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Effects of self-affine surface roughness on the friction coefficient of rubbers in the presence of a liquid interlayer

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In this article, we investigate how the friction coefficient is affected by the presence of a liquid layer in between a self-affine rough surface and a sliding rubber surface. The liquid layer will reduce energy dissipation from the small surface asperities and cavities of lateral sizes smaller than the healing length ζ and, therefore, will reduce the coefficient of friction. Significant coefficients of friction are attained for small healing lengths ζ ($\ll \xi$ with ξ the in-plane roughness correlation length). Finally, it is shown that the effect of the substrate roughness exponent *H* becomes less important especially for large healing lengths ($\zeta \ge \xi$). © 2004 American Institute of Physics. [DOI: 10.1063/1.1633338]

I. INTRODUCTION

Clearly, the frictional properties of a rubber body sliding onto a hard solid surface is a relevant issue for the car industry (i.e., tire construction and rubber wiper blades) and the cosmetics industry.^{1–5} The principal characteristics of rubber regarding frictional properties arise from its low elastic modulus *E* and high internal friction.² Moreover, sliding occurs on real solid surfaces, which are predominantly rough, with a significant degree of randomness.^{6,7} In particular, the influence of roughness has to be carefully considered in contact-related phenomena (i.e., friction and adhesion).⁵

The frictional force between a rubber body and a rough solid substrate has two contributions: A hysteric and an adhesive one.¹ The hysteric one arises from the oscillating forces that the surface asperities exert onto the rubber surface leading effectively to cyclic deformations, and energy dissipation due to internal friction.⁵ Thus, the hysteric contribution will have the same temperature dependence as that of a body of elastic modulus $E(\omega)$.⁵ The adhesive component is only important for clean and relatively smooth surfaces.⁵ When a rubber body slides with velocity V over a sinusoidal rough surface with period L, then it will feel fluctuating forces with frequencies $\omega \approx V/L$. The frequency $1/\tau = V/L$ falls in a regime between rubber (low ω) and glass (high ω) behavior.⁵ If the surface has a broad distribution of length scales L, then it will be present a broader distribution of frequency components in the Fourier decomposition of the surface stresses acting on the sliding rubber.⁵

An interesting case arises when a thin liquid layer is trapped within the rubber/solid interface. For fluid films in between rubber/glass, the rubber flexibility leads to liquid entrapment by elastic deformation. Similar effects have been observed for organic liquid films between mica surfaces.^{8,9} Under certain conditions thin and uniformly thick fluid films remain trapped at the rubber/substrate interface.^{10–12} The lat-

ter occurs when electrical charge is introduced upon contact of the surfaces leading to their electrical repulsion. The repulsive force can support a normal load ≤ 0.1 MPa.^{5,12} Typically, a liquid film of about 20 nm thick is stored between the surfaces. Similar effects can be important in biological systems (i.e., polyelectrolyte layers yield low friction in mammalian joints).^{4,5} It has been also found that in the presence of a uniform liquid film, the rubber/glass contact under shear is stable (under contact pressures of about 0.1 MPa). Moreover, friction measurements indicated that the electrolyte solution alone does not effectively lubricate the contact surfaces of sheared films thinner than 10 nm. However, if a surface-active agent is included in the electrolyte solution, monolayer protection prevents surfaces from coming into intimate contact at points where the separating liquid film is locally punctured.5

In any case, the formation and squeezing thin liquid layers between a rubber surface and a hard rough solid substrate is a complex problem. It is related to cavity connectivity, aperture distribution,^{5,13} and the hydrodynamic pressure distribution in the liquid film at the interface. All of these parameters depend on the local pressure and the squeezing time.⁵ So far, it has not been shown how a self-affine random rough surface can affect the coefficient of friction μ_f in the presence of a thin liquid film between the rubber and the solid substrate, which induces attenuation of replicated roughness on the rubber surface.¹⁴

II. THEORY OF FRICTION UNDER CONDITIONS OF INCOMPLETE CONTACT

We assume that sliding occurs with a liquid film confined between the rough solid substrate and the rubber surface (see Fig. 1). For a rubber body of Young's modulus *E* and Poisson's ratio ν that slides with velocity *V*, and provided that contact occurs up to a lateral length scale λ $= 2 \pi/q_{con}$, the coefficient of friction is given by⁵

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FIG. 1. Schematic of the system rubber/liquid film/solid substrate.

