

## Soft and aqueous lubrication

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Aqueous lubrication is of relevance for biocompatible lubricant systems and for the lubrication of soft solids. Computer simulations that generate a molecular understanding of lubricated soft systems are difficult because deformation processes in soft materials span many decades in time scale. This talk describes molecular simulations on three examples of soft and aqueous lubricant systems: Friction of hydrogels, ionic and anionic surfactants as friction modifiers and electrochemical control of friction in aqueous solvents. We find that friction mechanisms are determined by time scale of relaxation mechanisms, such as single chain relaxation in a hydrogel or adsorption time scales for surfactants.

**Keywords (from 3 to 5 max):** hydrogel, surfactant, water, electrochemistry

### 1. Introduction

Aqueous systems for lubrication have relevance in contact lenses, hip joints or for biocompatible lubricants. While these fields have seen much experimental activity, there are few instances of molecular calculations of aqueous and soft lubricated systems. One reason for this is that soft systems show a spectrum of relaxation times often exceeding typical molecular (all-atoms) methodologies.

### 2. Methods

We here employ all-atom molecular dynamics for studying adsorption films that require chemical specificity for predicting adsorbate morphologies. For voltage boundary conditions, we apply a variable charge scheme that allows to keep the two sliding partners at constant (but different) potentials. Soft bulk systems, such as hydrogels, cannot be modeled with all-atom approaches as the relaxation time scales are too long. We use bead-spring models with an implicit solvent for hydrogels to be able to span a reasonable range of time scales.

### 3. Discussion

The three distinct system studied by us reveal friction mechanisms of soft and aqueous systems. In the hydrogel system, friction is determined by the relaxation time of a single chain. During sliding, surface chains are stretched because of the external driving force. The friction coefficient increases with velocity as soon as the frequency of chain excitation exceeds the typical relaxation time of a surface dangling chain. In accordance with experiments [1], this gives rise to a velocity-independent friction coefficient at low velocity crossing over to a power-law behavior at high velocity. Our bead-spring model does not have an explicit solvent and therefore does not include hydrodynamic lubrication, yet it reproduces experimental behavior.

The hydrogel system shows that the surface is important for determining the friction coefficient, since our calculations reveal that the relaxation of the surface dangling chains and not the bulk chains is important. Surfaces can be functionalized with the addition of a

surfactant to the aqueous solution. We study the effect of the anionic SDS and the cationic CTAB on friction forces. Our calculations show a dependence of friction force on surfactant concentration and surface morphology.

Adsorption of SDS and CTAB can be tuned electrochemically and this has been experimentally shown to affect friction [2]. We probe this electrochemical control of friction using a simplified system, a Na-Cl electrolyte between two gold electrodes. Our calculations show [3] that the applied voltage has a pronounced effect on surface slip and this affects the friction coefficient in narrow channels.

These three systems are examples of advanced calculations of soft and aqueous molecular systems that show interesting frictional behavior. In combination with electrochemistry, they may prove useful for realization friction that is electrically tunable.

### 4. References

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