Adsorption-induced transient friction of hydrogels on hydrophilic countersurfaces

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ABSTRACT
Soft hydrated permeable surfaces of hydrogels exhibit unique lubrication behaviors, including frictional hysteresis found in tribo-rheometry measurements. A hydrogel lubrication model that describes the transient behavior was previously developed using the structure kinetics model in the field of rheology and rate-and-state friction model, where the friction change is described as a competition between buildup and breakdown rates. In this study, the model is further modified to include the effect of hydrophilicity of a countersurface. Ultraviolet (UV)/ozone treatment on an aluminum surface significantly removes organic materials, resulting in extremely hydrophilic surface. Friction response of a polyacrylamide hydrogel against untreated and UV/ozone-treated aluminum exhibited noteworthy difference in the trajectory of hysteresis. Model fits were conducted using the modified lubrication model on both hystereses, and the fitting parameters of both hystereses are compared with each other to identify a parameter addressing hydrophilicity. Based on the model fits, we suggest that the hydrophilicity of the countersurface initially prevents the adsorption on the hydrogel surface because it holds water better. However, once water goes out of the contact due to contact pressure, a stronger adsorption occurs, which increases friction and decreases the speed dependence of friction.

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I. INTRODUCTION
Due to its high water absorption, permeability, softness and flexibility, biocompatibility, and biodegradability, hydrogel is utilized in various fields, such as contact lenses, flexible electronics, tissue engineering, wound dressings, drug delivery, cosmetic industries, and agriculture engineering. For those applications of hydrogel, hydrogel experiences a number of situations regarding lubrication at the interface. Of particular importance in applications of hydrogel is, therefore, the lubricating performance of hydrogel surfaces and interfaces. Due to the importance of hydrogel lubrication, research efforts have intensively focused on understanding a lubrication curve that shows the relationship between friction and speed for soft hydrated gels in sliding by using several mechanics and models. Among those approaches for understanding hydrogel lubrication, the repulsion-adsorption model designed by Gong and co-workers is a representative model, which is based on Schallamach’s approach for rubber friction. In their studies, the model fits were performed only for the lubrication curve after reaching steady state, even though they reported the existence of transient response. However, it is frequent to experience transient response of lubrication in practical applications, and so it is required to handle transient behavior of hydrogel lubrication in model regarding hydrogel lubrication.

To understand the transient lubrication, Kim and Dunn deeply investigated the lubrication behavior of hydrogel on sliding interface. First, Kim and Dunn designed tribo-rheological experiments to understand the lubrication behaviors of hydrogel. They, for the first time, reported resemblance between the duration-dependent response of sliding interface and the stress response of thixotropic material to the stepwise reduction in the shear rate. Then, based on the time-dependent behavior of a hydrogel–aluminum interface, they...
established a model for hydrogel lubrication by using structure kinetics model in the field of thixotropy.\textsuperscript{25,26} The model suggested by those researchers agrees well with the lubrication hysteresis loops of the hydrogel–aluminum interface. In their study, it was inferred that the structural changes may be related to surface chain stretch or local dehydration. Furthermore, they expanded the model and suggested a clear physical meaning of structure parameter based on a comparison with the rate-and-state friction model used for geological events.\textsuperscript{27} In this study, the structure parameter is interpreted as the amount of adsorption sites, which is controlled by the amount of water at interface.

The complex fluid lubrication model, which is the name for the final version of model designed by Kim and Dunn, can provide a plausible explanation for the transient lubrication of hydrogel at sliding interface with a clear physical interpretation. However, the degree of hydrophilicity of the countersurface has not been considered in the model. Hydrogel mostly comprises water and water intervention is always accompanied during the lubrication. Furthermore, since the model postulates that the source of friction is attributed to the adsorption of the hydrogel surface to the countersurface, the degree of hydrophilicity of the countersurface is a major determinant of frictional response. For this reason, it is required to consider how hydrophilic the countersurface is for better understanding of hydrogel lubrication and mimicking the lubrication in practical applications. To this end, we aim to delve into the frictional hysteresis loops of hydrogel on more hydrophilic countersurface, which is treated by ultraviolet (UV)/ozone cleaner and modify the existing complex fluid lubrication model for better prediction of torque response measured on the hydrophilic surface.

