Electronic Supplementary Information
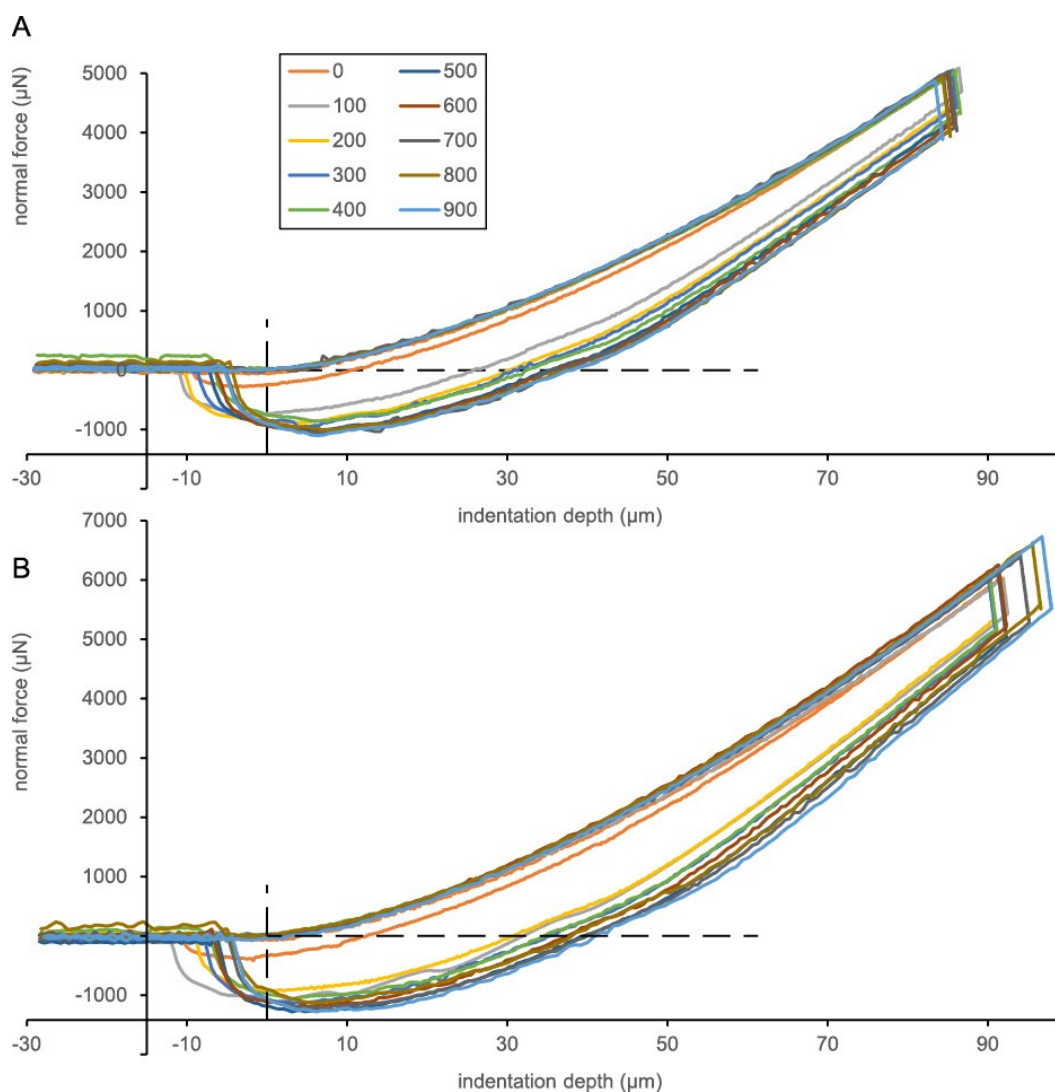
Calculation of Work of Adhesion from Indentations

To find the total adhesive energy of a glass probe sticking to the surface of a hydrogel surface, the area between the x-axis and normal forces less than zero is calculated using numerical integration from the raw data. All force-deflection curves were numerically integrated according to Eqn (1) in the main text, resulting in adhesion energies between 5 and 40 nJ depending on the hold time allowing the surface energy to increase. The number of data points used for integration was between 50 and 100 (Fig S1).

