Supplementary Materials

CONTENT

I. SMOOTH PROBE EXPERIMENTS

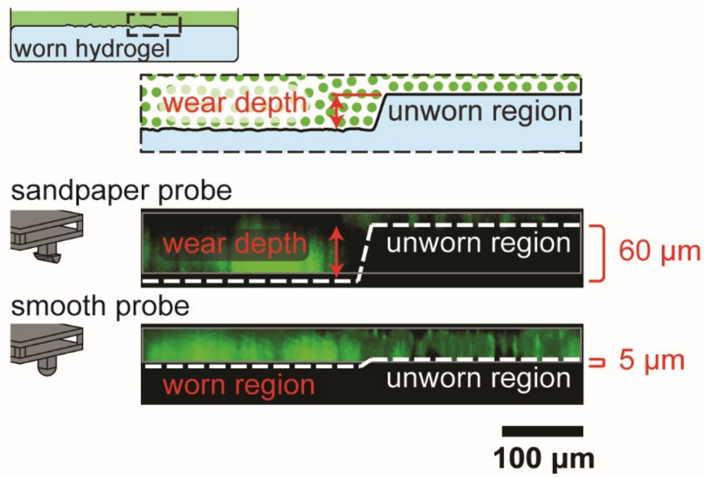

Figure S1. Surface Topography. The experimental procedure with the smooth probe is the same as that of the sandpaper probe, which we described in the paper. Hydrogels were polymerized 24 hours prior to the first experiment. The surface properties were characterized by microindentations and friction tests, followed by wear application with the smooth probe, and finally recharacterized by microindentations and friction tests. Wear was applied at a load of 10 mN, speed of 3 mm/s, and duration of 5,000 cycles. All surface characterizations and wear applications were performed with the samples submerged underwater. The top figure is a schematic of the method used to measure the wear depth of the hydrogels. After allowing the fluorescent microbeads to settle onto the surface of the worn hydrogel (~30 min), we imaged the xz-plane that included the location of the beads on the both the worn and unworn regions. The middle figure shows the results which we had presented in the manuscript, in which the surface of the hydrogel was worn with a probe covered in sandpaper. The bottom figure shows the location of the beads on a hydrogel worn with a smooth spherical steel probe. The wear parameters were the same between the two probes (load=10 mN, speed=3 mm/s, cycles=5,000), however, only two wear tests were performed before imaging for the smooth probe (total of 10,000 cycles). The wear rate of the smooth probe was calculated using the method outlined in the manuscript to be $1.9 \cdot 10^{-2}$ $mm^3N^{-1}m^{-1}$, which is about 3 times smaller than that of the sandpaper probe ($6.6 \cdot 10^{-2}$ $mm^3N^{-1}m^{-1}$).

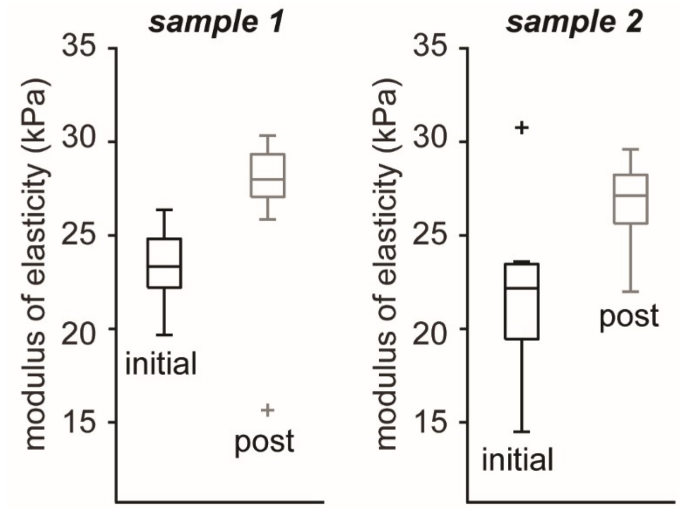

Figure S2. Stiffness Results. Two samples were worn with the smooth probe, and microindentations were performed before and after wear application. Each box plot shows the results of 10 indentations with a 1 mm radius steel ball probe. The probe indented to a load of 1 mN in 20 seconds, thus indenting at speeds of 8-17 μm/s. Both samples showed that wear increased their surface modulus.

