

# Molecular mechanisms of self-mated hydrogel friction

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We developed an approach to simulate self-mated hydrogel friction, based on a bead-spring polymer network. Our model is able to qualitatively reproduce the experimentally observed friction transition between a speed independent and dependent regime, as well as the collapse of the friction curves onto a single curve when the sliding velocity is rescaled. By using an implicit solvent, we disentangle the role played by hydrodynamic interactions and polymer dynamics in the frictional behavior of self-mated hydrogels.

**Keywords (from 3 to 5 max):** self-mated hydrogel, molecular dynamics, polymer

## 1. Introduction

Hydrogels are polymer networks swollen in an aqueous environment. Hydrogels sliding on hydrogels have been found to have a very small friction coefficient.[1] The friction coefficient of self-mated hydrogel systems is velocity independent at slow sliding speeds and transitions to a velocity dependent regime at a mesh size dependent velocity.[2] Current theories propose that at slow sliding speeds fluctuation induced polymer relaxation dominates over the time scale introduced by the shear rate.[2-3] Above the transition the increase of the friction coefficient has been attributed to either hydrodynamic lubrication[1] or non-equilibrium polymer dynamics.[3] The aim of this work is to contribute a computational perspective to current discussions driven by experimental results.

## 2. Methods

### 2.1. Model

Our approach is based on a mesoscopic description of the polymer network. A chain is divided into uncorrelated segments (Kuhn lengths) which are connected by nonlinear FENE bonds. Excluded volume interactions are included via a repulsive Lennard-Jones potential. We construct our network by placing crosslinkers on the lattice sites of the diamond crystal structure. Crosslinkers are then connected via chains of predetermined length. This allows us a precise control over the surface structure. By using periodic boundary conditions, we are able to shear the top surface against the bottom surface. An implicit solvent, induced by a DPD thermostat thermalizes the system.

### 2.2. Results

We run simulations using polymer networks with chain lengths ranging from 10 to 100 beads per chain whilst shearing both surfaces against each other at constant velocity. Our system showed a linear relation between the shear and normal stress. Looking at the friction coefficient, we found a transition between a velocity independent and dependent friction regime. We found the transition velocity to be chain length dependent. The power-law exponent of the velocity dependent regime differs from experimental observations. Rescaling the sliding velocity by the polymer relaxation time and radius

of gyration collapses the friction curves for our different networks (Fig. 1). We show that even simulations run at a lower temperature collapse onto the same curve.

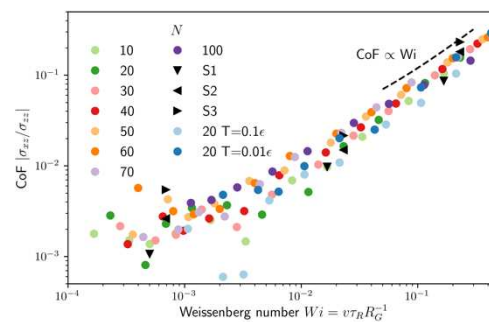


Figure 1: Coefficient of friction as a function of the Weissenberg number (sliding speed times the polymer relaxation time divided by the radius of gyration) for networks of chain length  $N$ .

## 3. Discussion

We have shown, that self-mated hydrogel contacts locally follow Amontons' law of friction, in contrast to experimental observations. This is consistent with current theories that experimental observations can be attributed to Hertzian contact mechanics and slider geometry. We further have developed a simulation model that successfully reproduces the friction transition. We confirm the thermal origin of the speed independent friction regime. Our model qualitatively reproduces a collapse of the friction curves with respect to the Weissenberg number observed in experimental data.[2]

## 4. References

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