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Normal Load Scaling of Friction in Gemini Hydrogels

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Abstract:

Health and physiology are critically dependent on the ability of soft, permeable, and aqueous materials (e.g. cartilage, cells, and extracellular matrix) to provide lubrication over a wide range of speeds and contact stresses. Living cells and tissues present tremendous handling and experimental challenges for fundamental biotribology studies. Synthetic high water content hydrogels, designed to share similar mechanical and transport properties of biomaterials, can provide fundamental insights into the basic dissipative mechanisms associated with aqueous lubrication. Recent studies investigating the response of self-mated (Gemini) hydrogels to a wide range of sliding speeds under constant load conditions revealed transitions in friction behavior that may be associated with polymer relaxation time and contact time for a surface mesh during sliding (mesh size divided by the sliding speed). Here, the extent to which contact pressure and contact area affect hydrogel friction behavior was explored by changing the applied load over two orders of magnitude (0.1–20 mN) and the sliding speed over four orders of magnitude (10 $\mu\text{m/s}$ –100 mm/s). Oscillating pin-on-disk microtribological experiments were performed in ultrapure water for Gemini polyacrylamide hydrogels (average mesh size ~ 7 nm). Friction coefficient decreased across all ranges of sliding speed with increasing applied load, consistent with predictions of contact area scaling non-linearly with applied load and pressure-independent surface shear stresses. The contact area for Gemini hydrogel interfaces under these conditions has been shown to follow Hertzian contact mechanics theory, and supports the scaling of friction coefficient in the speed-independent regime that follows $\mu \sim F_n^{-1/3}$.

1. Introduction:

The lubrication of biological interfaces is undoubtedly one of the most complicated and challenging systems to theorists and experimentalists; in part, because biology is fundamentally alive, evolving, and dynamic. In many ways high water content synthetic gels parallel these interfaces, and recent work with hydrogels in matched (Gemini) configurations has shed some light on the basic dissipative mechanisms associated with aqueous lubrication [1–3]. In these experiments the assumptions have been that although submerged and predominately aqueous, they are self-lubricated in direct contact not under hydrodynamic or fluid film lubrication. High water content hydrogels differ from biological systems in numerous important ways, including: being

manufactured from synthetic polymers without the addition of proteins, inanimate, homogeneous in composition, nanometer-scale surface roughness, isotropic mechanics, and neutral in charge [4–7]. In spite of these differences, hydrogels remain a convenient and potentially very useful system for theoretical and experimental studies of aqueous lubrication. The ability to specify the water content, polymer concentration, and mesh size, while maintaining optical transparency with excellent long-term stability [8] makes hydrogels nearly ideal specimens for tribological studies that aim to explore the sensitivity of gel friction to perturbations in stress, speed, and other contact conditions including surface roughness, materials, and contact aging [1–3,9,10].

One of the most often discussed aspects related to the tribology of cartilage, hydrogels, tissues, tissue microenvironments, and cell layers is poroelasticity and the role of water in supporting loads and providing low shear sliding interfaces [11–16]. Numerous sophisticated models of poroelastic deformation and predictions of tribological behaviors have been developed over the past 50+ years, often with the expressed purpose of determining the evolution in friction and contact area over time under conditions of either constant or dynamic applied loads [17–22]. In biological tissues, the permeability, modulus, and anisotropic mechanics add numerous levels of complexity and challenges (including opacity) making complementary measurements and model validations exceedingly difficult, and often preclude direct experimental quantification of the phenomena being modeled.

