



In Situ Measurements of Contact Dynamics in Speed-dependent Hydrogel Friction

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ABSTRACT

The friction behavior of soft, aqueous, biotribological contacts depends on contact geometry, speed, and pressure. Previous efforts to measure the surface profile of soft sliding contacts have been stymied by the matching index of refraction of aqueous materials submerged in water. Here, hydrogel surface deformations were imaged using confocal microscopy to experimentally investigate the contact geometry as a function of sliding speed. *In situ* fluorescence confocal microscopy measurements of the contact deformation during unidirectional friction experiments revealed the existence of a front/back asymmetry that increased with increasing sliding speed. A polyacrylamide hydrogel disk (96.5% water) was polymerized with fluorescent dyes and used as a rotating countersample below a polished glass hemispherical pin (1 mm radius of curvature). All experiments were performed submerged in a dilute (0.7 wt%) suspension of 1 μm red fluorescent microspheres in ultrapure water. Imaging of the contact was performed using a confocal microscope *in situ* with unidirectional sliding at 0.1, 1, 10, and 100 mm/s. The friction coefficient increased monotonically with increasing speed from $\mu \sim 0.04$ at 0.1 mm/s to $\mu \sim 0.20$ at 100 mm/s. All imaging was performed relative to the stationary glass probe under steady-state conditions. The contact line measured by connecting the leading and exiting contact points was nearly perpendicular to the loading direction at 0.1 mm/s sliding speed, but distorted as sliding speed increased. Consistent with the theories of viscoelastic contributions to polymer friction, the tangent of the contact-line angle correlated with friction.

1. Introduction

Aqueous lubrication with soft hydrogels, tissues, cartilage, and biopolymers frequently reveals a friction dependence on sliding speed. A number of different mechanisms have been suggested to explain this dependence, including a hypothesis of viscoelasticity in which a “plowing” component of friction is sensitive to sliding speed. As discussed by Bonnevie et al., the plowing contribution of friction is attributed to the integrated pressure distribution having a component parallel to the sliding vector but in the opposite direction [1]. Bonnevie et al. also discussed that these plowing contributions, “cannot be determined without known pressure distributions”. The prevailing theory is that viscoelastic effects lead to an asymmetry in the pressure distributions – viscoelasticity is frequently measured and modeled for cartilage, tissues, and high-water content gels [2–6].

A recent finding using self-mated contacts of identical hydrogel samples (Gemini hydrogels) revealed a surprising speed independence in friction coefficient that spanned over 2 orders of magnitude in sliding

speed [7], which was notable as the majority of healthy biological sliding interfaces are in Gemini configurations. This study raised questions as to whether or not *in vivo* biological interfaces experienced speed-dependent friction, or if they were effectively constant friction interfaces. In an effort to tease out the fundamental mechanisms underlying the friction behavior of biological interfaces, high-water content hydrogels have been used as convenient surrogates for *in vitro* studies [7–9]. The optical clarity, tunable mechanical and transport properties through changes in the mesh size (ξ), wide range of water content, and ease of fabrication have made these hydrogels nearly ideal experimental samples for studies of aqueous lubrication. One recent, and perhaps surprising, discovery was that friction coefficient decreased with increasing mesh size, and that this could continue into the regime of superlubricity ($\mu < 0.005$) [9,10]. As elastic modulus falls rapidly as mesh size increases (E proportional to $\xi^{-1/3}$) [11]. These superlubricious hydrogels were almost “slime-like” in their mechanics as elastic modulus falls rapidly with increasing mesh size ($E \sim \xi^{-1/3}$) [11], suggesting that the softest surface gels in bio-lubrication may be

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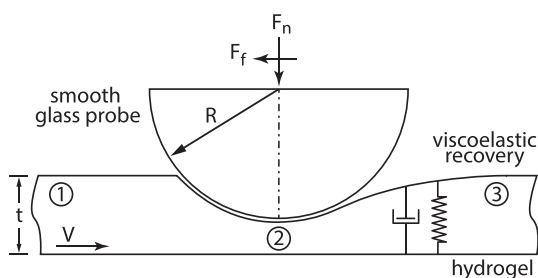


Fig. 1. An illustration of contact showing the modified Winkler foundation model which adds a viscoelastic component to the response of the material undergoing sliding friction. Considering a viscoelastic material undergoing dynamic deformation, three distinct regions of deformation become apparent: (1) the *inlet* region, or leading edge; (2) the *apex* region at the tip of the indenting probe; (3) the *exit* region, or trailing edge. Adapted from Rennie et al. [28].

responsible for the low friction coefficients that rival values found under hydrodynamic lubrication.

Physiological sliding speeds span a wide range from nm/s for cell migration to over 100 mm/s for blinking and joint movements. Gemini hydrogel experiments performed over a range in speed, 30 $\mu\text{m/s}$ to 100 mm/s, revealed a critical transition speed at which friction coefficient rises with increasing sliding speed, and this transition may be related to the polymer relaxation time [8,9]. Speed-dependent friction behavior has also been observed in cartilage systems [12], and the hypothesis for this behavior was a form of viscoelasticity in which fluid depressurization in the cartilage matrix due to fluid exudation in the wake of contact resulted in a time-dependent recovery following deformation. Bonnevie et al. argued that in a contact configuration of a rigid sphere sliding against a flat countersample of cartilage, the trailing half of the contact could not provide meaningful load support and thus only the front half supported the load. This concept was consistent with simulations by Kusche [13] that modeled the sliding of a probe over an indented viscoelastic half-space at sufficiently high sliding speeds (Fig. 1).