$$\mu_{f} = \frac{1}{2} \int_{q_{\text{con}}}^{Q_{c}} q^{3} C^{*}(q) P(q, q_{\text{con}}) dq$$
$$\times \int_{0}^{2\pi} \text{Im} \left[\frac{\tilde{E}(q V \tau \cos \phi)}{(1 - \nu^{2})\sigma} \right] \cos \phi d\phi, \qquad (1)$$

where

$$P(q,q_{\rm con}) = \frac{2}{\pi} \int_0^{+\infty} \frac{\sin x}{x} e^{-x^2 G(q,q_{\rm con})} dx,$$
 (2)

and

$$G(q,q_{\rm con}) = \frac{1}{8} \int_{q_{\rm con}}^{q} q^3 C^*(q) dq$$
$$\times \int_0^{2\pi} \left| \frac{E(qV\tau\cos\varphi)}{(1-\nu^2)\sigma} \right|^2 d\phi.$$
(3)

 $C^*(q)$ is the Fourier transform of the ensemble averaged correlation function $C^*(r) = \langle h^*(\mathbf{r})h^*(0) \rangle$ with $h^*(\mathbf{r})$ the roughness height fluctuation ($\langle h^* \rangle = 0$) at the liquid/rubber interface. σ is the macroscopic applied load and $\tilde{E}(\omega)$ is the complex conjugate of the Young modulus $E(\omega)$ which is given by⁵

$$E(\omega) = \frac{E_1(1 - i\omega\tau)}{1 + \alpha - i\omega\tau},\tag{4}$$

with $E(\infty) = E_1$, and $E(0) = E(\infty)/(1+\alpha)$ ($\alpha \approx 10^3$).⁵ $1/\tau$ is the flip rate of molecular segments (configurational changes) that are responsible for the rubber viscoelasticity.⁵

Furthermore, we have to relate the roughness spectrum $\langle |h^*(q)|^2 \rangle$ of the rubber/liquid interface with that of the substrate surface $\langle |h(q)|^2 \rangle$. Under conditions of complete wetting of the liquid layer onto the rough surface, we have¹⁴

$$h^*(q) = (1 + q^2 \zeta^2)^{-1} h(q), \tag{5}$$

with ζ the so-called healing length. The meaning of the latter is that roughness fluctuations of the substrate/liquid interface at length scales smaller than ζ are smoothened out, and thus they are not further replicated onto the rubber/liquid interface.¹⁴ The healing length ζ is calculated from the relation $\zeta = [\gamma/(\partial^2 U/\partial z^2)_{z=d}]^{1/2}$ with γ as the liquid interface energy at the rubber/liquid interface, d as the averaged liquid layer thickness, and $(\partial^2 U/\partial z^2)_{z=d}$ as the second derivative (calculated at z=d) of the attractive potential U(z) toward the solid flat surface.¹⁴ Here, it is assumed that only weak fluctuations exist, i.e., $w < \xi$, ζ with ξ the in-plane roughness correlation length. In the present case U(z) will also include



FIG. 2. Friction coefficient μ_f vs sliding velocity V for contact length scale $\lambda = 1000 \text{ nm} (\gg \xi)$, $E_1/\sigma = 1000$, $\tau = 2 \times 10^{-3} \text{ s}$, H = 0.4, w = 5 nm, $\xi = 100 \text{ nm}$, and healing lengths ζ as indicated.

the applied load σ . With increasing load, the liquid/rubber interface will increase its degree of conformity with the substrate roughness.

III. RESULTS AND DISCUSSION

As Eq. (1) indicates, in order to calculate the coefficient of friction μ_f the knowledge of the roughness spectrum C(q) is necessary. A wide variety of surfaces/interfaces in nature are well described by a kind of roughness associated with self-affine fractal scaling.⁷ In this case, C(q) scales as a power-law $C(q) \propto q^{-2-2H}$ if $q\xi \gg 1$, and $C(q) \propto \text{constant}$, if $q\xi \ll 1.^7$ The roughness exponent *H* is a measure of the degree of surface irregularity,⁷ such that small values of *H* characterize more jagged or irregular surfaces at short length scales ($<\xi$). The self-affine scaling behavior is satisfied by the equation¹⁵

$$C(q) = \frac{1}{2\pi} \frac{w^2 \xi^2}{(1 + aq^2 \xi^2)^{1+H}},$$
(6)

with $a = (1/2H)[1 - (1 + aQ_c^2\xi^2)^{-H}]$, if 0 < H < 1 (powerlaw roughness), and $a = (1/2)\ln[1 + aQ_c^2\xi^2]$ if H = 0 (logarithmic roughness).¹⁵ The parameter w is the root-mean-square (rms) roughness amplitude, and $Q_c = \pi/a_o$ with a_o of the order of atomic dimensions. For other correlation models, see also Refs. 16 and 17. Finally, from Eqs. (5) and (6), we obtain $C^*(q) = (1 + q^2\zeta^2)^{-2}C(q)$.