Hydrogels provide a unique platform in which to study aqueous lubrication. The mechanics of hydrogels are well studied, and a single parameter, mesh size (ξ), determines the hydraulic permeability (κ) [23], osmotic pressure (Π) [24], elastic modulus (E) [24], and recently was found to be the dominant parameter in the scaling of friction coefficient (μ) [3]. As friction coefficient is almost always a system parameter, and not a material property, the mesh size control of friction is somewhat misleading. However, it is an exciting concept that has revealed a surprising result: large mesh size and high water content hydrogels were the lowest friction systems under identical loads and sliding speeds for a wide variety of mesh sizes. Further, work exploring the response to Gemini hydrogel tribology over a wide range in sliding speeds has revealed transitions in behavior that are thought to be linked to the polymer relaxation times (τ) and the ratio of the mesh size to the sliding velocity (ξ/v) [2,3]. However, all of these previous studies were performed under constant load conditions.

In this manuscript, we examine the degree to which contact pressures and contact area will alter the friction behavior of hydrogels. This was done by changing the applied load over two orders of magnitude and the sliding speed over four orders of magnitude. All sliding experiments were performed completely submerged in ultrapure water in the Gemini configuration. The hydrogels were made from polyacrylamide ($\xi \sim 7$ nm) following a method previously used and characterized via small angle X-ray scattering (SAXS) as reported in [3].

2. Materials and Methods

Three identical sets of hydrogel probes and disks were used for these experiments. Hydrogel samples were prepared by polymerizing the following components: acrylamide (7.5 wt%), N,N'-methylenebisacrylamide (0.3 wt%), ammonium persulfate (0.15 wt%), and tetramethylethylenediamine (0.15 wt%) in ultrapure water (18.2M Ω)

as described in Urueña et al. [3]. The solid hemispherical hydrogel probes were cast in a diamond-turned polyolefin mold producing a radius of curvature of ~2mm and a surface roughness below 20 nm. Hydrogel disks were cast in a polished polystyrene dish (~60mm diameter and ~5mm thickness). After polymerization, the hydrogel disk was cut to a 40mm diameter and both the hydrogel probe and disk were allowed to equilibrate in ultrapure water for at least 24 h prior to experimentation.

Tribological experiments were performed on a high-speed, oscillating pin-on-disk microtribometer, shown schematically in Fig. 1a and previously described [2,3]. The hydrogel probe was polymerized around a stainless steel threaded fastener bonded to a titanium double leaf cantilever flexure that had a normal stiffness of 160 $\mu\text{N}/\mu\text{m}$ and a lateral stiffness of 75 $\mu\text{N}/\mu\text{m}$. Capacitance sensors (Lion Precision Elite Series, 5 $\mu\text{m}/\text{V}$ sensitivity, 20 V range) mounted axially and tangentially to the cantilever assembly above the hydrogel probe, were used to measure normal (F_n) and friction (F_f) forces, respectively. The combined standard uncertainties in normal and friction force measurements using this configuration were correspondingly $\pm 2 \mu\text{N}$ and $\pm 1 \mu\text{N}$. The friction coefficient is calculated as the ratio of the measured friction force divided by the normal force continuously, and the experimental uncertainties in friction coefficient are discussed in more detail in the literature [25,26].