The surface characterization based on contact angle and x-ray photoelectron spectroscopy is displayed in Sec. III A to verify an increase in the degree of hydrophilicity after the surface treatment and to understand the corresponding surface composition and chemistry. To show how the hydrogel lubrication changes after the UV/ozone treatment, the frictional hysteresis loops with five different step times measured on the UV/ozone-treated aluminum surface are visualized and their unique characteristics are enumerated in Sec. III B. Then, in Sec. IV, based on these characteristic visual aspects of hysteresis loops generated from the hydrogel on the UV/ozone-treated aluminum surface, the existing lubrication model is modified by introducing the concept regarding the separation of water layer at interface. In Sec. V, physical interpretation of the difference between hysteresis loops of the untreated and UV/ozone-treated countersurfaces is depicted.

II. MATERIALS AND METHODS

A. Hydrogel

Polyacrylamide hydrogel is used for all lubrication experiments. The hydrogel is synthesized by simultaneous polymerization and cross-linking using radical chain reactions. The synthesized hydrogel is prepared from a mixture of acrylamide (monomer; 7.5 wt. %), N,N\textsubscript{0}-methylenebisacrylamide (crosslinker), ammonium persulfate (radical initiator; 0.15 wt. %), N,N,N,N\textsubscript{0}-tetramethylethylenediamine (catalyst; 0.15 wt. %), and de-ionized water (solvent; ~92 wt. %). The mixture of hydrogel prepolymer solution is gently poured in a glass dish with 56 mm inner diameter and 11 mm depth. Then the top surface of the solution is covered with a flat polystyrene mold aligned parallel to the bottom surface of the glass dish. The prepolymer solution undergoes free radical chain reaction for 30 min at room temperature. After the polymerization, the surface mold is removed. The resulting hydrogel sample is a circular cylindrical slab with thickness of 8 mm and diameter of 56 mm fixed inside the glass. The remaining height of 3 mm is used to surround and hold water on top of the hydrogel.

B. Tribo-rheometry

All tribological measurements were performed on DHR-3 stress-controlled rheometer (TA instruments) at 15 °C by using parallel plate geometry (hard anodized aluminum) with a diameter of 40 mm (Part No. 513400.905). To provide a sliding interface with annular contact area that experiences an effectively uniform slip velocity, the aluminum annulus with 40 mm of outer diameter and 2 mm of wall thickness (see Fig. 2 in Kim and Dunn\textsuperscript{22}) was adhered to the top geometry. By adjusting gap height, aluminum–hydrogel contact with a normal force of 1.0 N was established before the rotational experiments; the gap height remained fixed during rotation. The experimental protocol of flow curve was applied to the hydrogel to measure frictional hysteresis loops. Each flow curve consists of 15 logarithmic steps from 0.05 to 50 rad/s, and an upward curve is followed by a downward curve. A pair of torque curves including an upward curve (upper curve) followed by a downward curve (lower curve) is considered as one cycle. Three cycles of experiments were performed for each step time. The hysteresis loops with step time of 9.0, 16.0, 28.5, 50.6, and 90 s are measured. Frictional torque under the sliding input was collected via the TRIOS software (TA Instruments). Each experiment was repeated three times and those are qualitatively identical.

C. Surface treatment and characterization

A UV/ozone cleaner (AH1700, AHTECH LTS) was used for the surface treatment of aluminum surface. The system is equipped with a low-pressure mercury vapor grid lamp capable of providing an ozone intensity of 28 mW/cm\textsuperscript{2}. The aluminum surfaces were processed under stable UV light irradiation with wavelengths of 184 and 254 nm for 10 min. To evaluate hydrophilicity, the static contact angle was measured by the touch automatic contact angle analyzer (Phoexnix 300 Touch, SEO) immediately after the surface modification processes. A droplet of de-ionized water was placed on the surfaces, and the droplet shape was captured with a CCD camera. The average contact angle of the five measurements was recorded. The surface chemical composition was investigated using x-ray photoelectron spectroscopy (Multilab 2000, Thermo Fisher) with an Al K\textsubscript{z} (1486.6 eV) x-ray source on a 200 μm\textsuperscript{2} point area.

III. EXPERIMENTAL RESULTS

A. Surface characterization

The purpose of surface treatment with the UV/ozone cleaner is to investigate the difference in the lubrication behavior of hydrogel with respect to the hydrophilicity of countersurface. It is, therefore, required to measure water contact angles on both untreated and UV/ozone-treated aluminum surfaces for quantifying the hydrophilicity. The water contact angles on untreated and UV/ozone-treated countersurfaces are shown in Fig. 1.

