Supporting Information

Human ability to discriminate surface chemistry by touch

Cody W. Carpenter,[a,1] Charles Dhong,[a,1] Nicholas B. Root,[b,1] Daniel Rodriquez,[a] Emily E. Abdo,[a] Kyle Skelil,[a] Mohammad A. Alkhadra,[a] Julian Ramírez,[a] Vilayanur S. Ramachandran,[b] and Darren J. Lipomi[a]*

[a]*Department of NanoEngineering, University of California, San Diego*
9500 Gilman Drive, Mail Code 0448, La Jolla, CA 92093-0448

[b]*Department of Psychology, University of California, San Diego*
9500 Gilman Drive, Mail Code 0109, La Jolla, CA 92093-0109

[1]These authors contributed equally to this work

*Author to whom correspondence should be addressed: dlipomi@eng.ucsd.edu

Methods

Participants.

The study was approved by the Institutional Review Board of the University of California, San Diego in accordance with the requirements of the Code of Federal Regulations on the Protection of Human Subjects (45 CFR 46 and 21 CFR 50 and 56), Project #170248S. Data were collected from a total of 23 healthy volunteers between the ages of 18 and 40.

Materials.

Both SiOH and FOTS samples were prepared using 0.625 mm thick (100) Si wafers (University Wafer). Sample dimensions for each psychophysical experiment: free exploration (2.54 cm × 7.62 cm); tapping only (2.54 cm × 2.54 cm); and ASCII alphabet studies (2 cm × 8 cm). Samples were washed in a 3-step process: 10 min sonication in water followed by compressed air drying; 10 min sonication in acetone followed by compressed air drying; and 10 min sonication in isopropyl alcohol followed by compressed air drying. Surfaces were activated using a plasma cleaner (Harrick, PDC-001) at 200 mTorr at 30 W for 5 min. Passivated samples were prepared using chemical vapor deposition (CVD) of trichloro(1H,1H,2H,2H-perfluorooctyl)silane (FOTS) purchased from Sigma-Aldrich. Following CVD surfaces were rinsed with isopropyl alcohol followed by DI water to remove unbound FOTS species and finally air dried with compressed air.

Static Water Contact Angle.

Static contact angle measurements of 2 μL DI water droplets on SiOH and FOTS surfaces were taken using an Automated Goniometer (Ramé-Hart, Model No. 290-U1).

Advancing and Receding Contact Angle.

Measurements of receding (θ_R) and advancing (θ_A) contact angles of DI water droplets (5 μL) on pristine and explored FOTS surfaces were performed to assess the persistence of receding and advancing contact angles after exploration. Explored FOTS surfaces (θ_R = 89.4° and θ_A = 112.5°) did not show an appreciable change compared to pristine FOTS surfaces (θ_R = 91.8° and θ_A = 114.7°). Previously explored samples were cleaned using isopropanol and a cleanroom wipe, followed by compressed drying, and a dry cleanroom wipe to remove finger residue. Measurements were taken with an Automated Goniometer (Ramé-Hart, Model No. 290-U1) using the add/ remove volume method.

ASCII Alphabet Sample Preparation.

Patterning was achieved by selective plasma treatment of FOTS slides using 1 × 2 cm PDMS blocks placed in conformal contact with slides to protect FOTS regions. Conformal contact was made by placing PDMS covered slides under house vacuum for 5 min. Surface activation of uncovered regions was performed using plasma treatment protocol above.

Finger Pad Moisture Measurements.

Measurement of skin moisture level were taken from the index finger of each subject's dominant hand using a Digital Moisture Monitor for Skin (Number-One, SK-1v).

Average of Subject Exploration Times.

Subjects were timed during individual free exploration, "odd-man-out," trials (8 per subject per condition, i.e., dry and wet) to track the inter-subject average rate of discriminability. Dry Average (13 subjects) = 49 s, Wet Average (15 subjects) = 32 s, time measurements were not recorded for two subjects in the dry condition. Exploration times ranged from 11 s – 88 s per set of samples.

Behavioral Experiments

Pre-experiment procedure.

Before each behavioral experiment, each subject washed their hands and allowed them to air dry, in order to reduce the likelihood that any residue on the fingers interfered with tactile perception. After being seated, subjects were instructed to put on a blindfold (except in the ASCII

Alphabet experiment) and a pair of noise-cancelling headphones, in order to isolate the sense of touch from visual and auditory feedback.

Experimental procedure.