Fig. S1 Force-deflection curves from the microindentation experiment used to calculate the work of adhesion upon probe retraction. Relaxation time after load application by a 0.5-mm radius glass probe was controlled between 0 and 900 seconds (as indicated in legend) to assess the effect on the work of adhesion. (A) Initial set, $F_{n,max} \sim 5$ mN (B) Confirmation set, $F_{n,max} \sim 7$ mN. Results are shown in Fig. 1 of the main text.

Method of calculating Coefficient of Diffusion

For a poroelastic material such as a hydrogel, the coefficient of diffusion describes the movement of fluid through the polymer mesh, and in this case, how the material relaxes over time. We find a coefficient of diffusion by fitting theory (Eqn (6) of main text) to experimental data of force relaxation.

Starting from Eqn (4) in the main text, the drop in normal force versus time can be predicted. The time constant was found by a linear least squares fit of Eqn (4) for each step indentation (Fig S2A). Six step indentations to varying depths were performed, and the resulting time constants report in Table S1.

After the characteristic poroelastic relaxation time constant is calculated, the contact area is found from Eqn (3), then used in Eqn (6) to fit for the coefficient of diffusion (Fig S2B). The value for effective surface energy density used in the area calculation (Eqn (3)) is the relatively high dehydrated value, on the basis that the experiment duration is long enough. This relationship is summarized in Eqn (S1).

$$D \equiv \frac{a^2}{\tau_p} = \frac{R\delta}{\tau_p}\left[1 + \frac{3\gamma_d \pi R}{F_n} + \sqrt{\frac{6\gamma_d \pi R}{F_n} + \left(\frac{3\gamma_d \pi R}{F_n}\right)^2}\right]^{2/3} \quad [S1]$$

Table S1. Summary of indentation parameters and fit parameters: indentation depth δ, poroelastic relaxation time τ_p, and contact radius a.

δ (μm)	$\tau_{p\,(s)}$	a (μm)
33.0	34.9	142.7
71.6	59.2	202.3
109.3	103.8	245.3
146.5	125.0	281.0
183.3	254.1	312.6
219.8	294.7	340.8
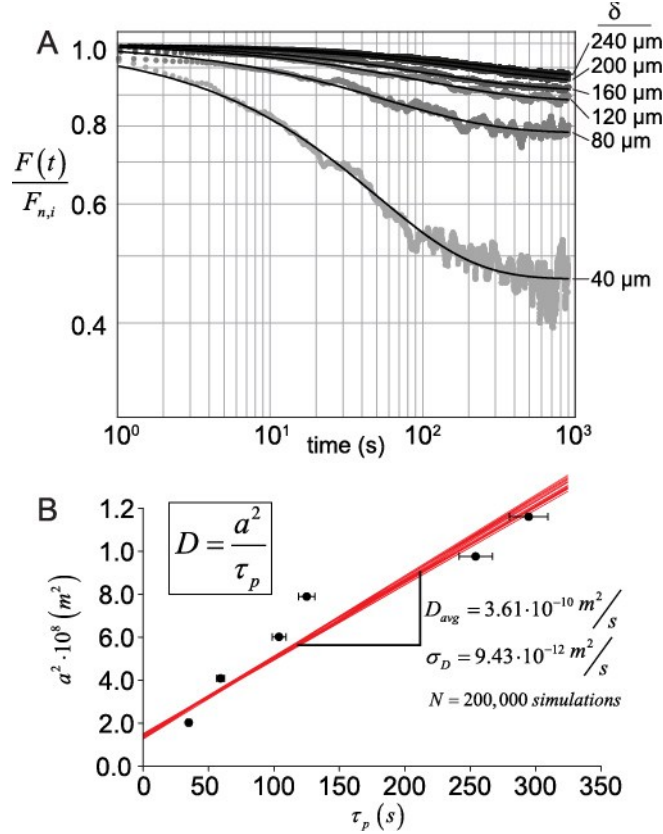
Fig S2. Poroelastic relaxation experiments following the force decrease over time for varying depths of indentation, δ. (A) Each curve was fit using Eqn. (4) with $F_{n,i}$, $F_{n,f}$, and τ_p as the free parameters by linear least squares regression. (B) Given the definition of coefficient of diffusion, a Monte Carlo simulation of the linear slope between the square of contact radius and poroelastic relaxation was used to determine the average coefficient of diffusion and the standard deviation of N=200,000 fits; red lines indicate 20 representative slopes, and the point (0,0) was also included in the simulations. The vertical error bars for a^2 are determined by propagation of error through measurement of indentation depth, and are covered by the data points. The horizontal error bars assume a systematic 5% error in the poroelastic relaxation times. See summary in Table S1.