The water contact angle on untreated aluminum surface was 74.2° and had significantly decreased to 36.80° after the UV/ozone treatment. The decrease in the water contact angle on the aluminum
surface after the UV/ozone treatment demonstrates significant increase in the degree of hydrophilicity.

Changes in surface compositions and chemistry after the UV/ozone treatment provide clues for significant changes in the degree of hydrophilicity. So, we examined the surface chemical composition of untreated and UV/ozone-treated aluminum surfaces by x-ray photoelectron spectroscopy (XPS), as shown in Fig. 2.

As shown in the survey spectrum in Fig. 2(a), there exists a significant decrease in C1s peak after the UV/ozone-treated aluminum surface. Atomic percentage of aluminum, carbon, and oxygen are 29.66%, 25.98%, and 44.36% before the surface treatment, respectively. After the UV/ozone-treatment, the atomic percentage of each element is changed to 36.71%, 8.11%, and 55.18%, respectively. When aluminum and oxygen are exclusively considered in atomic percentage, there is no change even after the surface treatment. It is possible to infer that the UV/ozone treatment effectively reduces the carbon content on aluminum surfaces. Interestingly, the intensities of O1s and Al2p are almost identical in the survey. In Al2p spectra, the Al metal and Al2O3 peaks are observed at 72.3 and 75.1 eV, respectively.28,29 Also, high resolution XPS core level Al2p and O1s spectra of the UV/ozone-treated surface mirror those of the untreated aluminum. Specifically, the integrated areas in Al2p spectra after deconvolution for aluminum metal and aluminum oxide are calculated as follows: 24.36% and 75.64% before the treatment and 24.81% and 75.19% after the treatment. With regard to C1s XPS spectra, a remarkable decrease in peak regarding C–C or C=C bond is observed; in that the C1s spectra consist of three peaks at 284.2, 285.7, and 288.3 eV, corresponding to C–C or C=C, C–O, and COOH, respectively.30,31 Taken together, UV/ozone functions as the removal of organic materials accumulated on the untreated aluminum surface. The contact angle returns to untreated state after few days, which indicates the accumulation of organic materials, such as volatile organic compounds (VOCs), from air. By eliminating the hydrophobic organic materials including C–C or C=C bond, it is possible to generate more hydrophilic surface via UV/ozone treatment.
B. Lubrication behavior of hydrogel on the UV/ozone-treated aluminum surface

The UV/ozone-treated aluminum surface is more hydrophilic than the untreated aluminum surface. It is, thus, expected that the UV/ozone-treated aluminum surface exhibits quite different interaction with hydrogel in comparison with the untreated aluminum surface, which is directly linked to the lubrication behavior. To deeply investigate how the degree of hydrophilicity of countersurface affects hydrogel lubrication, the torque responses with respect to angular speed are measured on both untreated and UV/ozone-treated aluminum surfaces. For the UV/ozone treated surface, all the lubrication experiments are performed immediately after the treatment. Experimental protocol of the transient flow curve is used with five different step times. ("Transient flow curve" is defined as stress responses from shear startup tests with different rates visualized in stress/rate space before reaching the steady state.") At the third cycle of lubrication, torque curve converges to the representative shape for each step time. So, torque curves on the untreated and UV/ozone-treated aluminum surfaces at third cycle are visualized in Figs. 3(a) and 3(b), respectively.

Frictional hysteresis loops on the untreated aluminum surface are adopted from the previous study (see Fig. 3 in the study of Kim and Dunn"). Friction responses of a polyacrylamide hydrogel against the untreated and UV/ozone-treated aluminum surfaces exhibit noteworthy difference in the trajectory of hysteresis as shown in Figs. 3(a) and 3(b). First, hydrogel on the UV/ozone-treated aluminum surface shows higher torques at low speed regime than hydrogel on the untreated surface. Second, the increasing rate of friction with angular speed is relatively low for the UV/ozone-treated surface than the untreated one under low angular speed. Third, there exists two slopes at the high speed regime of the downward torque curves on the UV/ozone-treated aluminum surface, while the downward torque curve on the untreated aluminum surface at high speed regime consists of only one slope. The physical interpretation for these unique features of the torque curve from the UV/ozone-treated surface will be discussed in Sec. V.