Free exploration, dry condition. Before the experiment began, skin moisture readings were taken using Digital Moisture Monitor for Skin (Number-One, SK-1v). At this point, the experiment began. In each trial, the subject was instructed to explore three prepared samples freely, for as long as they desired. If necessary, the subject could request that the researcher guide their hands to the samples. The subject's task was to identify which sample was unlike the other two: the "odd-man-out" test[1]. Each subject repeated this for a total of eight trials. For each trial, the identity (SiOH vs. FOTS) and location (left vs. middle vs. right) of the unlike sample was randomized.

Free exploration, wet condition. The procedure was identical to that of the dry condition, except that samples were submerged in deionized water.

Restricted exploration, tapping condition. The procedure was identical to the free exploration in the dry condition, except that subjects were instructed to explore samples using a tapping motion only, and to avoid dragging their finger along the sample.

Safety. Silane monolayers are covalently attached to activated silicon surfaces bearing a native layer of silicon dioxide. To address issues of possible removal of FOTS upon contact with human skin, prepared FOTS surfaces were assiduously rinsed with isopropanol, followed by deionized water, and dried using compressed air to remove any physisorbed material. Freshly prepared monolayer surfaces[2] contain on the order of 10^{14} molecules cm^{-2} and are adhered with an adhesive pressure >1 GPa, assuming a covalent bond force on the order of 1 nN. By way of comparison, we estimate the maximum pressure exerted by a human subject on the surface to be

≤10 kPa (≤100 g / cm^2). To measure possible removal of FOTS molecules, we measured the advancing and receding contact angles (methods above) before (θ_R = 91.8° and θ_A = 114.7°) and after (θ_R = 89.4° and θ_A = 112.5°) 50 swipes with approximately 10 - 40 g of applied mass and at 20 - 50 mm/s sliding velocity. Before measuring post exploration contact angles, previously explored samples were cleaned using isopropanol and a cleanroom wipe, followed by compressed drying, and a dry lint-free wipe to remove fingerprints. As a control, receding and advancing contact angles of untouched samples—exposed to the same cleaning procedures as touched samples—were taken before (θ_R = 93.4° and θ_A = 116.2°) after cleaning (θ_R = 90.0° and θ_A = 113.1°). A similar reduction in advancing and receding contact angles suggests that what FOTS is removed during the experiment is removed by mechanical abrasion of the surface by the wipes to remove fingerprints. We thus conclude that a negligible fraction of FOTS molecules are removed during exploration by the light touch used by the subjects during the psychophysical experiments. As an additional safety precaution, subjects washed their hands thoroughly after the psychophysical experiments. Hydrocarbon surface coatings are regularly used in non-stick pans (polytetrafluoroethylene, PTFE) and pose minimal risk of exposure by tactile contact.

Analysis.

Sensory discrimination performance: sensitivity index. For each subject, we used the method described by Craven[3] to calculate the sensitivity index d', a measure of behavioral performance commonly used in sensory discrimination experiments[4]. All reported P-values are two-tailed. When d-prime (d') is significantly greater than zero, this indicates that the subject can discriminate between two stimulus classes ($d' = 0$ corresponds to chance performance; in our odd-man-out, 3 alternative-forced-choice experiment, this is 33% accuracy). The statistical difference between the actual d-prime values and the expected d-prime values under the null hypothesis (d' = 0) can be calculated using a one-sample t-test. In all three experiments, subjects identified the odd-man-out significantly more often than predicted by chance (Table S1).

Sensory discrimination performance: generalized mixed model. Generalized mixed models (GMMs) enable repeated measures data with a categorical response (e.g., correct/incorrect responses, as in the present study) to be modeled, using a logit link function[5]. All reported P-values are two-tailed. A GMM is more powerful than the sensitivity index analyses above, because t-tests on d-prime are only sensitive to subjects' average accuracy, whereas GMMs accounts for subjects' performance on each trial (i.e., the number of trials matters). For each experiment (dry, wet, tapping), we fit a GMM, with trial outcome as the dependent variable, subject as random effect, and a logit link function. These GMMs yield evidence of above-chance discrimination if the intercept coefficient of the model, $\hat{\beta}_0$, is significantly greater than $\beta_{0,0}$ = logit(1/3) = -0.69; we used a Wald Z test to compute a significance value for this difference. In all three experiments (dry, wet, tapping), the intercept term was significantly higher (i.e., better performance) than predicted by chance (Table S2). One additional advantage of this analysis is that it is trivial (using the inverse logit function) to transform the confidence interval on the intercept term back to percent, as is plotted in Fig. 1D and 1E in the main text.