Area method for determining adhesion forces upon the initiation of slip

As slip is initiated after a long dwell time of applied pressure, the glass probe is translated to a nascent, fully hydrated surface that has not yet experienced pressure. An area-based rules of mixture between the dehydrated and hydrated regions of the interface is employed to predict the static coefficient of friction, as shown in Fig S3A.

An angle "θ" is defined as the angle between the x-axis and a radius terminating at the intersection of the initial and current contact areas. It indicates how far the contact area has moved compared to the contact radius during initial slipping of a surface in contact with a hydrogel.

$$\theta = \cos^{-1}\left[x/(2a)\right]; \ \ 0 \le \theta \le \pi/2 \tag{S2}$$

Before sliding, $\theta = \pi/2$ and $x/(2a) = 0$; when the contact area moves, the values approach $\theta \to 0$ and $x \to 2a$ as the overlap between the initial and current contact areas shrinks. The central region where the contact areas overlap is $A_{blue} = 2\theta a^2 - ax\sin\theta = 2a^2\left(\theta - \cos\theta\sin\theta\right)$. When $\theta = 0$, the contact area has

moved beyond its initial location, and $A_{blue} = 0$. The white area is the difference between the entire contact area and the blue portion; see Fig S3B for the area rules of mixture versus translation distance.

$$A_{white} = \pi a^2 - A_{blue} = \pi a^2 \left[1 - A_{blue} / \left(\pi a^2 \right) \right] = \pi a^2 \left[1 - \frac{2}{\pi} \left(\theta - \cos\theta \sin\theta \right) \right] \qquad [S3]$$
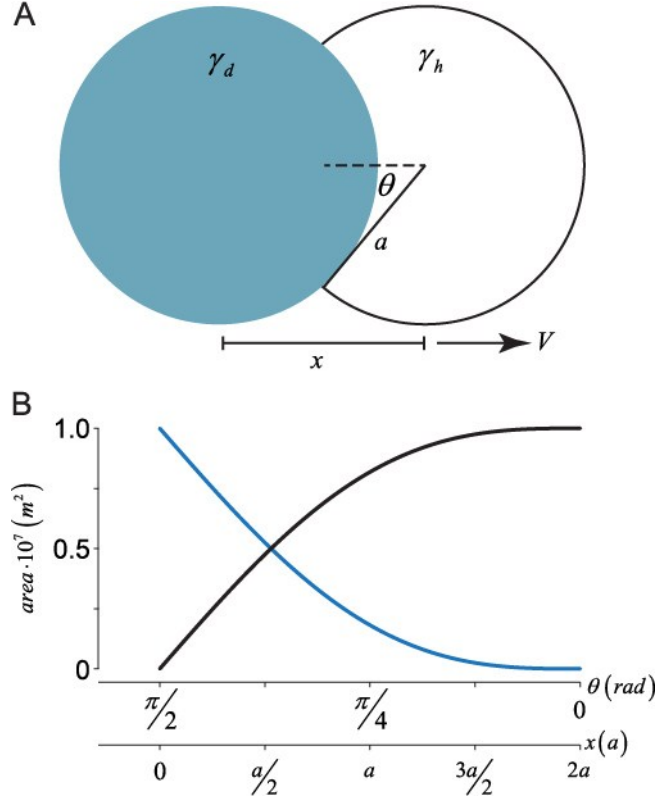
Figure S3. Schematic of area-based rules of mixture of surface energy densities to predict the static coefficient of friction after a long dwell time. (A) Schematic of a pressure-affected contact zone (left, shaded), characterized by a surface energy density in a dehydrated state, γ_d, and the contact area after the hydrogel surface has translated away from the probe a distance x at a velocity V (right, white with black outline). (B) The effective surface energy density of the interface as it moves away from a dehydrated zone can be calculated as the area-based rules of mixture between γ_d and γ_h. For full overlap before slip, $A_{white} = 0$ and $\theta = \pi/2$; when the interface has fully departed the dehydrated area, $A_{white} = \pi a^2$ and $\theta = 0$.