IV. MODIFICATION OF COMPLEX FLUID LUBRICATION MODEL

In this section, we modify the complex lubrication model suggested by Kim and Dunn. They adopted the structure kinetics model, which is the most prevailing model in the field of thixotropy, and tailored it for describing hydrogel lubrication based on time-dependent structure parameter, . The complex fluid lubrication model comprises constitutive equation of shear stress and kinetic equation of structure parameter. First, shear stress is described as the power law relation between friction and sliding speed as a Newtonian fluid flow with changing gap height, shown as follows:

\[
\sigma = \eta(\lambda) \gamma = \eta(\lambda) \frac{V}{h}\quad (1)
\]

where \(\eta(\lambda) = \eta_s + \lambda \eta_p\) and \(h(V) = CV^{1-a}\). In Eq. (1), \(\sigma\) is the fluid shear stress under sliding, \(\eta_s\) is the water viscosity, \(\eta_p\) is the viscous coefficient of the structure parameter, \(\gamma\) is the shear rate, \(V\) is the linear sliding speed, \(h\) is an effective gap height, \(a\) is the power law exponent found in lubrication curves at the high speed regime, and \(C\) is the proportional constant.

In structure kinetics models for thixotropy, the change in structure is described by breakdown and buildup of structure. Similarly, the complex lubrication model includes breakdown of contact area by sliding and buildup of adsorption sites by contact pressure. The evolution equation for structure parameter is given as

\[
\frac{d\lambda}{dt} = -k_1 \frac{V}{V_{max}} \lambda^a + k_2 (1 - \lambda)^b\quad (2)
\]

where \(k_1\) and \(k_2\) are the rate coefficients, \(a\) and \(b\) are power law exponents that determine the dependence of each rate term to \(\lambda\), and \(V/V_{max}\) is a normalized sliding speed.

In Sec. III, we portrayed the frictional hysteresis loops on the UV/ozone-treated aluminum surface and enumerated the unique features in comparison with the hysteresis loops on the untreated countersurface. For better model fits, we modify the existing lubrication model by focusing on those distinctive aspects of lubrication hysteresis on the
UV/ozone-treated aluminum surface. First, the power law exponents for sliding speed in fluid shear stress and frictional stress in Eq. (1) are separated as shown in the following equation, because the slope at low angular speed is significantly different with the slope in high speed regime:

\[
\sigma = \sigma_f + \sigma_p = (1 - \lambda)\eta \frac{V}{h(V)} + \lambda KV^\beta,
\]  

(3)

where \(\sigma_f\) is the fluid shear stress, \(\sigma_p\) is the frictional stress, \(\beta\) is the power law exponent found in lubrication curves at the low speed regime, and \(K\) is a proportional constant for frictional stress. The frictional stress term is proportional to structure parameter \(\lambda\), because friction of polyurethane-aluminum interface is affected by the degree of direct contact. The fluid shear stress term is proportional to \(1 - \lambda\) because fluid flow is expected to mostly occur on the area without contact, even though it is possible for water to flow through the contact region due to the water-polymer gradient existing on the surface of hydrogels.\(^{38,39}\) When \(\lambda\) approaches 1, the degree of direct contact of hydrogel aluminum interface increases, and therefore, the total stress becomes more dependent on the frictional stress. On the contrary, if \(\lambda\) is close to 0, then the total stress is more influenced by the fluid shear stress term because there is less direct contact due to a sufficient layer of water.

Second, downward torque curves formed by hydrogel lubrication on the UV/ozone-treated aluminum surface show two slopes at a high speed regime. It is, therefore, assumed that there exists an adjacent water layer on the aluminum surface held by stronger force created from higher hydrophilicity. We, therefore, introduce a minimum threshold of the lubrication film thickness as

\[
h(V) = \begin{cases} 
CV^{1-x} & (CV^{1-x} \geq h_0), \\
h_0 & (CV^{1-x} < h_0), 
\end{cases}
\]

(4)

where \(h_0\) is the thickness of the adjacent water layer on the aluminum surface. The same form of evolution equation of structure parameter shown in Eq. (2) is used in the modified model. The shear stress is converted to frictional torque as follows:

\[
T = \pi\left(\frac{r_o^2}{2} - r_i^2\right)\frac{r_o + r_i}{2} \sigma,
\]

(5)

where \(r_o\) and \(r_i\) are outer and inner radii of the annulus, respectively.

By using the modified lubrication model shown in Eqs. (2), (3), (4), and (5), we performed model fits to the hysteresis loops using MATLAB. In every iteration of model fits using lsqcurvefit function, Ode45, a function based on an explicit Runge–Kutta method, is used to solve the differential equation shown in Eq. (2). Parameters corresponding to the smallest residual sum of squares (RSS) were selected among the sets of parameters starting from various initial values selected by Latin hypercube sampling from a range spanning 1000-fold difference to select global optimum in the range.

