# Viscoelastic Tribopairs in Dry and Lubricated Sliding Friction - Supplementary Information -

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# Experimental

## (a) Dynamic viscosity of lubricant

The shear viscosity of glycerol was analysed using a dynamic shear rheometer (Kinexus Rotational Rheometer, Malvern Instruments) at 25 °C. A cup and bob geometry was used, with a rotating cylinder inserted inside of the cup. Glycerol was exposed to atmospheric conditions for 24 hours before friction tests due to its hygroscopic nature, so the glycerol used to measure viscosity was also prepared in the same manner. The temperature of glycerol was equilibrated for 5 min before testing. Shear viscosity was measured at different shear rates  $(0.1 - 650 \text{ s}^{-1})$ .

#### **Results and Discussion**

#### (a) Viscosity of lubricants

The shear viscosity of glycerol was measured over shear rates of  $0.1 - 650 \ s^{-1}$  (see Fig. S5). Due to its Newtonian behaviour, the viscosity remained constant at ~ 0.9002  $Pa \cdot s$ . Literature value for the shear viscosity of water (over a range of shear rates) of  $0.8 \cdot 10^{-3} Pa \cdot s$  was used in this work.<sup>1</sup>

## (b) Friction measurement between a PDMS probe and a glass substrate

To test the stability of our experimental set-up and compare our systems with previous research, which studied soft-hard contacts mostly, a glass substrate with an elastic modulus of 70,000

MPa,<sup>2</sup> replacing a PDMS substrate, was used as a bottom substrate in dry conditions. Friction traces were obtained like Fig. S1(b). In Fig. S1(c), as the preload decreases and the sliding velocity increases, the friction coefficient increases which is similar to the results from literature.<sup>3,4</sup> The standard deviations of experimental data obtained through three to four times repeated tests present have less than 5% on average.



Figure S1 Friction measurement between a PDMS probe and a glass slide in dry condition: the bottom glass substrate attached in a petri dish travels in x-axis against a stationary PDMS probe at a constant sliding velocity and a constant preload. (a) Experimental set-up. (b) A representative friction trace at a preload of 200 g and a sliding velocity of 0.1 mm/s. (c) The friction coefficient against the sliding velocity at the preloads of 25, 100, and 200 g. Error bars represent standard deviation, though some are smaller than the size of the data point.



Figure S2 Representative friction traces. In dry, (a) smooth sliding was obtained with a P20 substrate at the sliding velocity of 0.05 mm/s under the preload of 100 g, (b) irregular stick-slip friction was presented with a P30 substrate at the sliding velocity of 0.01 mm/s under the preload of 100 g, and (c) stick-slip friction was observed with a P10 substrate at the sliding velocity of 0.1 mm/s under the preload of 25g. In lubricated condition, (d) smooth sliding was obtained in the water with a P10 substrate at the sliding velocity of 0.005 mm/s under the preload of 25 g, (e) irregular stick-slip friction was shown in the water with a P10 substrate at 0.005 mm/s under the preload of 25 g, and (f) stick-slip friction was presented in the water with a P40 substrate at 0.005 mm/s under the preload of 100 g.



Figure S3 The plot of the friction coefficient (between a P10 probe and P25 – P40 substrates) against the sliding velocity. In dry conditions, the friction force was measured at the preloads of (a) 1.962, (b) 0.981, and (c) 0.245 N. In glycerol lubricated conditions, the friction force was examined at the preloads of (d) 1.962, (e) 0.981, (f) 0.245 N. In water lubricated conditions, the friction was tested at the preloads of (g) 1.962, (h) 0.981, and (i) 0.245 N. To improve readability, the friction coefficient between a P10 probe, and P10 and P20 substrates are not included in this figure. Error bars represent standard deviation, though some are smaller than the size of the data point.



Figure S4 The friction force  $({}^{F_{X}})$  against the preload (W) for each substrate at different sliding velocities. In dry condition, the friction force was measured with (a) P10, (b) P20, (c) P30, and (d) P40 substrate. In glycerol-lubricated condition, the friction force was measured with (e) P10, (f) P20, (g) P30, and (h) P40 substrates. In water-lubricated condition, the friction force was measured with (i) P10, (j) P20, (k) P30, and (l) P40 substrate.