Sensory discrimination performance, effect of skin moisture. We fit a GMM with trial outcome as the dependent variable, moisture as fixed effect, subject as random effect, and a logit link function. All reported P-values are two-tailed. The coefficient of the moisture fixed effect was trending lower than zero ($\hat{\beta}_{Moisture}$= -0.083, se($\hat{\beta}_{Moisture}$) = 0.045, Wald Z = -1.83, P = 0.067, 95% confidence interval [-0.17, 0.0058]), suggesting that subjects with higher skin moisture might perform worse on the discrimination task. That this is trending (rather than significant) does not affect our conclusion that subjects successfully ($P < 0.0001$) discriminated the surfaces; rather, it motivated running an additional experiment (the "wet" condition) to ensure that skin moisture was not a confounding factor in our interpretation of this result.

Sensory discrimination performance, wet vs. dry experiments. To determine whether accuracy was higher in the wet than in the dry condition, we fit a GMM with trial outcome as the dependent variable, condition as fixed effect, subject as random effect, and a logit link function. All reported P-values are two-tailed. The coefficient of the condition fixed effect was significantly different than zero ($\hat{\beta}_{ConditionWet}$= 0.82, se($\hat{\beta}_{ConditionWet}$) = 0.34, Wald Z = 2.45, P = 0.014, 95% confidence interval [0.16, 1.48]), suggesting that subjects were more accurate in the wet experiment. However, we cannot eliminate the potential confound of a training effect: all subjects in the "wet" experiment had previously experienced the discrimination task (in the "dry" experiment), so the increase in accuracy might plausibly have resulted from practice. However, it is clear that conditions unique to the "dry" experiment were not necessary to perform the discrimination task.

Sensory discrimination performance, tap vs. wet and tap vs. dry. To determine whether accuracy was higher in the tapping than in the free exploration condition, we fit two GMMs with trial outcome as dependent variable, condition as fixed effect, subject as random effect, and a logit link function. All reported P-values are two-tailed. For the tap vs. wet GMM, the coefficient of the condition fixed effect was significantly different than zero ($\hat{\beta}_{ConditionTap}$= -1.56, se($\hat{\beta}_{ConditionTap}$) = 0.38, Wald Z = -4.13, $P < 0.0001$, 95% confidence interval [-2.31, -0.82]), suggesting that subjects were more accurate in the wet condition. For the tap vs. dry GMM, the coefficient of the condition fixed effect was significantly different than zero ($\hat{\beta}_{ConditionTap}$= -0.90,

$se(\hat{\beta}_{ConditionTap})$ = 0.35, Wald Z = -2.58, P = 0.0098, 95% confidence interval [-1.59,-0.22]), suggesting that subjects were more accurate in the dry condition than in the tap condition.

Free exploration – ASCII Alphabet.

Procedure.

Silicon surfaces were patterned with FOTS to have alternating regions of silicon and FOTS to create "molecular-binary-bits," equivalent to 1s and 0s used in a ASCII binary code. In binary coding, each letter is represented by a sequence of eight bits. For example, the letters "A," "n," and "t" can be represented as "01000001," "01101110," and "01110100" to spell "Ant." Regions treated with FOTS were designated a binary value of "0," while un-patterned regions, SiOH, regions were designated a binary value of "1." Subjects were instructed to freely explore 3 patterned surfaces, record the 8 bits from each surface using a paper and pen, and finally asked to spell a word using the letters in order from left to right. Subjects used an ASCII binary code library to translate from binary to alphabetical characters. Subjects were given sunglasses rather than blindfolds to aid in recording bits and referencing the ASCII library. Treated and untreated surfaces did not differ in their visual appearance, but differed in the way residue deposited from touching the surfaces, so sunglasses served to reduce such visual feedback cues.

Analysis.

To determine whether subjects successfully decoded more bits than would be predicted by chance, we ran a binomial test for each subject, in which the number of successfully-decoded bits was compared with the number of bits that would be decoded by chance. All reported P-values are two-tailed. 10/11 subjects successfully decoded more bits than would be predicted by chance (all $P < 0.05$, except the one subject, who decoded only 8 bits, $P = 0.38$).

Experimental Apparatus.

Setup.

