Non-Amontons behavior of friction in single contacts

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Abstract. We report on the frictional properties of a single contact between a glassy polymer lens and a flat silica substrate covered either by a disordered or by a self-assembled alkylsilane monolayer. We find that, in contrast to a widely spread belief, the Amontons proportionality between frictional and normal stresses does not hold. Besides, we observe that the velocity dependence of the sliding stress is strongly sensitive to the structure of the silane layer. Analysis of the frictional rheology observed on both disordered and self-assembled monolayers suggests that dissipation is controlled by the plasticity of a glass-like interfacial layer in the former case, and by pinning of polymer chains on the substrate in the latter one.

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1 Introduction

Dry friction between macroscopic “hard” solids commonly involves multicontact interfaces, i.e. interfaces comprised of a set of micrometer-sized contacts between the asperities of the rough surfaces. In order to understand the physical mechanisms responsible for frictional dissipation under such conditions, recent experimental studies of static and low-velocity friction \((V \leq 100 \mu \text{m} \text{s}^{-1}, \text{i.e. negligible self-heating})\) have been performed. Two important features emerge from these studies [1]:

(i) at constant real contact area between the solids, the static threshold stress slowly increases with the time spent at rest, and its growth rate increases with the shear stress applied during ageing,
(ii) the steady-state sliding stress systematically exhibits, in the upper part of the velocity range investigated (typically \(1–100 \mu \text{m} \text{s}^{-1}\)), a regime of quasi-logarithmic increase with velocity.

This phenomenology has been shown to be consistent with the following picture, akin to that of ageing and plasticity of glassy media: frictional dissipation localizes in an amorphous interfacial layer of nanometric thickness, adhesively pinned to the substrates. At rest, this layer is the seat of a glass-like relaxation process that gives rise to static ageing. Sliding rejuvenates the layer, and dissipation occurs by thermally assisted depinning and rearrangement of structural units of volume \(\sim \text{nm}^3\), leading to the observed velocity-strengthening friction at velocities large enough for rejuvenation to be fully effective.

On the other hand, many tribological studies rely on the surface force apparatus (SFA) technique, which involves a well controlled single contact configuration. They mostly focus on the frictional behavior of fluids solidified under the effect of confinement down to molecular thicknesses [2,3], and, recently, on dry friction between thin films of glassy polymers [4,5]. In the former systems, frictional dissipation takes place in the nanometer-thick confined layer. In the latter ones, it involves molecular rearrangements in a nanometer-thick interfacial region between the polymer films, as in the multicontact situation above.

The behaviors observed in SFA and multicontact experiments do share some qualitative features, e.g. static ageing. However, several contrasting results emerge: in particular, SFA studies always evidence unstable (stick-slip) sliding up to a system-dependent critical velocity \(V_c\) in the range \(\sim 0.1–10 \mu \text{m} \text{s}^{-1}\). For \(V > V_c\), friction is always found to be velocity-weakening. This is one of the most striking differences between SFA and multicontact experiments, and two questions arise from it:

(i) To what extent is the sliding dynamics sensitive to the chemical nature of the confined medium on the one hand, and to the strength of its interactions with the confining surfaces on the other hand?
(ii) Does the level of confining pressure affect the dissipative mechanisms, and if so, how?

The latter question is of particular importance. Indeed, in SFA experiments, the confining pressure can be varied typically up to, at most, 10 MPa. This contrasts with the situation at multicontact interfaces, where pressure is self-adjusted and considerably higher: due to the randomness of surface profiles, the number of...
microcontacts adjusts so that, at essentially all usual apparent pressure levels, the pressure exerted on the microcontacts reaches a quasi-constant level close to the yield stress of the material [7]. For a glassy polymer like poly(methylmethacrylate) (PMMA), this pressure is \( \sim 300 \, \text{MPa}, \) i.e. one to two orders of magnitude higher than the pressure in a SFA. This in turn raises the following question: is it legitimate to “extrapolate” the results obtained in SFA experiments to macroscopic situations where the pressure is much higher?

Briscoe and Tabor already addressed, in a pioneer work, the question of the pressure dependence of polymer friction [6]. They studied the shear behavior of glassy polymer thin films, confined between a flat glass substrate and a spherical glass slider. They claimed that their data supported the existence of an Amontons-like linear dependence of the sliding stress on the contact pressure, a statement which has become common wisdom. However, as will be discussed below, close inspection of their results gives no clear evidence for such a linear relationship. We believe that their work suffered from two main drawbacks: poor control of the physico-chemical state of the glass surfaces used, and lack of measurement of the contact area between the solids (their estimate of shear and normal stresses, based on the assumption of a non-adhesive hertzian contact, may thus be biased).

For these reasons, we have revisited the question of the influence of pressure on dynamic friction, taking care to avoid the problems that render the conclusions of Briscoe and Tabor questionable. We report, in this article, on friction experiments performed between a PMMA lens and a flat silica substrate on which an alkylsilane layer is chemically grafted. We have developed an experimental setup similar to that of Vorvolakos and Chaudhury [8], which allows for direct optical measurement of the contact area. This sphere/flat single contact configuration enables us to work at applied pressures in the range 10–70 MPa, intermediate between SFA and multicontact levels.

Under these conditions, we show that, for PMMA sliding on a disordered layer of trimethylsilane (TMS), i.e. the same interface as in previous multicontact experiments [1]:

(i) In contrast with the conclusions of Briscoe and Tabor, the shear stress \( \sigma \), measured in steady sliding at constant velocity, does not increase linearly with pressure. The increase of \( \sigma \) with \( p \) is found to be strongly sublinear in the range of pressure 10–70 MPa. Moreover, extrapolating this nonlinear dependence up to \( p \approx 300 \, \text{MPa} \) leads to a shear stress compatible with the friction coefficient measured in the multicontact configuration.

(ii) The shear stress \( \sigma \), which velocity dependence has been fully investigated at two different pressures, is found to increase logarithmically with velocity over the range 0.1–100 \( \mu \text{m} \, \text{s}^{-1} \). Analysis of these data leads to conclude that the activation volume characteristic of elementary dissipative processes at such an interface is essentially insensitive to pressure in the range 35–300 MPa.

The substrates are 2” silicon wafers covered by a silane layer. The wafers are cleaned as follows: rinsing with...