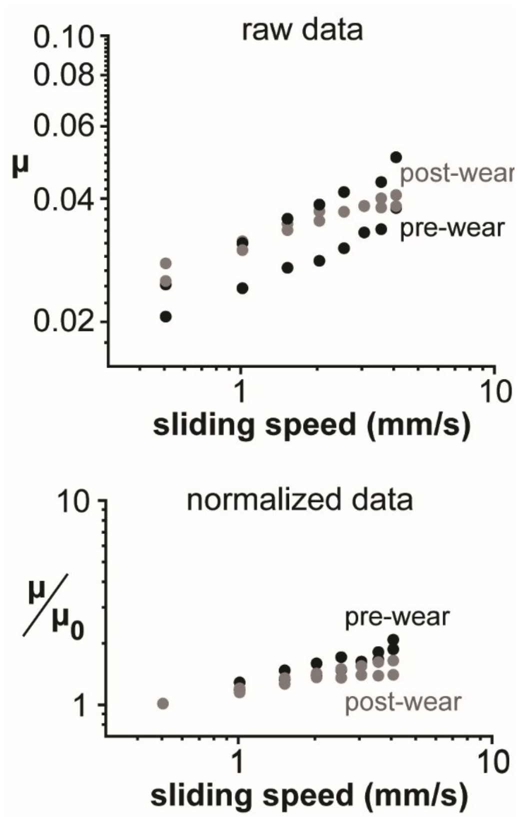

Figure S3. Lubrication Raw Data and Results. Friction tests were performed on the two hydrogel samples before and after wear. The friction coefficient was measured at 8 speeds from 0.5 mm/s to 4 mm/s at increments of 0.5 mm/s. A steel ball probe with a 1 mm radius applied a 1 mN load at the surface of the hydrogels. A reciprocating stage held the sample, and travelled a stroke distance of 1.5 mm 100 times at each speed. The average of the 100 friction coefficient values for each speed are graphed above. The black data points are from pre-wear measurements, and the grey points are from post-wear measurements. The top graph shows the friction coefficient values measured by the instrument. The bottom graph shows the friction coefficient values normalized by the friction coefficient of the lowest speed (μ_0), which was also the lowest friction coefficient value. The slopes of the curves were calculated, and we found that the slopes decreased with wear by about 41.2% and 30.3% for the two samples.