We used a pull-test to mimic the free exploration of a finger on FOTS and SiOH-treated surfaces. In place of a finger, we used a 30:1 (~100 kPa) PDMS block, sized 1 × 1 × 5 cm and supported by a 3D printed, PMMA bone. This artificial finger was attached to a load cell (Futek LSB200, $k = 1.38$ kN/m) and the load cell was attached to a linear stage (Newmark model ET-100–11). During the pull-test, the PDMS finger was brought into contact with the same silicon wafer surfaces as in the psychophysical tests (FOTS or SiOH). Then the finger was pulled across a surface at constant drive velocity for a distance of 4 mm and the force was recorded (~50 readings per second, *via* Kiethley 2611b, LabVIEW). This was done for 4 pulls at each clean spot, and repeated for 3 spots. The very first pull was always ignored from analysis, since it always involved the step of bringing a finger into contact with the test surface. We varied the drive velocity¸ v (1 mm/s, 2.5 mm/s, 7.5 mm/s, 10 mm/s) and varied the applied mass, M (0 g, 25 g, 75 g, 100 g) on the finger. The stick-slip motion is possible because the compliance in the load cell allows the finger to oscillate in velocity, relative to the motor[6].

Rationale.

The goal of these experiments was to mimic free exploration in two ways. First, we set the experimental variables to match physiological ranges in applied mass, pull velocity as well as the finger's dimensions and mechanical properties. Controlling for these variables, we believed that any differences in forces between the SiOH and FOTS-coated surfaces would be readily apparent because human subjects were able to consistently tell a difference between SiOH and FOTS-coated surfaces in free exploration. Thus, the experimental system should not be particularly sensitive to the way we conducted the pull-test. In this vein, we did not control the time in-between the three pulls on a single run (which ranged from 1-6 s). We also did not control the real contact area of the finger with the test surface and instead brought the finger to an estimated, apparent contact area of 1×1 cm^2.

Mathematical Model.

We modeled the friction between the model finger and substrate as a one-dimensional rigid block sliding on a smooth surface, where the differences in friction could be linked to SiOH and FOTS. We use the model by Ruina[7], which incorporates a state-variable (θ) to account whether the finger is in a microscopic stick (akin to a situation where the static friction must be overcome) or slip event (akin to using the kinetic friction, once the block has begun to move), or some value in between. The advantage of this model is that we can use the friction parameters of FOTS and SiOH surfaces across multiple testing conditions, which is usually not appropriate using a friction coefficient because it is not a rigorously defined property of a material[8]. More complicated models exist which can account for lubricated surfaces[9], elastic deformation[10] and so forth. The friction coefficient, μ, is assumed to be a function of the velocity (v), the state (θ), critical slip distance (D_c) and the friction parameters, (A and B) as such:

$$\mu = \mu_o + A \ln\left(\frac{v}{v_o}\right) + B \ln\left(\frac{v_o \theta}{D_c}\right) \quad (1)$$

A and B are unique to the material to a first order[11], and sensitive to the measuring conditions to a second order. A and B were experimentally determined by a silicon microcantilever tip on silanized and oxidized silicon wafers. The critical slip distance is the microscopic distance needed to break contact between the two surfaces[6] and is a function of the materials and topography of both surfaces – thus, we assume that D_c is constant between both SiOH and FOTS and constant at all testing velocities and applied masses.

The governing equations for a sliding block are as follows:

$$M\frac{dv}{dt} = ku + \theta + A\ln\left(\frac{v}{v_o}\right) \quad (2)$$

$$\frac{du}{dt} = v - v_o \quad (3)$$

$$\frac{d\theta}{dt} = \left(-\frac{v}{D_c}\right)\left(\theta + B\ln\left(\frac{v}{v_o}\right)\right) \quad (4)$$

Equation 2 is a force balance of the spring force and friction. Equation 3 relates the slip of the block, relative to the motor, and Equation 4 is the semi-empirical relationship between the state and friction parameters.

Scoring Metric.

Both the experimental and theoretical model were reduced to two metrics to determine whether or not, for a given velocity and mass, would the FOTS and SiOH surfaces be discriminable.

For the experimental results, as shown in equation (5), these are weighted sums of the normalized skew and the normalized correlation.

$$Score = 1.4\times\widehat{Skew} + 0.3\times\widehat{Correlation} \tag{5}$$

For the theoretical results, as shown in equation (6), it is a combination of the difference in the magnitude of the force ($\Delta_{\text{Magnitude}}$) and the difference in oscillations made about 0 force ($\Delta_{\text{zero crossings}}$). $\Delta_{\text{Magnitude}}$ is defined as the percentage of the force trace where the FOTS and SiOH forces vary in magnitude a factor of 5 and $\Delta_{\text{zero crossings}}$ is defined as the difference in number of times the FOTS or SiOH force traces cross 0 force, which corresponds to a change in direction of the slider, relative to the motor (the slip). $\Delta_{\text{Magnitude}}$ is equivalent to its normalized value, since it has been described as a percentage and $\Delta_{\text{zero crossings}}$ is normalized by the percentage of the entire force trace, multiplied by 2 because a zero crossing takes two points, so the maximum a trace could oscillate would mean crossing zero 50% of the time.

$$Score = 0.5\times\widehat{\Delta_{Magnitude}} + 1.0\times\widehat{\Delta_{\text{zero crossings}}} \tag{6}$$

Force Traces and Correlations.

