscosity of the glycerol-water mixture in the full course of experiments is within $O(0.1)$, so is the possible effect on the possible shift of $V_c$.

The phenomena described by Fig.3-1 and Fig.3-2 suggest that the stick-slip behavior serves as a sensitive indicator for the gradual change of conditions on the PDMS surfaces. This can be the consequence of the absorption of molecules from the fluid due to the long-time immersion, in combination with the wearing over a large distance of sliding. See subsequent section for further discussions.
Discussion and Summary

Comparison between FD and SP experiments --- Both Fig.2-1 and Fig.2-2 show a substantial decrease of tangential force with driving speed $U$ away from the slow limit. Limited by the data available at this point, our data only shows the transition between solid-to-solid contact and mixed lubrication in the Stribeck diagram. As for the anticipated increase of tangential force at the range of higher speeds, which is often referred to hydrodynamic lubrication in the literatures, demands further works to completed.

In SP experiments, the tangential force as function of the driving speed shows a sharp decay when $U$ above a certain value, which we define $V_c \sim 3$ mm/s as the onset of transition between solid-to-solid contact and mixed lubrication. However, in the FD experiment, the decay of the tangential force is not so sharp that we can’t find an obvious critical driving speed. Instead, we find the occurrence of stick-slip patterns to be a convenient indicator for a transition from solid-to-solid contact to mixed lubrication. In fact, recalling Fig.2-3, we have verified that stick-slip patterns do consistently occur for $\kappa > \kappa_c^{FD}$, but not otherwise. We can regard the occurrence of stick-slip patterns as a generic indicator for the regime transition, even without an obvious “kink” on the dependence of mean force on the driving speed $U$.

Signs of change in the surface condition --- We speculate that the absence of the kink in FD experiments can be the consequence of the gradual change of surface condition, because the data in these experiments are averaged over much longer time and sliding distance, than those in SP experiments. The gradual change of $V_c$, as shown by Fig.3-1 and Fig.3-2, do suggest changes of surface condition. However, these data do not offer direct evidence to distinguish whether this change comes primarily from wearing or the absorption of molecules from the fluid on to PDMS surfaces due to the long-time immersion. It is likely that both factors are in effect. We do find visually identifiable change on the spot of contact on the upper sample (in the FD experiments) from its initial state. That suggests the long-distance sliding does produce change on the surface at the length scale comparable to visible light $\sim O(400)$nm. Microscopic characterizations of the change await further studies. More information is available at the Online Supplement (http://www.phys.sinica.edu.tw/jctsai/SBD/).
To summarize, we designed an experiment for measuring time-resolving contact force between two curved PDMS surfaces that are sliding against each other with sphere-to-sphere and the cylinder-to-cylinder geometries. We find the tangential force as function of driving speed in both geometries decays when driving speed $U$ beyond a certain value $V_c$, which shows the transition between solid-to-solid contact and mixed lubrication in the Stribeck diagram. On the other hand, the normal force appears in sensitive to the driving speed and agree with Hertzian contact. In the FD experiment, the decay of tangential force against $\kappa$ is not sharp enough to decide $V_c$ easily. However, we find the minimum $\kappa$ where the stick-slip patterns occur in timeseries is a good indicator of the transition from boundary regime to mixed lubrication, defining this $\kappa$ as $\kappa_{cFD}$. We have verified that stick-slip patterns do consistently occur for $\kappa > \kappa_{cFD}$ in all FD experiments. Moreover, we find not only the spatial extent of stick-slip patterns, but also the $V_c$ can shift over the repetition of experiments, which suggest that the stick-slip behavior serves as a sensitive indicator on surface conditions that has a significant different during multiple times of the sliding in glycerin.
References

(1) Tsai, J.-C.; Huang, G.-H.; Tsai, C.-E. Rate-Dependent Stickslips in Steady Shearing: Signature of Transition between Granular Solid and Fluid. *arXiv:2002.07329 [cond-mat, physics:physics]* 2020.

(2) Kim, J. M.; Wolf, F.; Baier, S. K. Effect of Varying Mixing Ratio of PDMS on the Consistency of the Soft-Contact Strubeck Curve for Glycerol Solutions. *Tribology International* 2015, 89, 46–53. https://doi.org/10.1016/j.triboint.2014.12.010.

(3) Bongaerts, J. H. H.; Fourtouni, K.; Stokes, J. R. Soft-Tribology: Lubrication in a Compliant PDMS–PDMS Contact. *Tribology International* 2007, 40 (10–12), 1531–1542. https://doi.org/10.1016/j.triboint.2007.01.007.

(4) Cristiani, T.; Cadirov, N.; Ehrman, M.; Kristiansen, K.; Scott, J.; Jamadagni, S.; Israelachvili, J. Characterizing Dynamic, High-Frequency Friction in Lubricating Complex-Fluid Thin Films Between Viscoelastic Surfaces. *Tribol Lett* 2018, 66 (4), 149. https://doi.org/10.1007/s11249-018-1093-z.

(5) Maru, M. M.; Tanaka, D. K. Consideration of Strubeck Diagram Parameters in the Investigation on Wear and Friction Behavior in Lubricated Sliding. *J. Braz. Soc. Mech. Sci. & Eng.* 2007, 29 (1). https://doi.org/10.1590/S1678-58782007000100009.

(6) Drummond, C.; Israelachvili, J.; Richetti, P. Friction between Two Weakly Adhering Boundary Lubricated Surfaces in Water. *Phys. Rev. E* 2003, 67 (6), 066110. https://doi.org/10.1103/PhysRevE.67.066110.

(7) Ben-David, O.; Rubinstein, S. M.; Fineberg, J. Slip-Stick and the Evolution of Frictional Strength. *Nature* 2010, 463 (7277), 76–79. https://doi.org/10.1038/nature08676.

(8) Drummond, C.; Israelachvili, J. Dynamic Phase Transitions in Confined Lubricant Fluids under Shear. *Phys. Rev. E* 2001, 63 (4), 041506. https://doi.org/10.1103/PhysRevE.63.041506.

(9) Persson, B. N. J. *Sliding Friction: Physical Principles and Applications*; Springer: Berlin; London, 2011.

(10) Bayer, R. G. *Mechanical Wear Fundamentals and Testing*, 2nd ed., rev.expanded.; Mechanical engineering; M. Dekker: New York, 2004.

(11) Røn, T.; Lee, S. Influence of Temperature on the Frictional Properties of Water-Lubricated Surfaces. *Lubricants* 2014, 2 (4), 177–192. https://doi.org/10.3390/lubricants2040177.

(12) Pitenis, A. A.; Urueña, J. M.; Schulze, K. D.; Nixon, R. M.; Dunn, A. C.; Krick, B. A.; Sawyer, W. G.; Angelini, T. E. Polymer Fluctuation Lubrication in Hydrogel Gemini Interfaces. *Soft Matter* 2014, 10 (44), 8955–8962. https://doi.org/10.1039/C4SM01728E.

(13) Fernandez, N.; Mani, R.; Rinaldi, D.; Kadau, D.; Mosquet, M.; Lombois-Burger, H.; Cayer-Barrioz, J.; Herrmann, H. J.; Spencer, N. D.; Isa, L. Microscopic Mechanism for Shear Thickening of Non-Brownian Suspensions. *Phys. Rev. Lett.* 2013, 111 (10), 108301. https://doi.org/10.1103/PhysRevLett.111.108301.