Our calculations were performed for $a_o = 0.3$ nm, Poisson's ratio $\nu = 0.5$ (ignoring any weak frequency dependence),⁵ and relatively weak applied loads $\sigma (E_1/\sigma \ge 1)$. Moreover, as is shown in Ref. 5, the factor $P(q,q_{con})$ can be well approximated by the extrapolation formula $P(q,q_{con}) = \{1 + [\pi G(q,q_{con})]^{3/2}\}^{-1/3}$, which makes the calculations of the friction coefficient μ_f simpler. Finally, we consider contact lengths $\lambda \ge \xi$, because it is more physically to occur in the presence of a liquid layer rather than in the case of direct contact of a rubber on a hard solid substrate.

As is shown in Figs. 2 and 3, with increment of the healing length ζ and thus smoothing of the substrate features, which are replicated on the rubber body, the friction coefficient drops very drastically. This is more clearly shown in



FIG. 3. Friction coefficient μ_f vs sliding velocity V for various correlation lengths ξ , contact length scale λ =1000 nm ($\geq \xi$), E_1/σ =1000, τ =2 $\times 10^{-3}$ s, H=0.4, w=5 nm, and healing length ζ =30 nm.

Figs. 4 and 5, where for healing lengths $\zeta > \xi$, the friction coefficient becomes negligible $(\mu_f \ll 1)$. Clearly, the effect of the roughness exponent *H* is rather small, as Fig. 4 indicates. Similar is the situation for the correlation length ξ , which, however, has a larger influence than the roughness exponent *H* for healing lengths practically $\zeta < \xi/2$. Indeed, the dominant influence of the healing length ζ is due to the fact that $C^*(q)$ depends on ζ as an inverse square power law $C^*(q) \propto (1+q^2\zeta^2)^{-2}$ leading to a higher decay rate than the exponent 1+H(H<1) of the roughness contribution through the spectrum C(q).

For large contact length scales $(\lambda \gg \xi)$, the friction coefficient μ_f is given by the simpler form

$$\mu_{f} = \frac{1}{2(1-\nu^{2})} \frac{E_{1}}{\sigma} \int_{0}^{Q_{c}} q^{3}C(q)(1+q^{2}\zeta^{2})^{-2}dq$$
$$\times \int_{0}^{2\pi} \frac{\alpha q V \tau \cos^{2} \phi}{(1+\alpha)^{2}+(qV\tau)^{2}\cos^{2} \phi} d\phi.$$
(7)

Since $C(q) \propto w^2$, the influence of the rms roughness amplitude w on the friction coefficient μ_f is rather simple $(\mu_f \propto w^2)$. Therefore, any more complex dependence on the substrate surface roughness will arise solely from the rough-



FIG. 4. Friction coefficient μ_f vs healing length ζ for sliding velocity $V = 2 \times 10^{-4}$ m/s, $\xi = 100$ nm, contact length scale $\lambda = 1000$ nm ($\gg \xi$), $E_1/\sigma = 1000$, $\tau = 2 \times 10^{-3}$ s, w = 5 nm, $\zeta = 30$ nm, and various roughness exponents *H*.



FIG. 5. Friction coefficient μ_f vs healing length ζ for sliding velocity $V = 2 \times 10^{-4}$ m/s, various correlation lengths ξ , contact length scale $\lambda = 1000$ nm ($\gg \xi$), $E_1 / \sigma = 1000$, $\tau = 2 \times 10^{-3}$ s, w = 5 nm, $\zeta = 30$ nm, and H = 0.4.

ness parameters *H* and ξ (or the ratio w/ξ). If $V\tau \leq \alpha/Q_c$ in Eq. (4), then a simpler form for the friction coefficient μ_f can be found:

$$\mu_{f} \cong \frac{\pi \alpha}{2(1-\nu^{2})(1+\alpha)^{2}} \frac{E_{1}}{\sigma} (V\tau) \\ \times \int_{0}^{Q_{c}} q^{4} C(q) (1+q^{2}\zeta^{2})^{-2} dq.$$
(8)