The lubrication hysteresis loops on untreated and UV/ozone-treated aluminum surfaces and their corresponding fitted curves from the modified lubrication model are displayed in Fig. 4. The fitted curves from the modified model agree well with the experimental data for both hysteresis loops. The fitting parameters for the modified lubrication model are tabulated in Table I.

V. DISCUSSION AND CONCLUSION

Kim and Dunn\(^{27}\) suggested the physical interpretation for the structure parameter in the complex fluid lubrication model as the degree of the direct contact between the polymer and hard surfaces (see Fig. 5 of Kim and Dunn\(^{27}\)). The degree of the direct contact is determined by the amount of water at the interface. The formation of thick water layer with water lubrication occurs under the high sliding speed, leading to the prevention of direct contact. On the other hand, the contact area increases under low sliding speed with the water drainage at the interface due to the compression. The grown contact area induces the macroscopic adhesion. It is, therefore, important to understand how water intervention affects the friction response of hydrogel–aluminum interface.

For the physical interpretation of lubrication behavior on the UV/ozone-treated aluminum surface, we select the frictional hysteresis loop with 90 s of step time as representative shape. For comparison between lubrication behavior on untreated and UV/ozone-treated aluminum surfaces, frictional hysteresis loops and related schematics are visualized in Figs. 5(a) and 5(b), respectively.

Based on the complex fluid lubrication model, the transition from increase to decrease in torque in upward curve is considered as rapid introduction of water and the transition from decrease to increase in torque in downward curve is considered as the start of direct contact between the two surfaces. From this point of view, the downward curve in Fig. 5(b) consists of two slopes before the start of direct contact, while the downward curve of the untreated surface shown in Fig. 5(a) presents only one slope in this regime. The slope of downward curve at high speed regime in Fig. 5(a) is identical to that of the UV/ozone-treated surface in Fig. 5(b), and so they are colored as green dashed lines. Then, the two slopes in Fig. 5(b) are denoted as green and red dashed lines. Unlike the untreated surface, the UV/Ozone treated surface can resist squeeze flow of water at lower sliding speeds due to hydrophilicity. The remaining excess water at lower speeds can effectively maintain gap height, and we consider this as an adjacent water layer on the treated aluminum surface. The thickness of the adjacent water layer represents the degree of hydrophilicity and determines lower limit of lubrication film thickness. Since the thickness of the layer is negligible for the untreated surface, the single slope of downward curve at high speed regime in Fig. 5(a) is observed. The two slopes in Fig. 5(b) correspond to two states of water lubrication. The schematics for water at the interface for both stages of lubrication are illustrated in Fig. 5(b). The adjacent water layer is denoted as dark blue and the rest of water in the lubrication film is colored with light blue. When lubrication film is thicker than the adjacent water layer, the torque response is identical to the torque curve of hydrogel on the untreated aluminum surface since water is abundant. The slope is changed when the hydrogel begins to contact the adjacent water layer, and this is the point when hydrophilicity of countersurface starts to affect the lubrication behavior and maintains the effective lubrication film thickness.

As mentioned in Sec. III, one of most remarkable features in the frictional hysteresis loop of hydrogel on the UV/ozone-treated aluminum surface is relatively high torque values at the low speed regime, which are colored as orange in Figs. 5(a) and 5(b). As shown in contact angles in Fig. 1, the UV/ozone-treated aluminum surface is more hydrophilic than the untreated one. It is therefore required to exert higher torque on the hydrogel to reach an applied sliding speed in low
sliding speed regime, where the direct contact between hydrogel and the UV/ozone-treated aluminum occurs. Once water is removed from the contact due to the contact pressure, a stronger adsorption occurs between hydrogel and the UV/ozone-treated aluminum, which increases friction in comparison with the untreated countersurface.