The friction force is the combination of hydrated and dehydrated surface energies, and the area fractions allow for the average surface energy to be approximated during the transition between stationary and sliding contact. And as one area is expressible in terms of the other, the friction force can be expressed as a function of A_{blue}.

$$F_f = \frac{\gamma_h A_{white} + \gamma_d A_{blue}}{l} \qquad [S4]$$

$$F_f = \frac{1}{t} \left[\gamma_h \pi a^2 \left(1 - \frac{A_{blue}}{\pi a^2} \right) + \gamma_d A_{blue} \right] \qquad [S5]$$

Reorganizing the equations to be only a function of A_{blue} results in S6.

$$F_f = \frac{1}{l}\left[\gamma_h \pi a^2 + A_{blue}(\gamma_d - \gamma_h)\right]$$ [S6]

Rearranging Equation S3 and plugging it into S6:

$$F_f = \frac{a^2}{l}\left[\gamma_h \pi + 2(\gamma_d - \gamma_h)(\theta - \cos\theta\sin\theta)\right]$$ [S7]

When the probe first begins moving $\theta = \pi/2$ and the friction force $F_f = (\gamma_d \pi a^2)/l$, as the entire area is in contact with the dehydrated gel surface because the probe has not left its starting position. For $\theta = 0$, when there is no area of overlap and only contact with the hydrated gel surface, $F_f = (\gamma_h \pi a^2)/l$. To represent the change in friction force between these two states as a function of distance moved, x, we convert the key variable from θ to x using the relationship in Equation (S2).

$$\theta - \cos\theta\sin\theta = \cos^{-1}\left(\frac{x}{2a}\right) - \frac{x}{2a}\frac{\sqrt{4a^2 - x^2}}{2a} = \cos^{-1}\left(\frac{x}{2a}\right) - \sqrt{\left(\frac{x}{2a}\right)^2 - \left(\frac{x}{2a}\right)^4}$$ [S8]

Substituting the dimensionless factor $b = x/(2a)$ for simplicity, the friction force vs. position can be approximated as Eqn (S9).

$$F_f = \frac{a^2}{l}\left[\gamma_h \pi + 2(\gamma_d - \gamma_h)\left(\cos^{-1}b - \sqrt{b^2 - b^4}\right)\right]$$ [S9]

Time method for determining adhesive friction in steady-state sliding

In the direction of motion, the leading edge of the interface begins at the hydrated surface energy, γ_h, and then surface energy increases corresponding to the duration of applied pressure during the passing of the probe. The circular geometry shows the points along each curve are all the same distance from the outer edge in the direction of the probe's relative motion, and with the entire contact area moving at a particular speed, each curve relaxes for the same amount of time depending on the angle φ, the angle between the y-axis and the line from the origin to the end of an arc (see schematic in Fig 4A of the main text). Each dashed line represents a curve where the surface energy remains the same. The time each curve spends in contact, $t_{contact}$ will control the surface energy along each arc, as shown by the dependence $t_{contact} = (2a\sin\varphi)/V$, where a is the radius of contact and V is the sliding speed. The longest contact time occurs when $\varphi = \pi/2$ when the hydrogel lies beneath the full diameter of the contact area, $t_{max} = (2a)/V$.

When the contact time is determined, it can be compared to the characteristic poroelastic relaxation time, as shown in the exponent of Eqn (S10), with a description of the surface energy density for any location in the interface.

$$\gamma(\varphi) = \gamma_d - (\gamma_d - \gamma_h)e^{-\alpha\left(\frac{t_{contact}}{\tau_p}\right)^\beta}$$ [S10]

The effective average surface energy density is required to determine the coefficient of friction of the entire interface. To find this, the average contact time of the interface is calculated, then used in Eqn (S10).

$$arc\ length = a\left(\pi - 2\varphi\right) \tag{S11}$$

$$area\ element = a^2\left(\pi - 2\varphi\right)\cos\varphi\,d\varphi \tag{S12}$$

$$\overline{t}_{contact} = \frac{1}{\pi a^2}\int_0^A t_{contact}\,dA = \frac{2a^3}{\pi a^2 V}\int_0^{\pi/2}\left(\pi - 2\varphi\right)\sin\varphi\cos\varphi\,d\varphi \tag{S13}$$

$$\overline{t}_{contact} = \frac{a}{2V} = \frac{t_{max}}{4} \tag{S14}$$

This equation applies to contact between a flat surface and a spherical indenter producing a circular contact area, but the concept of averaging the time of contact in this manner could be applied to other shapes.