Supplementary Information

Friction through molecular adsorption at the sliding interface of hydrogels: Theory and experiments

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1 Scaling of friction force with contact radius

In what follows, we detail the relationship between the steady-state friction force F_t and the contact radius a as a function of the physical-chemistry of the silica lens. In addition to data obtained with the rotational setup described in the main text, we also report experiments carried out using a rectilinear sliding setup fully described in reference[1] which was operated with the same silica lenses and PDMA film.

Rotational sliding

In Fig. SI.1a, F_t values under rotational sliding are reported as a function of the velocity V for normal loads ranging from 50 to 200 mN (from light to dark colors). In the velocity range under consideration, F_t is monotonically increasing with V in a way which strongly depends on the physical chemistry of the lens (colors in Fig. SI.1) and on the normal load (shades). Steady state values of the contact radius a are reported as a function of velocity for a normal load $F_N = 200$ mN in Fig. SI.1b. It can be seen that the steady-state contact radius is slightly increasing with velocity, *i.e.* with the frictional force. The maximum increase in a is about 5 µm which corresponds to an increase in the indentation depth of a about 30 nm for a static indentation depth $\delta_0 \approx 410$ nm. This increase in the contact size upon sliding is evidencing a coupling between normal and lateral load components which is discussed in the main text within the framework of the adsorption/desorption model.
Fig. SI.1c shows that all the $F_t(V)$ data for a given physical chemistry of the lens collapse on a single master curve if F_t/a^4 is plotted as a function of V. In other words, the average frictional stress σ varies as a^2 or equivalently as the indentation depth δ ($\delta = a^2/R$). This scaling will be the topic of a forthcoming paper where it will be discussed within the framework of a modified Schallamach model taking into account pinning/depinning events within a layer of finite molecular thickness.

Rectilinear sliding

Similar data are reported in Fig. SI.2 for the case of rectilinear sliding, as sketched in this figure. Although a small increase in a with velocity can still be observed (especially for propyl) when $V < 10^{-5}$ m s^{-1}, the main effect here is a strong decrease in the contact radius above this velocity threshold. As fully described elsewhere[1], this decrease in a originates in the poroelastic transport resulting from the displacement of the glass lens on the PDMA film when $Pe > 1$, where Pe is the Péclet number defined as the ratio of the time τ needed to drain the water out of the contact region (the so-called poroelastic time) to a contact time a/V. As discussed in reference[1], the reduction in the contact size and the associated decrease in F_t when $Pe > 1$ can