A correlation is a common technique used to identify whether or not two signals are similar in shape or not. In our case, our signals are the force traces from the model finger and we are comparing whether the model finger is similar or different on SiOH versus FOTS at different velocities and masses by using a cross-correlation. A force trace from SiOH and FOTS would have a higher cross-correlation if they both increase from their mean at the same time and same magnitude. Conversely, they would have a low correlation if, at a given time, SiOH decreases relative to its mean and FOTS does not change relative to its mean.

The correlation analysis can also be applied within the force traces of FOTS alone to gauge the internal consistency of the data. This is not an “autocorrelation”, another commonly used application of correlation analysis, because we are not comparing a single signal to itself to find internal patterns. Instead, we apply a cross-correlation of the different FOTS traces. These are shown in the second panel in each of the following figures, directly under the force traces.

The correlation is calculated by:

$$Cross\ Correlation\colon \Sigma[(SiOH(t) - \overline{SiOH}) * (FOTS(t - lag) - \overline{FOTS})]$$

where $\bar{x}$ represents the mean value, *t* is the time, and *lag* refers to a dummy *x*-variable used to compare the two data sets at all combinations of time.

We choose to normalize this cross-correlation by whichever dataset has a higher correlation *within* itself, whether it is from the SiOH or FOTS for a given condition. Assuming SiOH is higher, it is computed as:

$$Cross\ \widehat{Correlation} = \frac{Cross\ Correlation}{(SiOH(t) - \overline{SiOH})^2}$$

Fingers were tested at four masses at four velocities and within each test, the fingers were pulled on both FOTS and SiOH for a minimum of three runs each and this was repeated for three spots. Sample cross-correlations from a single spot from FOTS and SiOH are shown for four of the 16 conditions (varying velocity, constant applied mass). Each force curve from the FOTS is compared with each force curve from the SiOH condition. Then each spot of FOTS, with the three force traces inside, is compared with each spot of SiOH for each velocity and mass (creating 9 × 9 cross-correlations). The correlations are then averaged together to represent the correlation of SiOH and FOTS at a given mass and velocity.

Supplemental figures and tables

Table S1: Significance tests for discrimination, using sensitivity index. Columns depict the mean accuracy, mean d-prime value (***d′***), degrees of freedom (**d.f.**), test statistic (***t***), p-value (***P***), and 95% confidence interval on d' estimate (**95% C.I.**).

Experiment	**Accuracy (mean)**	***d′*** **(mean)**	**d.f.**	***t***	***P***	**95% C.I.**
Dry	71.67%	3.29	14	5.68	$P < 0.0001$	[2.05, 4.53]
Wet	84.17%	4.20	14	8.63	$P < 0.0001$	[3.16, 5.25]
Tapping	56.25%	2.09	13	3.66	$P = 0.0029$	[0.86, 3.32]

Table S2: Significance tests for discrimination, using GMM. Columns depict the coefficient of the intercept term (in units of log odds); standard error ($\sigma_{\bar{x}}$) of the estimate of this coefficient (the square root of the diagonal of the variance-covariance matrix); the Wald Z statistic $Z = (\hat{\beta}_0 - \beta_{0,0})/\mathrm{se}(\hat{\beta}_0)$, p-value, and 95% confidence interval on the intercept term $\hat{\beta}_0$ (recall that the null hypothesis of chance performance would yield $\hat{\beta}_0 = -0.69$.

Experiment	$\hat{\beta}_0$	$\mathrm{se}(\hat{\beta}_0)$	Wald Z	P	95% C.I.
Dry	1.33	0.50	4.07	$P < 0.0001$	[0.36,2.31]
Wet	1.90	0.41	6.40	$P < 0.0001$	[1.11,2.70]
Tapping	0.33	0.38	2.73	$P = 0.0063$	[-0.40,1.07]

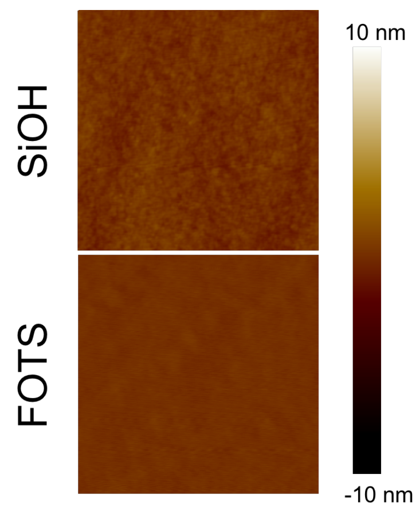


Figure S1: Surface Characterization. Atomic Force Microscopy (AFM) images of SiOH (top, R_a = 0.203) and FOTS (bottom, R_a = 0.206 nm) surfaces. AFM measurements were taken using a Veeco Scanning Probe Microscope in tapping mode. Data was analyzed using NanoScope Analysis v1.40 software (Bruker Corp.).