Equation (8) results in the following two expressions for exponents H=0 and H=1:

$$\mu_{f(H=0)} \cong \frac{\alpha}{4(1-\nu^{2})(1+\alpha)^{2}} \frac{E_{1}}{\sigma} (V\tau) w^{2} \xi^{2} (\zeta^{2}-a\xi^{2})^{-1} \\ \times \left[\frac{Q_{c}}{2\zeta^{2}(1+Q_{c}^{2}\zeta^{2})} -\frac{(3\zeta^{2}-a\xi^{2})}{2\zeta^{3}(\zeta^{2}-a\xi^{2})} \tan^{-1}(\zeta Q_{c}) -\frac{1}{\xi\sqrt{a}(\zeta^{2}-a\xi^{2})} \tan^{-1}(\xi\sqrt{a}Q_{c}) \right], \qquad (9)$$

$$\mu_{f(H=1)} \approx \frac{\alpha}{8(1-\nu^{2})(1+\alpha)^{2}} \frac{E_{1}}{\sigma} (V\tau) w^{2} \xi^{2} (\zeta^{2}-a\xi^{2})^{-2} \\ \times \left[\frac{Q_{c}}{(1+Q_{c}^{2}\zeta^{2})} + \frac{Q_{c}}{(1+a\xi^{2}Q_{c}^{2})} - \frac{(3\zeta^{2}+a\xi^{2})}{\zeta(\zeta^{2}-a\xi^{2})} \tan^{-1}(\zeta Q_{c}) + \frac{(\zeta^{2}+3a\xi^{2})}{\xi\sqrt{a}(\zeta^{2}-a\xi^{2})} \tan^{-1}(\xi\sqrt{a}Q_{c}) \right].$$
(10)

Equations (9) and (10) set analytic physical limits of the friction coefficient so that $\mu_{f(H=0)} \leq \mu_{f(H)} \leq \mu_{f(H=1)}$. These are more important for low healing lengths ($\zeta \leq \xi$) and sig-

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FIG. 6. Friction coefficient μ_f vs sliding velocity V for contact length scale $\lambda = 1000 \text{ nm} \ (\geq \xi), E_1/\sigma = 1000$, various relaxation times $\tau, H = 0.4, w = 5 \text{ nm}, \xi = 100 \text{ nm}, \text{ and healing lengths (a) } \zeta = 10 \text{ nm and (b) } \zeta = 30 \text{ nm}.$

nificant substrate roughness $(w/\xi \sim 0.1)$, since in the opposite limit $(\xi \ll \zeta)$, the friction coefficient is negligible $(\mu_f \ll 1)$; see Figs. 4 and 5).

Clearly, the presence of the liquid layer will remove the contribution to the energy dissipation from the small surface asperities and cavities of lateral sizes smaller than the healing length ζ and, therefore, will reduce the coefficient of friction. Nevertheless, for lower healing lengths (or higher conformity degree of the liquid/rubber interface to the substrate roughness), the effect of the rubber relaxation time on the friction coefficient is more pronounced, especially for relatively high sliding velocities as is shown in Fig. 6.

IV. CONCLUSIONS

In summary, we have shown that the presence of the liquid layer between a self-affine rough surface and a sliding rubber surface will reduce energy dissipation from the small surface asperities and cavities of lateral sizes smaller than the healing length ζ and, therefore, the coefficient of friction will

be reduced. Significant coefficients of friction are attained for small healing lengths ($\zeta \ll \xi$). Finally, it is shown that the effect of the substrate roughness exponent *H* becomes less important, especially for large healing lengths ($\zeta \approx \xi$).

Although, we considered contact lengths $\lambda \ge \xi$ which is likely to occur for a liquid layer in direct contact with a rubber surface under pressure, in future studies, also the case of incomplete contact (between liquid/solid and/or rubber/ liquid) has also to be investigated. Moreover, despite the fact that our work is theoretical, it would be interesting to be tested by rubber sliding experiments on well-defined random rough surfaces in the presence of a liquid layer between rubber/substrate. The substrate roughness can vary from micrometers in size (produced, i.e., by scratch/wear testers) down to nanometers (i.e., polished glass surfaces⁵ or surfaces fabricated by metal evaporation onto metallic substrates, etc.).⁷

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