Electronic Supplementary Information

Eliminating cohesive failure in pH-switchable adhesion
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Modulus and swelling properties of the hydrogels

![Graphs showing modulus and swelling properties](image)

**Figure S1.** The Hertz equation was used to calculate the pH-dependent modulus of the DN hydrogel (a) and the SN hydrogel (b). The modulus is the gradient of a plot of $PR$ as a function of $a^3$, where $a$ is the contact radius; $P$, the load; and $R$, the radius of the gel. The elastic moduli (and maximum swelling) of the gels are included in **Table S1**

**Table S1.** Elastic modulus and swelling ratio of the gels used in this work

<table>
<thead>
<tr>
<th>pH</th>
<th>Modulus DN gel (MPa)</th>
<th>Swelling ratio DN gel</th>
<th>Modulus SN gel (MPa)</th>
<th>Swelling ratio SN gel</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1.18 ± 0.02</td>
<td>1.4 ± 0.1</td>
<td>0.42 ± 0.08</td>
<td>7.3 ± 0.3</td>
</tr>
<tr>
<td>5.8</td>
<td>1.09 ± 0.01</td>
<td>6.5 ± 0.6</td>
<td>0.30 ± 0.04</td>
<td>13.6 ± 0.4</td>
</tr>
<tr>
<td>12</td>
<td>1.00 ± 0.02</td>
<td>24.3 ± 1.2</td>
<td>0.17 ± 0.01</td>
<td>48.6 ± 1.4</td>
</tr>
</tbody>
</table>

Table S1 is identical to Table 1 in the main article, except for the addition here of the values for pH 12.
Time before gels are detached

(a)

Figure S2. (a) Force-distance curves for a DN hydrogel (previously attached to the brush in pH 6 solution), which was removed from the brush surface at pH 1, after waiting for different periods of time. (b) Force-distance curves for a DN hydrogel brought into contact with the surface at pH 1 and allowed to remain there for 10 min or 2 h before being separated at the same pH. These data correspond to the results shown in Figure 5 of the article

Less-crosslinked DN gels

(a)

Figure S3. Optical microscopy images for the loosely crosslinked DN gel (the amount of crosslinker was decreased by a factor of 25) when it was retracted from the PDEAEMA brush underwater at pH 6. The applied force used was 0.5 N to bring the DN hydrogel in contact with the PDEAEMA brush for 2 min before (a) being retracting with a speed of 50 mm/min. Since this DN hydrogel was malleable, it underwent significant stretching (b, c) before it fractured (d), resulting in cohesive failure. These images are taken from the associated video.