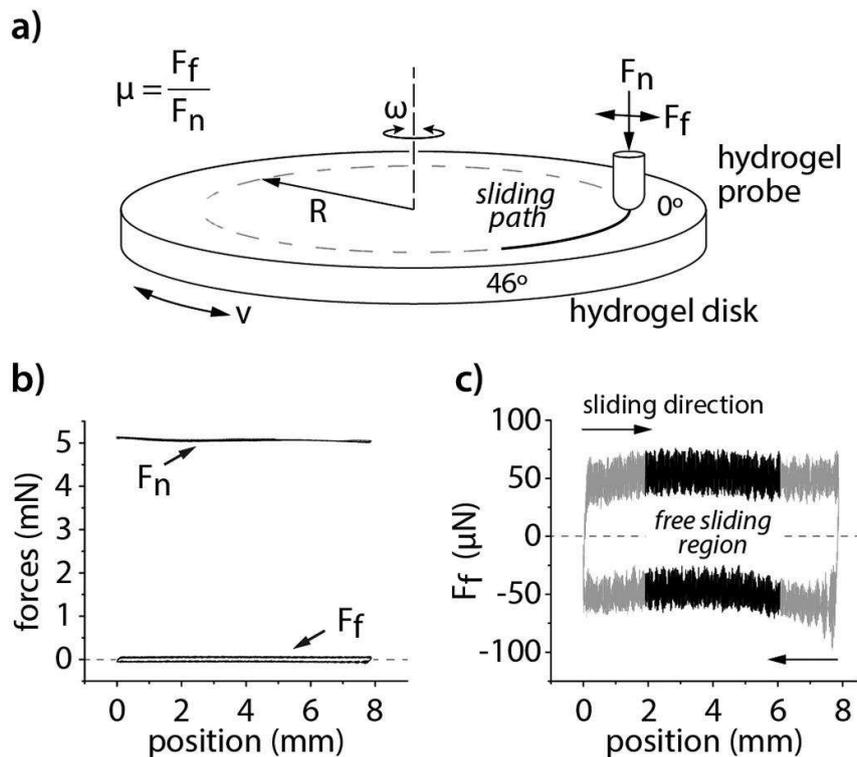


Fig. 1. a) The Gemini hydrogel configuration consisted of a stationary hydrogel probe (2mm radius of curvature) mounted to a cantilever and loaded against an oscillating hydrogel disk (40mm diameter, ~5mm thickness). Both the hydrogel probe and the disk were composed of 7.5 wt% polyacrylamide, 0.3 wt% bisacrylamide with a mesh size of 7 nm[3]. The dark line illustrates the oscillating sliding path from 0° to 46° and back to 0° per cycle. b) Normal (F_n) and friction (F_f) forces for a reciprocating cycle at a sliding speed of 1 mm/s and nominal normal force of 5 mN. c) Detail of the friction force data shown in b), which were two orders of magnitude lower than the normal force. The horizontal arrows indicate the forward and reverse sliding directions. The black data points highlight the free sliding region, over which the average friction coefficient was calculated for each cycle.

The hydrogel disk was mounted to a piezoelectric rotary stage (Physik Instrumente M-660.55, 4 μ rad resolution), capable of rotational speeds from 0.001 to 720°/s. In this study, the stage oscillated over 46° (Fig. 1a) and two track radii were used to achieve the large range of sliding speeds: 5mm for lower speeds (0.01–0.5 mm/s), and 10mm for higher speeds (1–100 mm/s). The oscillatory path allowed for ample free sliding regions of constant speed between reversals: ~4mm for the low speed experiments, and ~8mm for the high speed experiments. The error analysis for friction measurements within small wear tracks was studied by Krick et al. [27] and was included in the uncertainty analysis of these data. To reduce normal force variations associated with angular misalignments, the hydrogel disk was levelled through fine adjustments around the periphery of the holder until a suitably low level of run-out was achieved. A typical trace of normal forces and friction forces during a reversal are shown in Fig. 1b.

To evaluate the effect of increasing normal force on Gemini hydrogel lubrication, eight different normal forces spanning two orders of magnitude (100 μ N to 20 mN) were each tested across thirteen different sliding speeds spanning four orders of magnitude (0.01 to 100 mm/s). Experiments were performed from high to low sliding speeds, and from low to high normal forces. For each experiment, the hydrogel probe was brought into contact with the hydrogel disk and loaded to the prescribed normal force. The normal force was maintained for approximately 300 s prior to sliding. For sliding speeds between 200 μ m/s and 100 mm/s, the duration of the sliding experiment was 20 cycles. The slower speed experiments (1 μ m/s and 100 μ m/s) were performed over 10 cycles. Fig. 1b shows the stability of normal and friction forces over the entire sliding path during a single oscillation. Average friction coefficients for each cycle were calculated from the free sliding region of each friction force trace (Fig. 1c) using the methods in Burris et al. [26]. The average friction coefficient was calculated over 20 cycles for the faster experiments and over 10 cycles for the slower experiments.