II. SURFACE VS BULK EXPERIMENTS

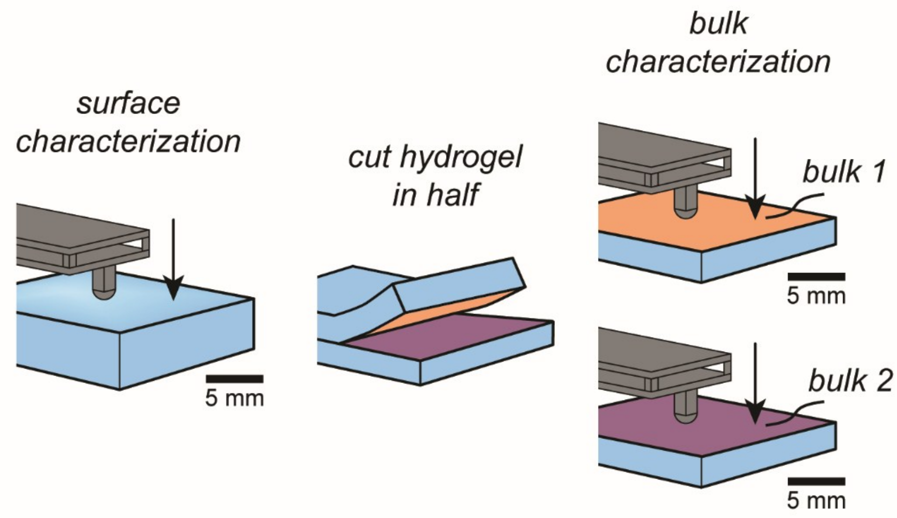

Figure S4. Methods Schematics. Hydrogels for the following experiments were created using the same method detailed in the manuscript. To compare the surface of the hydrogel to the bulk, we first performed microindentations on the surface of the hydrogel samples, which were submerged underwater. We then used a razor blade attached to a custom instrument to smoothly cut the hydrogels in half. The two halves of the hydrogel were then resubmerged underwater, and microindentations were performed immediately (within 5 minutes) afterwards on the two newly revealed bulk surfaces. We performed this experiment on two hydrogels.

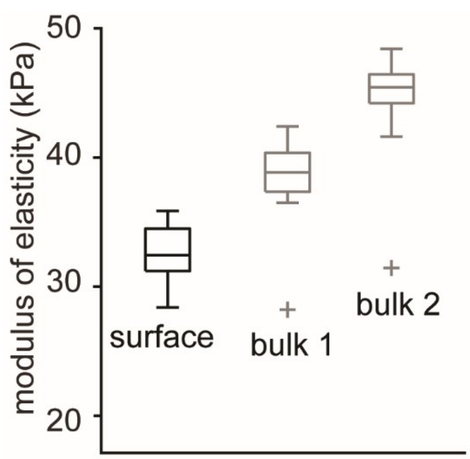

Figure S5. Stiffness Results. The plots above show the stiffness values measured for two samples that were cut in half. The two measurements of the bulk are greater than the surface. Thus, both samples confirmed that the surface of the hydrogel is significantly softer than the bulk.

III. SANDPAPER PROBE EXPERIMENTS

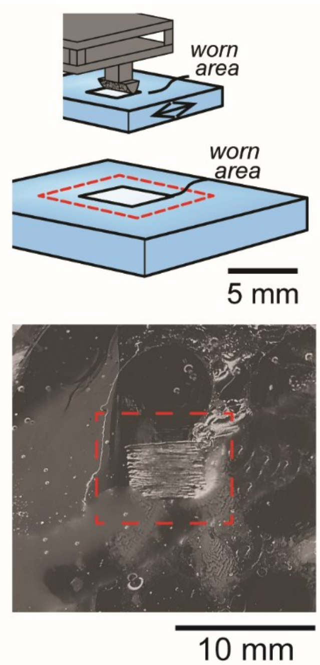

Figure S6. Worn Hydrogel Surface Image. As confirmation that the hydrogel was worn using the sandpaper probe, we captured an image of the surface using a camera. The schematics illustrate the location of the wear scar to provide a reference for the camera image.