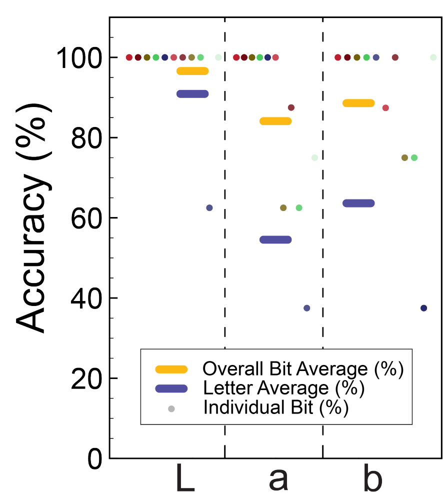


Figure S2: Psychophysical Results (ASCII Alphabet). Individual subject bit accuracy (colored dots), overall average bit accuracy (gold line) and overall letter accuracy (purple line).

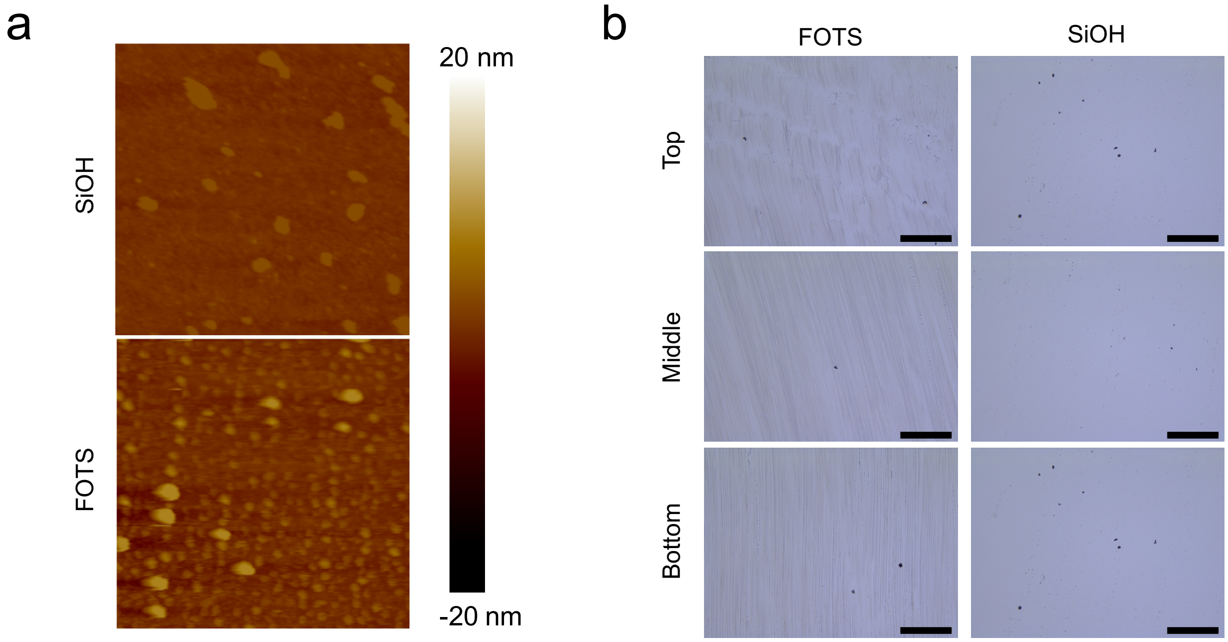
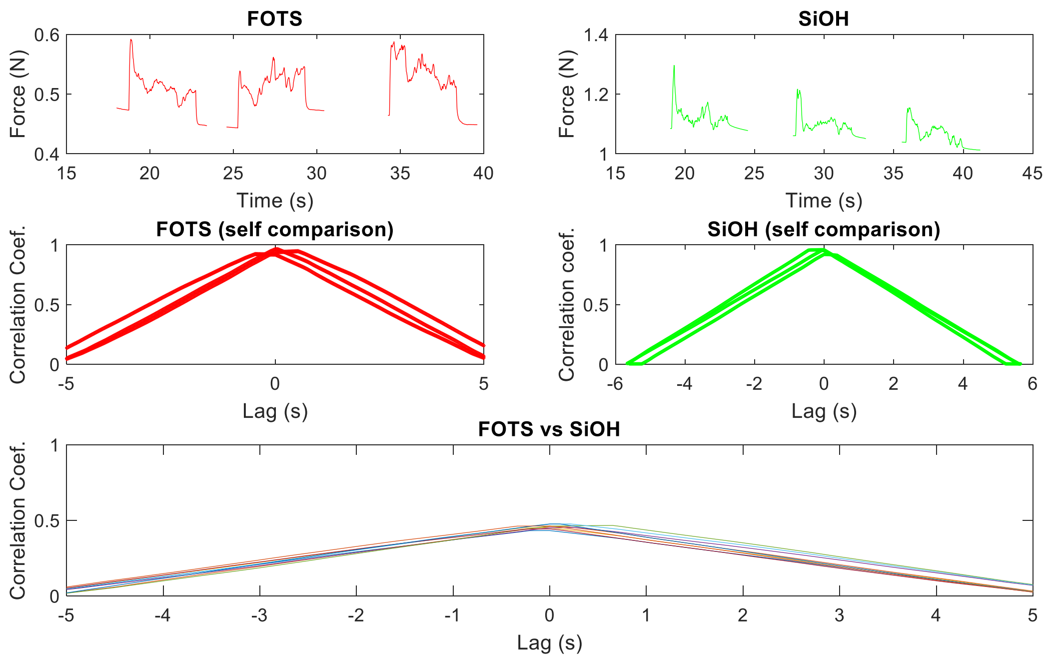
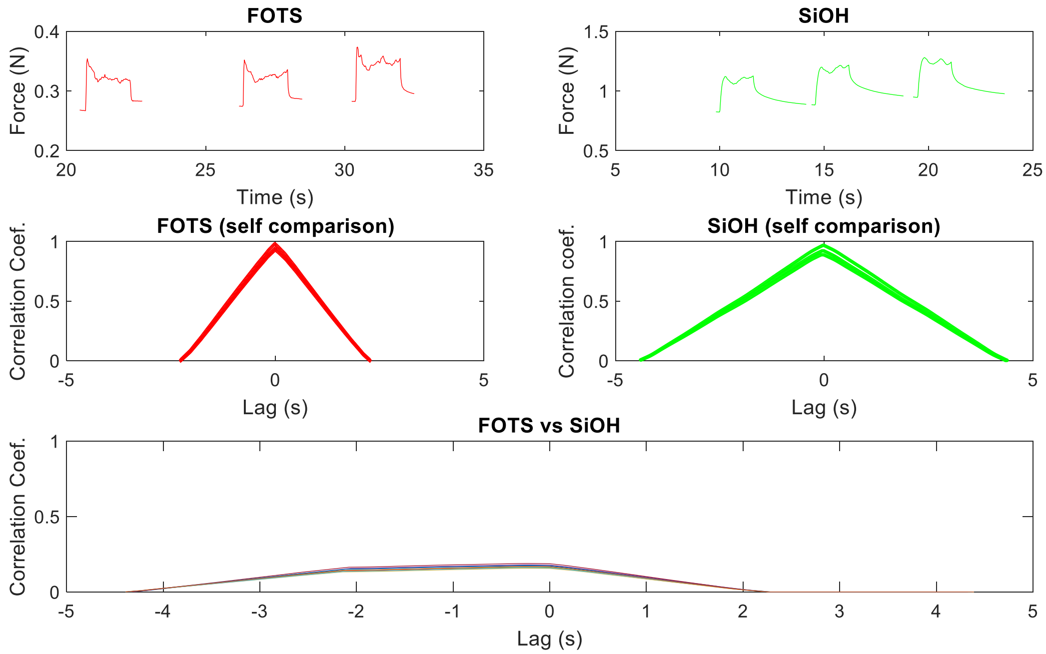


Figure S3: Post Exploration Surface Characterization. (a) Atomic force microscopy images of surface height topography (1 µm × 1 µm scan dimensions) of SiOH and FOTS surfaces post exploration (b) Optical micrographs (500 µm scale bars) of finger deposition on FOTS (left) and SiOH (right) surfaces (top to bottom) after a single swipe across a distance of 2.5 cm.