During all tribological experiments, the hydrogel probe and disk were fully submerged in a bath of ultrapure water (pH~7). The rotary microtribometer apparatus was mounted on a passive vibration isolation stage set on an optical table and housed within a polymethylmethacrylate (PMMA) chamber to reduce noise associated with ambient air currents and acoustic sources.

3. Results and Discussion

When using an appropriate sample thickness such that the radius of contact area remains small relative to the thickness of the sample, the contact mechanics of these high water content hydrogels have recently been shown to follow Hertzian behavior [28] for indentations lasting $<10^3$ s [29–31]. This is an important finding for experimental studies, because performing in situ observations of contact area during sliding introduces significant experimental challenges [32]. In these experiments, we examine one of the oldest hypotheses in tribology: is friction force linearly dependent on the applied load? [33–36]. For these sphere-on-flat configurations, the expected behavior is the classic example of decreasing friction with increasing applied load if they follow the expected Hertzian mechanics and the dissipation is related to a shear stress at the surface of the gels that is not pressure-dependent. Numerous researchers have predicted and found this scaling for ideal sphere-on-flat

configurations for elastomers [37], polymers [38,39], metals [39], and natural materials including minerals [40].

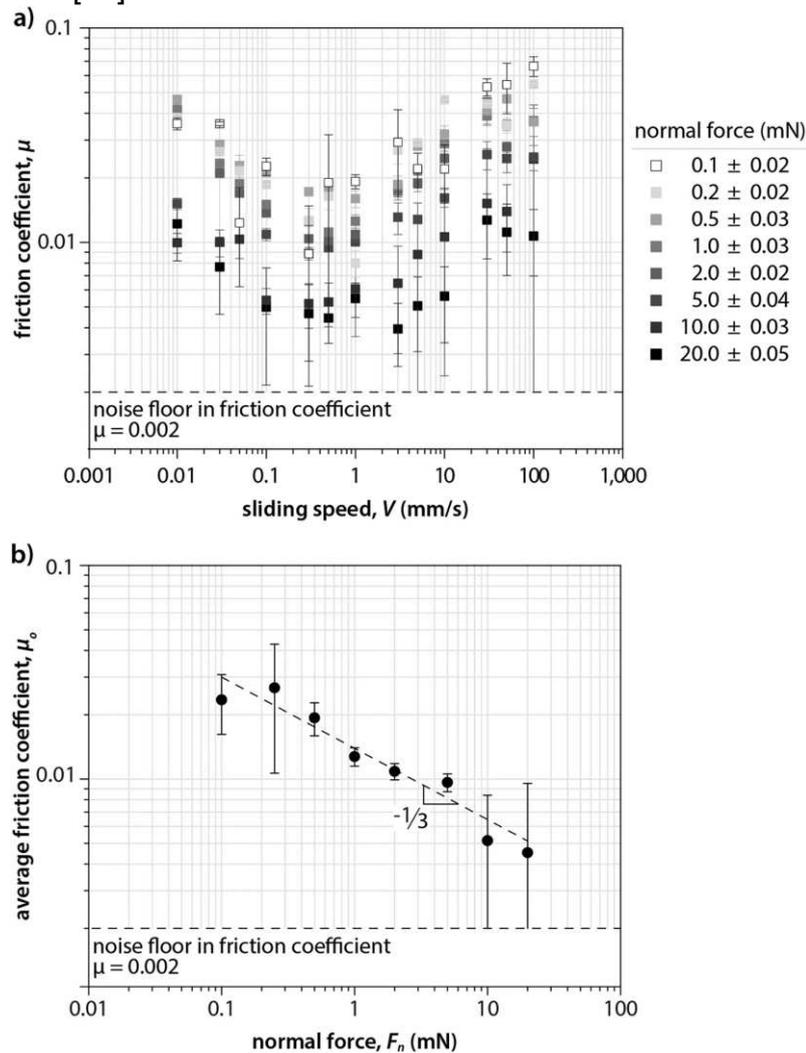


Fig. 2. a) The average friction coefficient, μ , is plotted versus sliding speed for eight different normal loads from 0.1 to 20 mN. The average normal forces and respective standard deviations over each experiment are reported in the adjacent legend. The friction behavior of each normal force experiment displays three distinct lubrication regimes from the