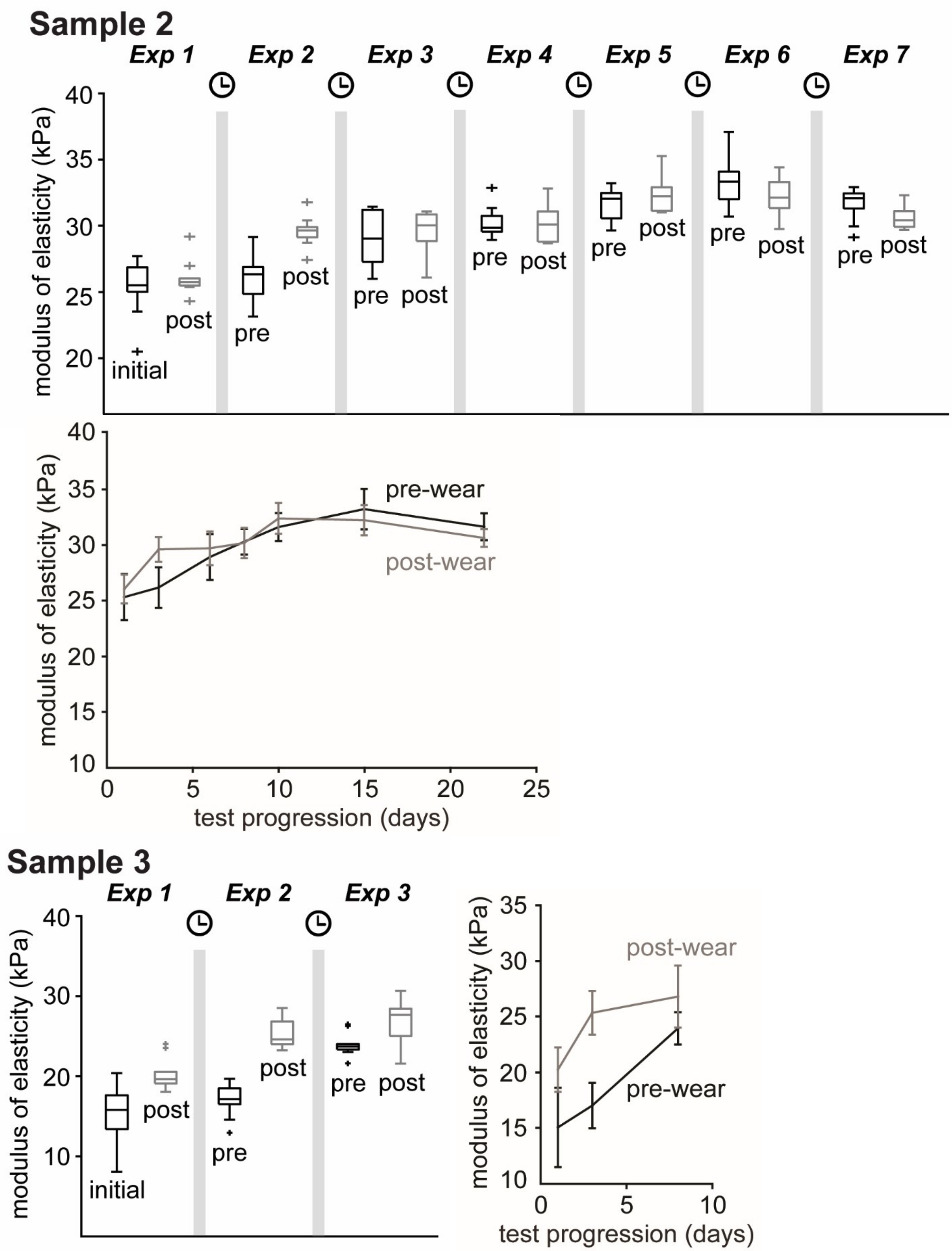

Figure S7. Stiffness Results. For this work, we performed wear tests using the microtribometer on three hydrogels. We present the results for one hydrogel in the manuscript, and provide the results for the other two samples here. The experimental procedure was the same for all samples, which we had detailed in the manuscript. We performed 7 wear experiments on Sample 2, and 3 wear experiments on Sample 3. The box plots show the stiffness changes with each experiment. We recorded the days in which each experiment was performed and show the evolution of stiffness over time in days with line plots.