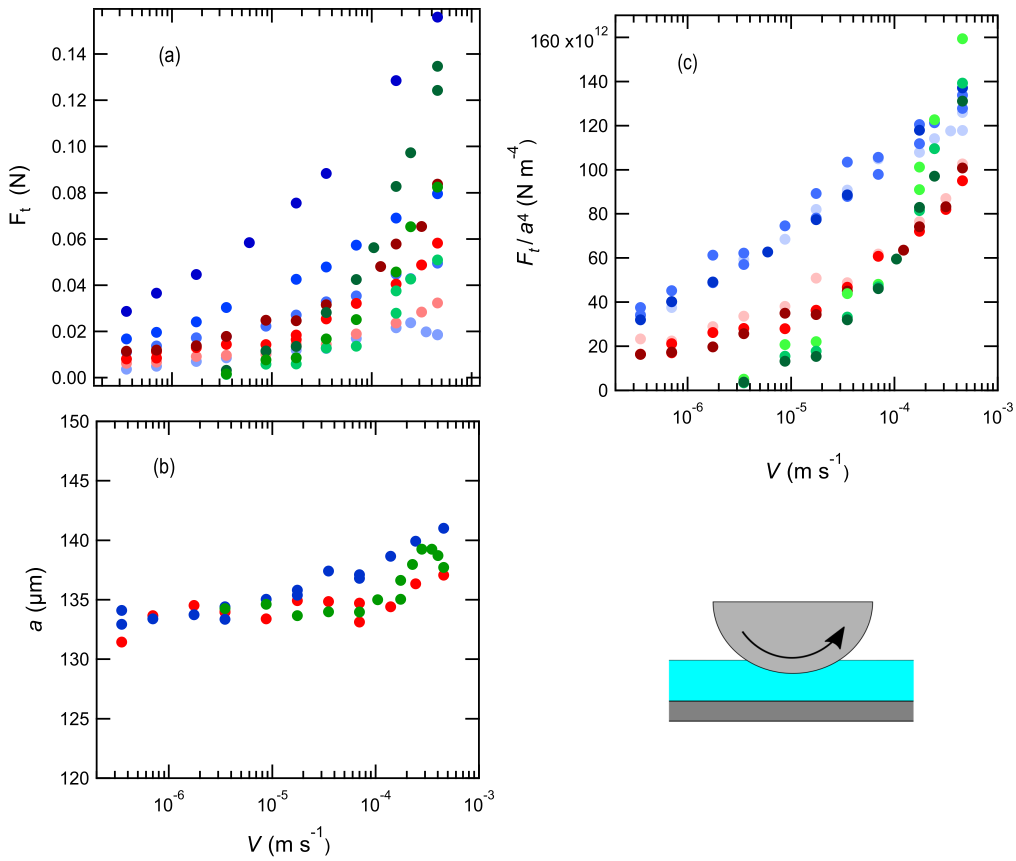


Figure SI.1: Steady-state friction force F_t and contact radius a for rotational sliding. (a) F_t as a function of the sliding velocity V for various normal load F_N and for silica lenses treated with different silanes : propyl (blue), aminopropyl (red) and octadecyl (green). (b) Contact radius as a function of velocity V for $F_N = 100$ mN. (c) Scaled frictional force F_t/a^4 as a function of velocity V. From light to dark colors: $F_N = 50$, 100 and 200 mN (propyl and octadecyl); $F_N = 50$, 100 and 150 mN (aminopropyl).

be accounted for by a pore pressure imbalance between the leading and trailing edges of the contact.

Noticeably, the scaling of the reduced friction force F_t/a^4 with V is preserved under rectilinear sliding even in the presence of poroelastic flows. Some comments are also in order regarding the magnitude of the reduced F_t/a^4 values for rectilinear and rotational sliding. These values are of the same order of magnitude, which indicates that friction by adsorption/desorption predominates over viscous dissipation from poroelastic flows for which a power law $F_t(a)$ with exponent -4.5 had been obtained[2]. The present results suggest the exponent is -4 instead. The two exponent values being very close, our experimental accuracy do not allow us to separate them in rectilinear friction experiments.