lowest to highest sliding speeds tested. At the lowest speeds, the friction coefficient is initially high, then falls with increasing speed to a minimum in the speed-independent regime (generally ~ 0.1 to ~ 5 mm/s), and rises again with increasing sliding speed. The error bars represent the standard deviation over the 20 reciprocating cycles per experiment. b) The average friction coefficient in the speed-independent regime, μ_0 , plotted against normal force, F_n , scales like a $-1/3$ power. The error bars represent the standard deviation in the speed-independent regime.

The effects of normal load on friction have been previously explored for hydrogel substrates against smooth polished spherical glass probes. In 2005, Rennie et al. found that the friction of soft hydrogel contact lenses decreased with increasing load ($\mu \sim F_n^{-1/2}$) when tested against smooth glass probes; the authors suggested that this was due to Winkler-like mechanics of contact area scaling to the square-root of load [41]. Gong et al. performed numerous studies on hydrogels under varying loads and composition, and suggested that the friction coefficient was dictated in part by the surface chemistry [16,42]. All indications from the work of the groups from Archard [39], de Gennes [24], Persson [43], Müser [44], Israelachvili [45], Klein [46], and

Salmeron [47] suggest the potential for a relatively simple hypothesis regarding the scaling of friction for gels: to the first approximation, the friction coefficient will be proportional to the real area of contact. Therefore, in these ultra-smooth and soft hydrogel interfaces where the surface roughness is on the order of the mesh size, the real area of contact is approximately equal to the nominal area of contact determined from the Hertzian contact analysis. Therefore, the expected scaling of friction coefficient with applied load is to the $-1/3$ power.