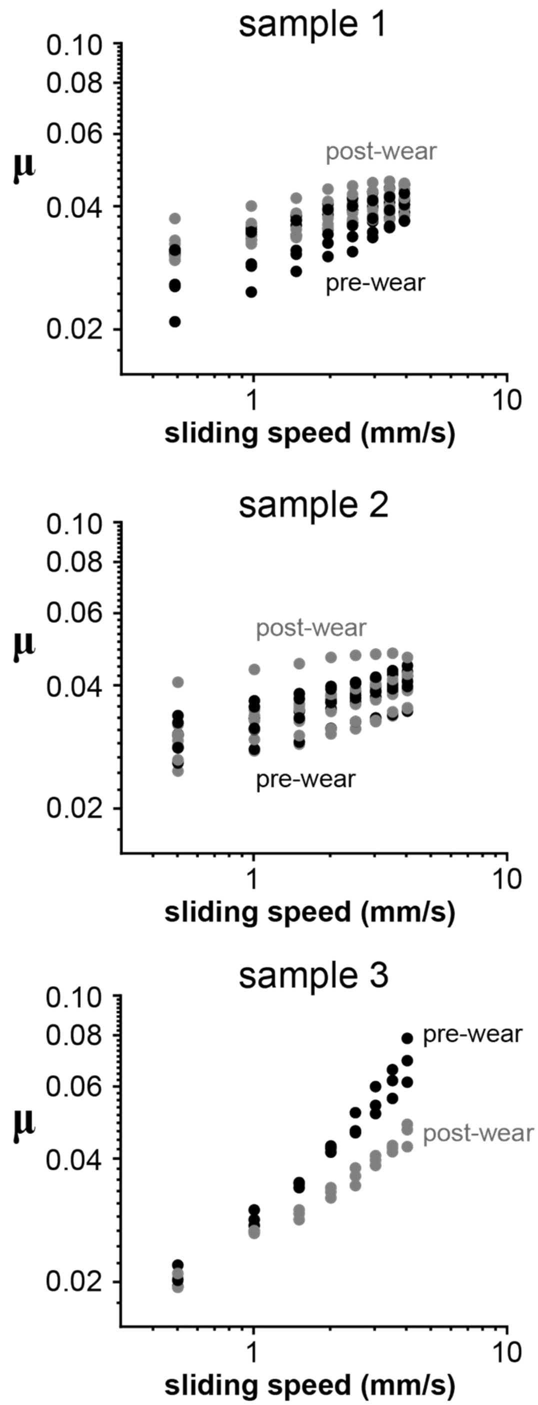

Figure S8. Friction Raw Data. We present the raw friction data we acquired for all three samples. The black data points are from pre-wear measurements, and the grey points are from post-wear measurements. We performed 6 wear experiments on Sample 1, 7 wear experiments on Sample 2, and 3 wear experiments on Sample 3. The majority of the friction coefficient values ranged from 0.02 to 0.05 for both pre-wear and post-wear measurements. This indicates that surface wear does not compromise the effective lubricity of the hydrogel.

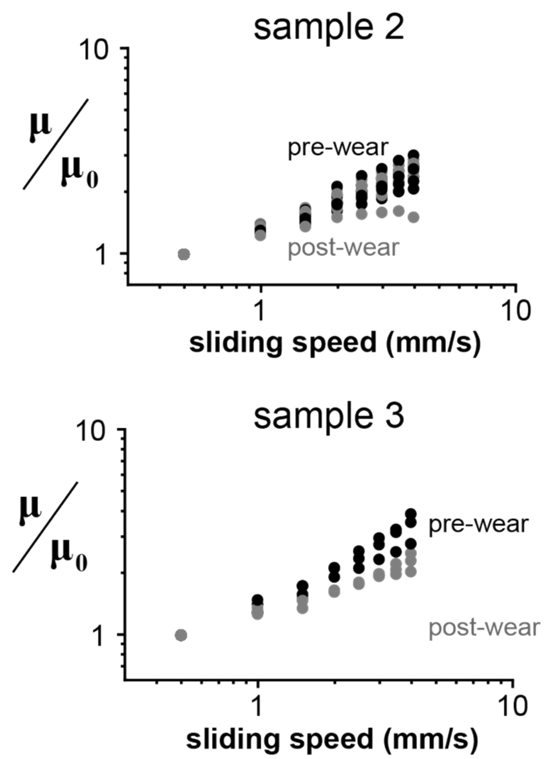

Figure S9. Friction Results. To compare the slopes between the pre-wear and post-wear lubrication curves, we normalized the friction coefficient values by dividing them by the friction coefficient at the lowest speed (0.5 mm/s) of that individual curve. From these graphs, it is visually evident that the pre-wear lubrication curves have a greater slope than the post-wear lubrication curves. We measured the difference between the slopes of the pre-wear and post-wear curves from these two samples, and included this data in Figure 12 of the manuscript.