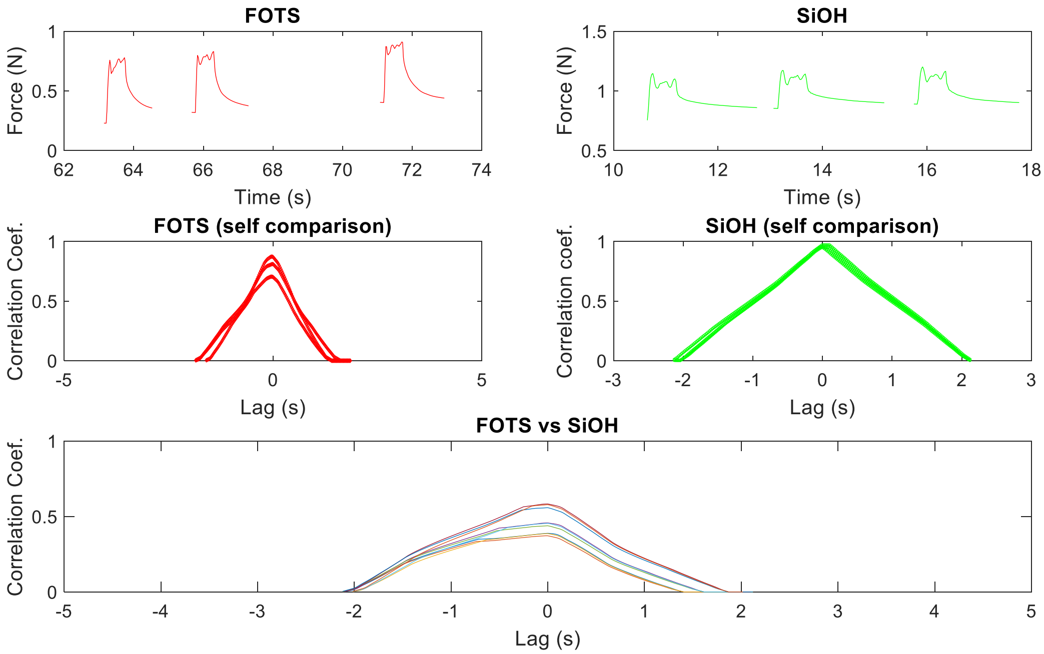
1 mm/s, 25 grams



2.5 mm/s 25 grams



7.5 mm/s, 25 grams



1 cm/s, 25 grams

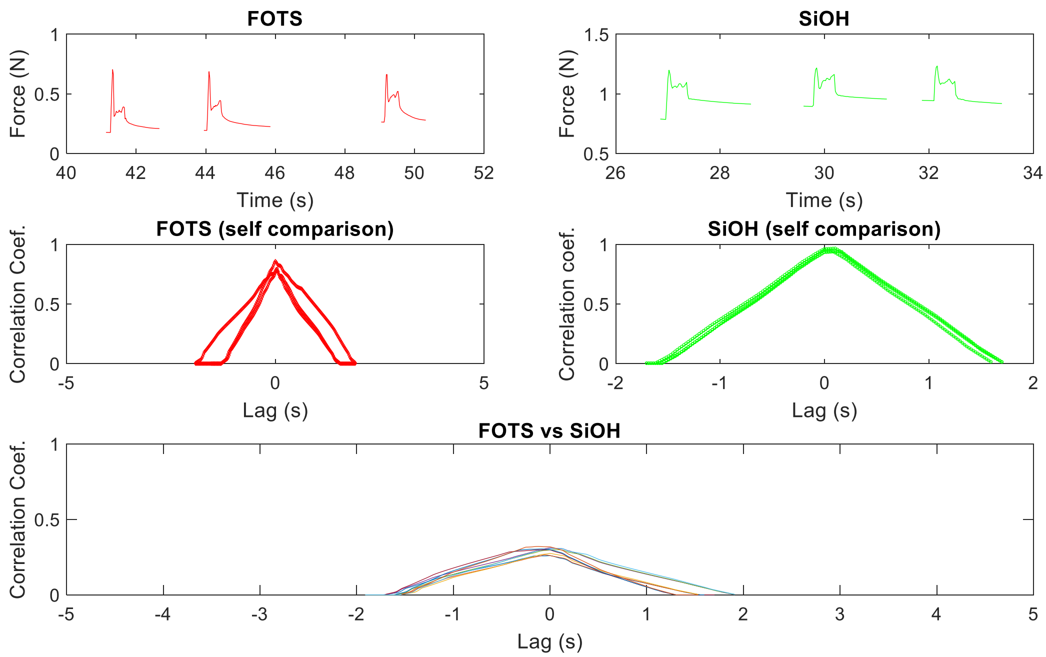


Figure S4: Force and correlation plots at 25 grams of applied load.

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