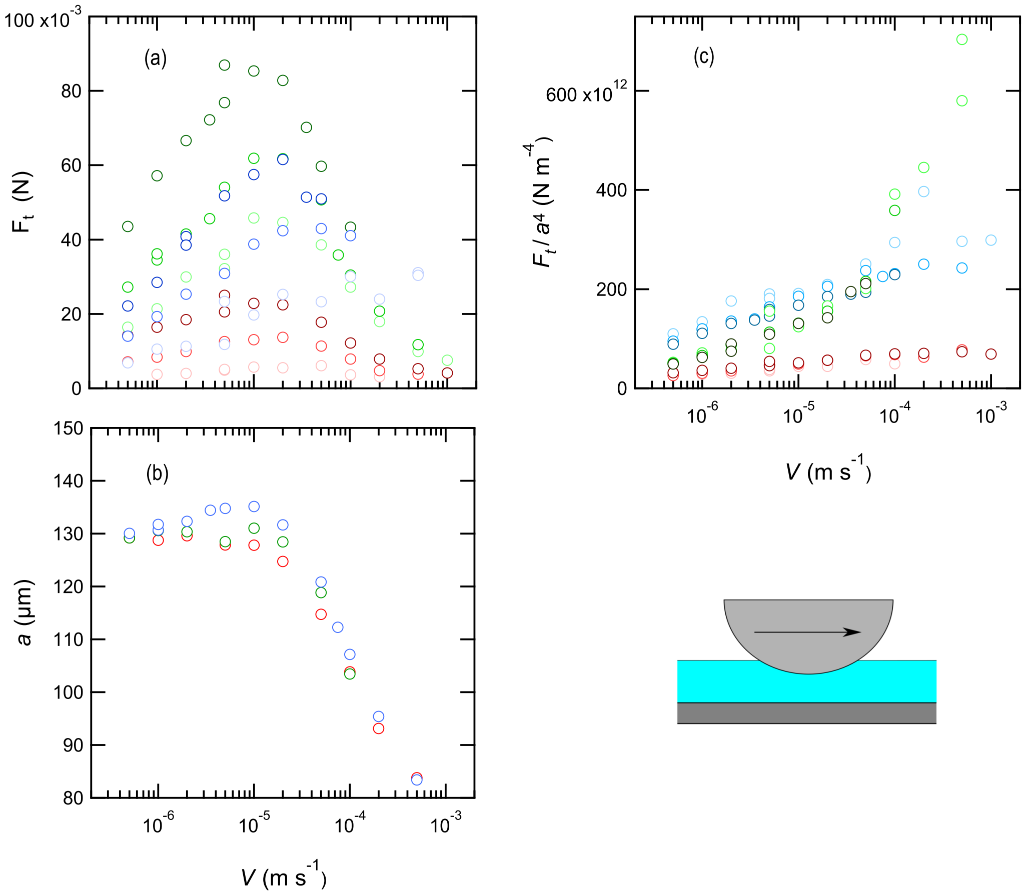


Figure SI.2: Steady-state friction force F_t and contact radius a for rectilinear sliding. (a) F_t as a function of the sliding velocity V for various normal load F_N and for silica lenses treated with different silanes : propyl (blue), aminopropyl (red), octadecyl (green). (b) Contact radius as a function of velocity V for $F_N = 100$ mN. (c) Scaled frictional force F_t/a^4 as a function of velocity V. From light to dark colors: $F_N = 50$, 100 and 200 mN.

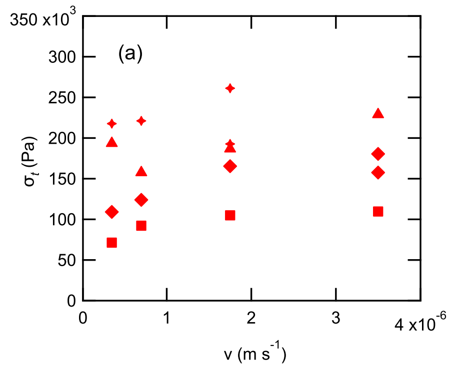
2 Frictional shear stress in the low velocity regime.

We detail here the experimental results obtained in the low velocity regime, *i.e* when the sliding velocity $V \ll uV^*$. From Fig. 4c,d in the main article, such a regime can be identified for the silica lens treated with aminopropylsilane or octadecylsilane where a deviation from the logarithmic behaviour is clearly evidenced at low velocity. According to our friction model, the frictional shear stress in this regime is expected to increase linearly with velocity as

$$\sigma_t = \frac{N_0 kT}{\lambda} \frac{V}{uV^*} = N_0 M V \tau_{ads} \qquad \text{(SI1.1)}$$

In Fig. SI.3, $\sigma_t(V)$ data of Fig. 4c,d of the main text have been reported in a lin-lin representation in the low velocity range. For the aminopropyl surface treatment, there is no evidence of a vanishing frictional shear stress at zero velocity. Moreover, σ_t is dependent on the applied normal load.

For the octadecyl lens, a linear increase in σ_t with V is more likely to occur without any dependence on the normal load. However, a linear fit of the experimental data in this velocity regime (dotted line in Fig. SI.3) provides a slope of 8.10^9 Pa m^{-1} s which is about ten times higher than



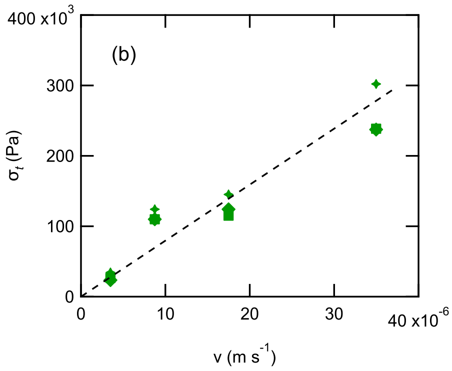


Figure SI.3: Steady state frictional shear stress σ_t as a function of velocity V (linear scale) and applied normal force F_n in the low velocity regime. Silica lens treated with (a) aminopropylsilane; (b) octadecylsilane. Normal load F_n: (●) 20 mN; (■) 50 mN; (♦) 100 mN; (▲) 150 mN; (✦) 200 mN. The dotted line is a linear fit with a slope 8.10^9 Pa.s.m^{-1}.

that predicted with Eq. SI1.1. Taking into account the values of N_0 and τ_{ads} measured in the logarithmic velocity regime and for the chain stiffness $M \approx \frac{kT}{\nu_c b^2}$ (with $\nu_c \sim 10$ and $b \sim 1$ nm), the slope should indeed be of the order of 10^{11} Pa m^{-1} s.

References

[1] J. Delavoipière, Y. Tran, E. Verneuil, B. Hertefeu, C.-Y. Hui and A. Chateauminois, *Langmuir*, 2018, **34**, 9617–9626

[2] L. Ciapa, J.Delavoipiere, Y.Tran, E. Verneuil, and A.Chateauminois, *Soft Matter*, 2020, **16** 6539–6548