Micro-scale friction mechanisms of rough stearic acid monolayers

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Fatty acid monolayers are frictional systems that can exhibit rate-and-state characteristics. Using an experimental approach in conjunction with atomistic and continuum computational methods, we investigate the micro-scale mechanisms that result in macroscopic rate-and-state friction for stearic acid deposited onto a surface with sub-nanometric roughness .

Keywords (from 3 to 5 max): stearic acid, monolayer, roughness, friction, aging

1. Introduction

Fatty acids, used in conjunction with lubricants, are often employed as friction-modifying agents in industrial applications to minimize energy dissipation. Their carboxyl group allows them to bind to surfaces and self-assemble into a compact monolayer, which is responsible for the drastic alteration of the friction properties at the contact interface.

In order to identify the underlying physical mechanisms of friction, we combined an approach mixing experimental and numerical friction simulations of well-defined stearic acid boundary layers on surfaces with sub-nanometric roughness.

2. Stearic acid monolayer behavior – exp. simulation

Experiments with the ATLAS tribometer in LTDS showed that stearic acid, and its unsaturated variants, exhibit rate-and-state friction behavior [1]. These results were obtained after deposition of a 2mM solution of fatty acid in dodecane between two Cobalt-coated surfaces of sub-nanometric roughness [1]. Figure 1 shows that the steady-state friction force is velocity dependent, and that changes in the sliding velocity v cause the friction force to peak and adjust over a sliding distance $D_0 \sim 5$ nm. This is typically described with the evolution equation of the age variable ϕ:

$$\frac{d\phi}{dt} = 1 - \frac{v\phi}{D_0}$$

where D_0, in the context of our experiments, was interpreted as the average size of junctions between the surfaces [1].

Rate-and-state behavior of fatty acid monolayer systems can be rationalized by competing viewpoints: one can postulate phase transitions from liquid-like to solid-like states of the monolayer, or surmise an out of equilibrium glassy state which ages and rejuvenates. The former states that the friction force is the sum of contributions of the solid zones of the monolayer, which grow with time when the interface is at rest, and undergo melting when the sliding occurs [2]. The latter viewpoint attributes aging and rate dependency to aging and rejuvenating observed in glassy systems,
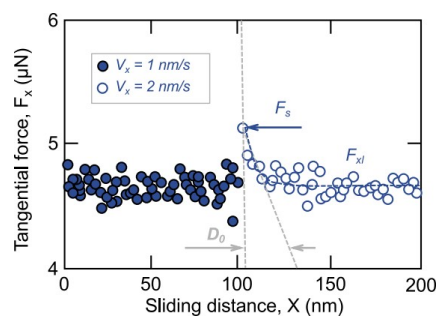

Figure 1: Transient behavior of friction force with increasing velocity on stearic acid boundary layers and under constant normal force (from [1]).

e.g. how the yield stress can be accurately described by a rate-and-state model in glassy polymers [3].

3. Molecular and continuum simulations

In order to validate our interpretation, we employed both molecular dynamics simulations, with coarse-grained potentials for alkane chains to represent the stearic acid molecules, and continuum rough contact simulations using a boundary integral method. Atomic force microscopy measurements of the roughness of surfaces used in experiments were used to evaluate the distribution of micro-contacts and to generate realistic synthetic roughness profiles for the molecular system.

We first showed that monolayers atop flat surfaces with full surface coverage exhibit neither aging of the interface nor friction peaks after changes in sliding velocity. The flexibility available in simulations helps us to identify which source of disorder among the roughness, the imperfect surface coverage of the monolayer or the lubricant gives rise to rate-and-state friction.

4. References

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