Figure S5 Dynamic viscosity of glycerol (after being exposed in the air for 24 hours) as obtained from the rheometer over shear rates of  $0.1 - 650 \text{ s}^{-1}$ .

Experimental conditions			Dry	Glycerol	Water
Substrate	Preload, W (g)	Sliding velocity, U (mm/s)		Friction mode	2
<b>D10</b>	25	0.005	Smooth	S-S	Smooth
		0.01	S-S	Smooth	I s-s
		0.05	Smooth	Smooth	S-S
		0.1	S-S	I s-s	S-S
	100	0.005	Smooth	I s-s	Smooth
		0.01	Smooth	S-S	Smooth
110		0.05	Smooth	S-S	Smooth
		0.1	Smooth	S-S	I s-s
	200	0.005	Smooth	Smooth	Smooth
		0.01	Smooth	Smooth	Smooth
		0.05	Smooth	Smooth	Smooth
		0.1	Smooth	Smooth	S-S
	25	0.005	Smooth	Smooth	Smooth
		0.01	Smooth	Smooth	Smooth
		0.05	Smooth	Smooth	Smooth
		0.1	Smooth	I s-s	Smooth
	100	0.005	Smooth	S-S	Smooth
D <b>2</b> 0		0.01	Smooth	I s-s	Smooth
F 20		0.05	Smooth	I s-s	Smooth
		0.1	Smooth	I s-s	Smooth
	200	0.005	Smooth	Smooth	Smooth
		0.01	Smooth	Smooth	Smooth
		0.05	Smooth	Smooth	Smooth
		0.1	Smooth	Smooth	Smooth
	25	0.005	Smooth	S-S	I s-s
		0.01	I s-s	S-S	Smooth
P30		0.05	S-S	I s-s	Smooth
		0.1	S-S	I s-s	Smooth
	100	0.005	I s-s	S-S	I s-s
		0.01	I s-s	S-S	I s-s
		0.05	I s-s	I s-s	I s-s

Table S1 Friction modes of each friction trace for dry and lubricant contacts: "smooth" denotes smooth sliding, "s-s" denotes stick-slip sliding, and "I s-s" refers to irregular stick-slip sliding.

		0.1	S-S	I s-s	I s-s
	200	0.005	Smooth	S-S	Smooth
		0.01	Smooth	Smooth	Smooth
		0.05	Smooth	Smooth	Smooth
		0.1	Smooth	S-S	Smooth
P40	25	0.005	Smooth	Smooth	S-S
		0.01	Smooth	Smooth	Smooth
		0.05	I s-s	Smooth	Smooth
		0.1	I s-s	I s-s	Smooth
	100	0.005	I s-s	S-S	S-S
		0.01	I s-s	Smooth	Smooth
		0.05	Smooth	S-S	Smooth
		0.1	I s-s	Smooth	Smooth
	200	0.005	Smooth	I s-s	Smooth
		0.01	Smooth	I s-s	Smooth
		0.05	Smooth	I s-s	I s-s
		0.1	Smooth	I s-s	I s-s

Table S2 Regression equations between the empirical friction coefficient ( $\mu$ ) and other key parameters (the sliding velocity (U), the preload (W), the lubricant viscosity ( $\eta$ ), the reduced elasticity ( $E^*$ ), the loss tangent (tan  $\delta$ ), the storage modulus of the substrate (E')): the material properties of the tribopairs ( $E^*$ , tan  $\delta$  and E') obtained at 0.1 and 1 Hz.

Hz	Dry condition	Adjusted R <sup>2</sup>	Lubricated condition	Adjusted $R^2$
0.1	$\mu_{dry} \sim \frac{E^{*4.123} U^{0.175}}{E^{'3.781} (tan\delta)^{3.827} W^{0.451}}$	0.813	$\mu_{lubricated} \sim \frac{U^{0.151}}{\eta^{0.011} E^{'0.163} (tan\delta)^{0.286} W^{0.333}}$	0.89
1	$\mu_{dry} \sim \frac{E^{*3.363} U^{0.175}}{(E' tan \delta)^{3.754} W^{0.451}}$	0.813	$\mu_{lubricated} \sim \frac{U^{0.151}}{\eta^{0.011} (E' tan \delta)^{0.281} W^{0.333}}$	0.888

# References

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