The hypothesis of a viscoelastic effect on friction for soft biomaterials is essentially one of leading/trailing edge contact asymmetry. Earlier studies have investigated the deformed surface profile of flat transparent elastomers loaded under a clean glass hemispherical probe and used Newton's rings to determine contact area in static and dynamic conditions [14]. Barquins and Courtel directly observed an asymmetric contact patch of a soft elastomer under hemispherical contact develop as sliding speed increased but were unable to image the surface profile. Schulze et al. [15] observed a complex speed-dependent folding and contact patch evolution when sliding soft rubber spheres against smooth glass countersurfaces. Krick et al. and Persson et al. used similar methods under liquid-lubricated conditions to resolve changes in contact area during the sliding of soft hemispherical probes against smooth glass surfaces [16,17]. The insight provided by these experiments for understanding elastomer tribology suggests that direct visualization of the contact patch and adjacent regions during sliding may also be necessary to elucidate the mechanisms of speed-dependent friction at hydrogel surfaces. Further, direct observations and measurements of hydrogel surface profiles during loading and sliding may reveal insights into the nature of biological sliding contacts, such as those found in cartilaginous joints or the cornea-eyelid interface.

Here we describe *in situ* experiments measuring the surface profile of a deforming hydrogel sample sliding against a spherical glass probe. These experiments were performed and imaged using 3D confocal laser-scanning microscopy under a condition of dynamic equilibrium where fluorescent particle dispersions were used to delineate the borders between the hydrogel surface, water, and the glass probe. Imaging was performed relative to the stationary spherical probe that was loaded into contact with a submerged hydrogel countersurface spinning under steady angular speed. The deformation profiles over a range of sliding

speeds from 0.1 mm/s to 100 mm/s were analyzed and compared to changes in friction coefficient as a function of sliding speed.

2. Materials and Methods

2.1. Hydrogel Preparation and Characterization

Polyacrylamide (pAAm) hydrogel disks (~ 10 mm thick) were polymerized at room temperature from a precursor solution, reported as weight-solute per weight-total percentages: 3.75% acrylamide monomer, 0.15% *N,N'*-methylenebisacrylamide cross-linker, 0.15% ammonium persulfate initiator, 0.15% tetramethylethylenediamine catalyst in ultrapure water (18 M Ω). FluoroMax green fluorescent microspheres (5 μm diameter) were included in the precursor composition at 0.002 wt% in solution. To ensure flatness and low surface roughness, disks were cast between two polystyrene culture dishes ($R_a \sim 20$ nm). After curing, the disk diameter was reduced to 24 mm and equilibrated in ultrapure water for ~ 5 days prior to experimentation. The elastic modulus of the gel ($E \sim 500$ Pa) was determined from indentation experiments following the methods in Pitenis et al. [8], except with a hemispherical borosilicate glass probe (1 mm radius of curvature) in this study.

2.2. In Situ Friction Experiments

Unidirectional sliding was performed using a pin-on-disk microtribometer mounted to a confocal laser-scanning inverted microscope (Nikon C2). A double-leaf cantilever (normal stiffness of 148 $\mu\text{N}/\mu\text{m}$, and lateral stiffness of 69 $\mu\text{N}/\mu\text{m}$) with two 3 mm capacitive displacement sensors were used in combination to measure normal and lateral forces during contact and sliding. The cantilever was secured below an XY-axis translation stage (OptoSigma TAM-602 high precision linear stages), which was mounted in place of the microscope condenser head. A high-speed piezoelectric rotary stage (Physik Instrumente M-660.55, 4- μrad resolution) was fixed to the microscope stage. The hydrogel disk was placed in a custom dish with an optical window and mounted to the rotary stage. A 17.5 wt% pAAm was polymerized in the free space around the soft sample and thus secured it in place. A hemispherical borosilicate glass probe (1 mm radius of curvature) was attached to the cantilever. During sliding, the hydrogel disk and glass probe were completely submerged in a 0.7 wt% dispersion of FluoroMax red fluorescent microspheres (1 μm diameter) in ultrapure water, to provide an additional image channel to delineate the deformed hydrogel surface.

The experiments described herein were very sensitive to misalignments so accurate measurements of hydrogel surface deformations depended on adequate leveling prior to sliding. The hydrogel disk was leveled with respect to the probe to within ± 20 μN by loading the glass probe into contact with the gel to a normal force of 1 mN, then rotating the disk in 360° increments and performing fine adjustments using three leveling screws on the custom dish. After the leveling procedure, the glass probe was unloaded and the hydrogel disk equilibrated for ~ 2000 s. A reference surface was established by imaging the undeformed surface of the rotating hydrogel disk at the prescribed constant speed (0.1, 1, 10, or 100 mm/s).

Before and after each set of sliding speed experiments, the hydrogel disk was allowed to equilibrate for ~ 2000 s before reimaging to confirm a level surface. The glass probe was translated using the XY-axis stage to a new location on the hydrogel and loaded to a normal force of 1 mN. The hydrogel rotated beneath the probe at the prescribed constant speed (0.1, 1, 10, or 100 mm/s) with an 8 mm radius contact path and a contact width of ~ 1.5 mm, which ensured negligible errors in friction coefficient [18]. The normal force was not controlled during sliding, which allowed free amplitude variations due to potential rate-dependent deformations of the hydrogel surface.

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