Morphology, concentration, potential:
Exploring tunable adsorption film friction with molecular dynamics

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Ionic solutes in aqueous solution aggregate on immersed surfaces. The composition and structure of compact adsorption films and the complementing diffuse layers influence interfacial friction. Active friction control is possible by tuning this adsorption behavior. We focus on three parametric dimensions governing film composition and morphology, namely the involved ions themselves, ion concentrations, and electrostatic potential, and show their impact on friction mechanisms on the molecular level by means of probe-on-flat and flat-on-flat shear simulations via molecular dynamics.

Keywords: active friction control, boundary lubrication, molecular origins of friction, molecular dynamics

1. Introduction

Surfactant adsorption films at the solid-liquid interface undergo phase transitions controlled various environmental parameters [1]. The anionic model surfactant sodium dodecyl sulfate (SDS) forms flat-lying monolayers at low concentrations at the aqueous solution–gold interface. With increasing surface coverage, stripe-like aggregates of hemicylindrical nature assemble. Sufficiently strong attractive electrostatic potential induces another transition towards densely packed bilayers. Active tuning of concentrations and electrostatic potential allows for precise friction control at the nanoscale by navigating the adsorption film's phase diagram [2]. Moreover, the background electrolyte's nature and concentration is known to alter surfactant aggregation behavior as well [3]. We explore the molecular mechanisms behind concentration- and potential-dependent friction response in the model.

2. Methods

By means of all-atom molecular dynamics simulations, we slide an atomic force microscope (AFM) tip model laterally across SDS films at the H_2O–Au(111) interface. Thereby, we shine a light upon the molecular mechanics behind distinct friction force responses of different film phases under shear. Figure 1, for instance, compares sliding on a dense monolayer (a) with sliding across hemicylinders (b) on the basis of friction and load evolutions at the onset of lateral motion. In another batch of simulations, we use a constant potential approach by dynamic charge optimization to investigate potential-dependent friction of sodium chloride, a commonly encountered background electrolyte, in aqueous solutions confined within a nanogap.

3. Discussion

In the particular comparison between sliding on different aggregates shown in Figure 1, the hemicylindrical configuration exhibits less resistance under comparable load. The model allows us to attribute changes in tribological response directly to structural properties, such as molecular alignment within the surfactant film or potential-induced alteration of the saline solution's electrochemical double layer. Automized screening of the parametric space spanned by concentrations and electrostatic potentials enables us to identify parameter-dependent friction trends.

4. References

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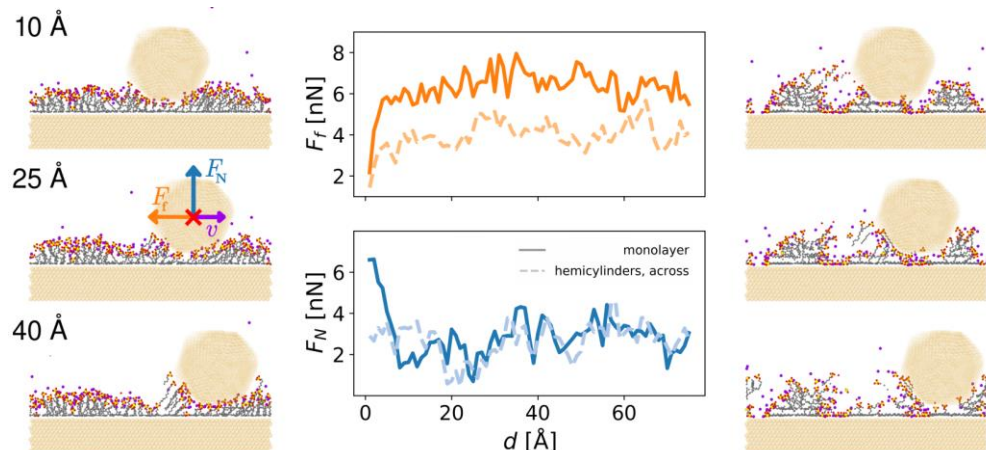
Figure 1: Friction $\mathbf{F_f}$ (top, center) and load $\mathbf{F_N}$ (bottom) felt by AFM tip (radius $r = 25$ Å) during initial sliding phase on SDS monolayer (solid lines in plot and conceptual sketch on the left) and across hemicylinders (dashed lines, right sketch) at surface packing density $n = 3\ nm^{-2}$, fixed surface–surface distance $a = 10$ Å and velocity $v = 10\ m\,s^{-1}$. Labels in left hand side conceptual sketch indicate laterally travelled distance d for each snapshot.