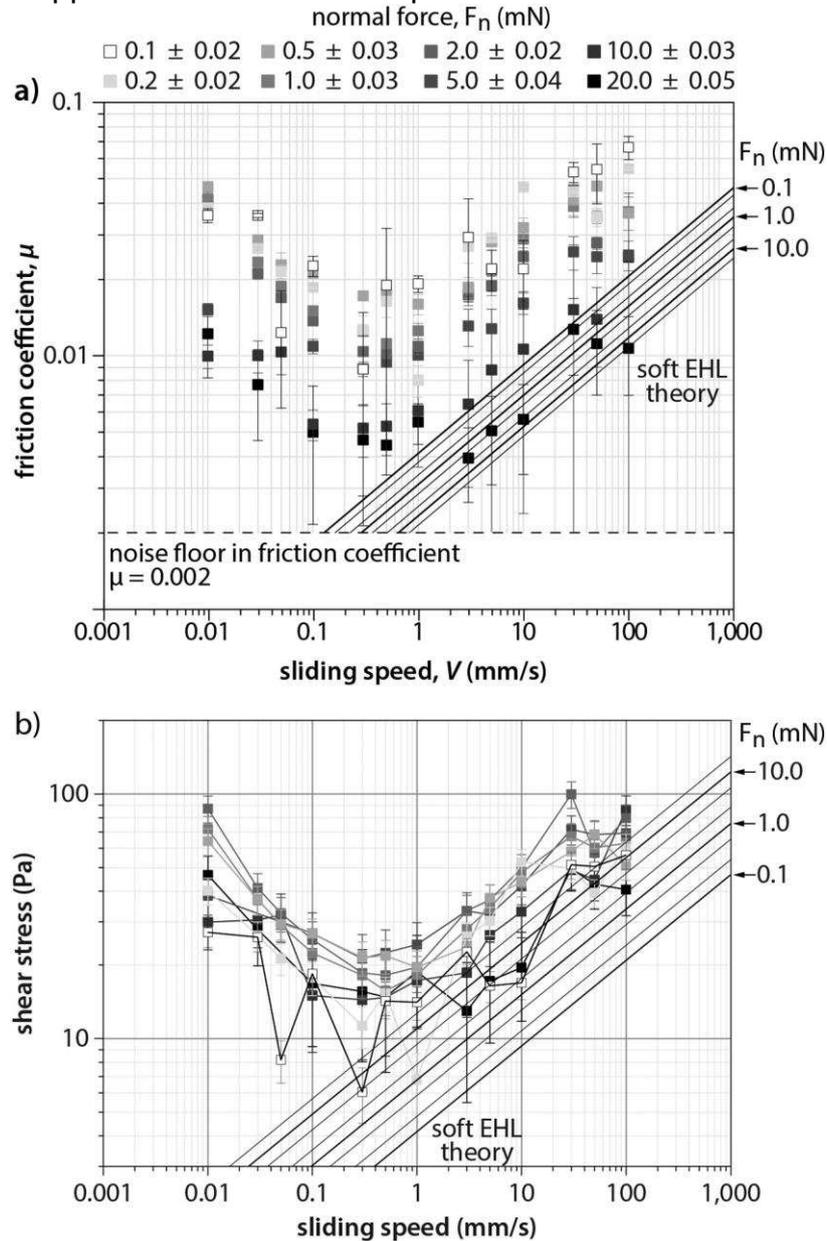


Fig. 3. a) The average friction coefficient, μ , plotted versus sliding speed. Solid lines are the theoretical predictions from soft EHL theory at the corresponding loads and speeds. b) Shear stress, τ , as predicted assuming Hertzian contact mechanics and uniform shear stress is plotted versus sliding speed for eight different normal loads from 0.1 to 20 mN. Solid lines again are based on the soft EHL theory under the corresponding loads and speeds.

As shown in Fig. 1b and c, the Gemini hydrogel configuration is capable of providing very low friction coefficients ($\mu < 0.01$) at low contact pressures and low sliding speeds,

which is in agreement with previous findings [1–3]. For each of the eight normal forces in this study (0.1 to 20 mN), the full range of sliding speeds revealed speed-dependent friction coefficient behavior, which is the subject of previous [3] and ongoing investigations [48]. Following the observations and methods set forth by Dunn et al. [1] and Urueña et al. [3], a speed-independent regime of friction ($v \sim 0.1$ to ~ 5 mm/s) was identified in these data. This regime was used to report the nominal friction coefficient, μ_0 , which enabled comparisons of the scaling behavior of friction as a function of applied load, essentially independent of the effects of sliding speed. It is clear from Fig. 2a that the friction coefficient drops across all ranges of sliding speed with increasing applied load, as would be predicted from considerations of contact area scaling non-linearly with applied load. The average friction coefficients in the speed-independent regime for each normal force experiment are given in Fig. 2b, along with error bars that represent ± 1 standard deviation for the set of cycles performed at each normal load.