In addition, the increasing rate of torque with angular speed for the UV/ozone-treated aluminum is less than the rate in the torque curve measured with the untreated aluminum in the regime of direct contact. The slopes of upward curves in this regime shown in Figs. 5(a) and 5(b) are denoted as orange dashed lines, and the slope of upward curve in Fig. 5(b) is gentler than the slope of upward curve in Fig. 5(a) measured with the untreated aluminum surface. The gentler slope in this regime after the surface treatment is due to the stronger instantaneous adsorption, or higher adsorption energy, between hydrogel and the UV/ozone-treated surface originated from the higher hydrophilicity. Based on the adsorption friction model proposed by Gong and Osada, the hydrogel friction against a countersurface is determined by instantaneous percentage of adsorbed polymer chains with respect to the total surface chains per unit area. The adsorbed chains stretch elastically, which causes resistance to sling or friction. In the model, the desorption of chains is described by the Polanyi–Wigner equation, which shows a simple kinetics of desorption with a rate constant derived from the Arrhenius equation. Difference between adsorption energy and elastic energy of a stretched chain is used as an activation energy in the Arrhenius equation. For a stronger adsorption, the adsorption energy will be higher. The desorption kinetics determines average bonding time and viscoelastic recovery of a single chain determines time in a free state. The two characteristic times are used to estimate the average number of adsorbed chains in unit area, which is directly proportional to frictional stress. The simulation

<table>
<thead>
<tr>
<th>Parameter</th>
<th>(k_1)</th>
<th>(k_2)</th>
<th>(a)</th>
<th>(b)</th>
<th>(C (m^{0.7} s^{0.5}))</th>
<th>(K (Pa \cdot s^b / m^d))</th>
<th>(\alpha)</th>
<th>(\beta)</th>
<th>(h_0 (nm))</th>
</tr>
</thead>
<tbody>
<tr>
<td>Untreated</td>
<td>0.0102</td>
<td>0.000747</td>
<td>0.0176</td>
<td>4.75</td>
<td>(2.33 \times 10^{-7})</td>
<td>64545</td>
<td>0.70</td>
<td>0.578</td>
<td>30</td>
</tr>
<tr>
<td>UV/ozone</td>
<td>0.0193</td>
<td>1.34 \times 10^{-3}</td>
<td>0.0176</td>
<td>4.75</td>
<td>(2.33 \times 10^{-7})</td>
<td>11039</td>
<td>0.70</td>
<td>0.147</td>
<td>157</td>
</tr>
</tbody>
</table>
result based on this model shows a decrease in speed dependence of friction at higher adsorption energy in the speed regime correspond to low speed regime in the current study.\textsuperscript{13}

In the future work, the hydrogel lubrication model can be further improved by considering the permeability of the surface of hydrogel. In the current study, the fluid shear stress is modeled with no-slip boundary condition. However, since the surface of hydrogel is porous and brushy and has polymer density gradient,\textsuperscript{38,39} fluid flow can occur through the subsurface of hydrogel. Therefore, if the farthest boundary from the bulk of hydrogel is considered a nominal surface, then the slip-flow boundary condition can be applied.\textsuperscript{40,41} With this boundary condition, the effective gap height $h$ with power law speed dependence can be modeled with better physical interpretation considering penetration depth of the flow. Furthermore, the permeation through the subsurface can be another origin of time dependence, and therefore, some of the complicated features of lubrication hysteresis curves can be more precisely predicted.

In conclusion, this study expands the understanding of complex fluid lubrication model by addressing the hydrophilicity of countersurface. When the countersurface for hydrogel lubrication is significantly hydrophilic, it is considered that there exists the adjacent water layer near the countersurface strongly held by the hydrophilic surface. This concept of separating water layer at interface is reflected in the modified lubrication model, and so the transient torque responses are successfully predicted by model fits. The enhanced model and insight will help in designing a hydrogel and material for countersurface for better performance in practical applications. Moreover, we expect that a more general model for hydrogel lubrication will be available in the future with this study as a cornerstone.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Ethics Approval

This material has not been published in whole or in part elsewhere. The manuscript is not currently being considered for publication in another journal. All authors have been personally and actively involved in substantive work leading to the manuscript, and will hold themselves jointly and individually responsible for its content.

Author Contributions

Jiho Choi and Kwangmo Yang contributed equally to this work.

Jiho Choi: Conceptualization (lead); Data curation (lead); Formal analysis (supporting); Investigation (equal); Validation (equal); Writing – original draft (lead). Kwangmo Yang: Conceptualization (equal); Data curation (equal); Formal analysis (lead); Investigation (lead); Validation (lead); Writing – original draft (supporting). Youn-Ki Lee: Investigation (supporting). Sungho Lee: Writing – review & editing (supporting). Kunsik An: Writing – review & editing (supporting). Sung-Soo Kim: Conceptualization (equal); Supervision (equal); Validation (equal); Writing – review & editing (equal). Jiho Kim: Conceptualization (equal); Data curation (equal); Investigation (equal); Supervision (equal); Validation (equal); Writing – review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding authors upon reasonable request.

REFERENCES