The finding that the high water content hydrogels used in this study can be described by Hertzian contact mechanics theory [29] is entirely consistent with the finding that the scaling of friction coefficient in the speed-independent regime follows $\mu \sim F_n^{-1/3}$ (Fig. 2b). One intriguing aspect of this finding is that in the speed-independent regime there appears to be a constant shear stress. Based on the calculations of contact area from Schulze et al. [29] we compute that the shear stress is $\tau = 18.4 \pm 3.5$ Pa for all of these samples. One postulated mechanism for the dissipation in large mesh size hydrogels is that the shear stress arises from the shearing of the solvent within the hydrodynamic penetration depth of the gel, which is the effective depth into the gel that the solvent is shearing as a result of a fluid shear at the gel surface. The viscosity of the solvent within the hydrogel is likely greater than the bulk viscosity of water due to considerations of bound water in the vicinity of polymer chains [49]. Considerations of fluid relaxation times would predict that the solvent viscosity could be an order of magnitude higher than the bulk viscosity. Milner [50] and Klein [51] have argued that to a good approximation, the hydrodynamic penetration depth is on the order of the mesh size. However, such analysis fails to predict a speed-independent zone, and actually predicts that the shear stress should increase with increasing sliding speed. At these sliding speeds ($v = 0.1$ to 10 mm/s) and for a hydrogel mesh size of 7 nm [3], the resulting shear rates are order $\dot{\gamma} = 10^4$ – 10^6 s $^{-1}$ with resulting shear stress (assuming the viscosity of water) of $\tau = 10$ to 1000 Pa. The measured shear stresses from these experiments are generally lower than the predicted shear stress from the hydrodynamic penetration over the majority of this sliding regime. One potential hypothesis is that there is a fluid film thickness that is significantly larger than the hydrodynamic penetration depth and shearing is occurring across this fluid film. Following the soft Elasto-Hydrodynamic Lubrication (EHL) models of Hamrock and Dowson [41,52], a computation for the friction coefficient and shear stress as a function of applied load and sliding speed are shown as solid lines in Fig. 3a and b, respectively, based on soft EHL calculations of fluid film thicknesses reported in the Supplementary Material. In Fig. 3a, the weak dependence of friction coefficient on load for EHL is clearly seen by the crowded parallel lines of friction coefficient. The measured friction coefficient data is spread over a much wider range, and the scaling with load at the higher speeds is stronger than would be predicted by soft EHL considerations, and at the lower speeds actually flat or reversed. In Fig. 3b, the experiments varying load and sliding speed collapse into a regime of strongly

overlapping uncertainty intervals over the entire range of experimental conditions. This collapse is counter to the trend that would be predicted by soft EHL considerations, which would further spread these data and predict the greatest shear stresses for the highest loads. Taken together the experiments and models suggest that the friction is lower than would be predicted from the shearing of a fluid through a single mesh, yet greater than the predictions from soft EHL considerations.

Qualitatively, these gels appear to behave like low friction polymers with a constant value of shear stress across the interface in the speed-independent regime. The estimation of a constant value of shear stress of $\tau=18.4 \pm 3.5$ Pa, adds a new and additional clue to investigations aimed at unlocking the physics of hydrogel tribology. It would be interesting to study the dependence of this shear stress on the mesh size. For these gels, the ratio of the computed shear stress to the osmotic pressure (or elastic modulus) is approximately 1%. This ratio, while outside of the range of viscous shearing for a hydrodynamic penetration depth on the order of the mesh size, is well within the range that one would expect for repeated shear strains in polymer chains to viscoelastically dissipate energy, similar to friction in elastomers [43,53].

A more complete explanation of the velocity dependence of friction in Gemini hydrogels remains an elusive quest, and an active area of research. The findings of Hertzian contact mechanics and Hertzian scaling of friction coefficient within Gemini hydrogel experiments are an important contribution to these investigations.

4. Concluding Remarks

In these experiments, soft high water content hydrogels were molded and tested in a Gemini configuration over a wide range of speeds and loads. The scaling of friction coefficient was consistent with the elastic theories of Hertz, and predict that there is a constant value of shear stress within the contact of $\tau=18.4 \pm 3.5$ Pa. This shear stress is smaller than the calculated value of fluid shear within a hydrodynamic penetration depth on the order of the mesh size. This finding, along with the observation of a speed-independent friction coefficient over a wide range of sliding speeds, challenges the hypothesis that the origin of dissipation in these gels is defined by isoviscous shear within the surface gel layer or fluid film lubrication.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.biotri.2018.01.002>.

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