Supporting Information

Shape-change Programming of Zwitterionic Hydrogels via Chemical Gradients Directed by Surface Energy

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Table. S1. Optimization of parameters contributing to the efficiency of shape-change in gradgels.

<table>
<tr><th>Input</th><th colspan="7">Output</th></tr>
<tr><td rowspan="2">Ratio of Comonomers (MAA:DMAPS (wt.%))</td><td colspan="3">1:2</td><td colspan="4">3:1</td></tr>
<tr><td colspan="3">Isotropic Swelling</td><td colspan="4">Significant Bending</td></tr>
<tr><td rowspan="2">CNC Concentration (wt.%)</td><td colspan="2">1</td><td colspan="3">2</td><td colspan="2">3</td></tr>
<tr><td colspan="2">Large Deformation-Fast Relaxation</td><td colspan="3">Large Deformation-Slow Relaxation</td><td colspan="2">Small Deformation-Slow Relaxation</td></tr>
<tr><td rowspan="2">Thickness (µm)</td><td>125</td><td colspan="2">250</td><td colspan="3">500</td><td>750</td></tr>
<tr><td>Fast Relaxation</td><td colspan="2">Fast Relaxation</td><td colspan="3">Slow Relaxation</td><td>Slow Relaxation</td></tr>
<tr><td rowspan="2">Cell Size (width×length (mm^2))</td><td colspan="2">20×25</td><td colspan="3">20×37.5</td><td colspan="2">20×75</td></tr>
<tr><td colspan="2">Consistent Separation</td><td colspan="3">Inconsistent Separation</td><td colspan="2">Inconsistent Separation</td></tr>
<tr><td rowspan="2">Hydrophobic Treatment</td><td colspan="4">Gas Phase Silanization</td><td colspan="3">Liquid Phase Silanization</td></tr>
<tr><td colspan="4">Inconsistent Separation-Small Deformation</td><td colspan="3">Consistent Separation-Large Deformation</td></tr>
</table>

Table. S 2. Hansen solubility parameters for hydrogel components.

	δ_D	δ_P	δ_{hb}	δ_{Total}
DMAPS	22.1049	28.1924	6.5846	36.425197
MAA	15.8	2.8	12	20.0369658
Water	15.5	16	42.3	47.8073216

Table. S 3. Values for the Miscibility parameter of the comonomers in water at bulk.

$\chi_{DMAPS\ in\ Water}$	$\chi_{MAA\ in\ Water}$	$\chi_{DMAPS\ in\ MAA}$
9.44056E-07	5.62167E-06	9.14326E-06

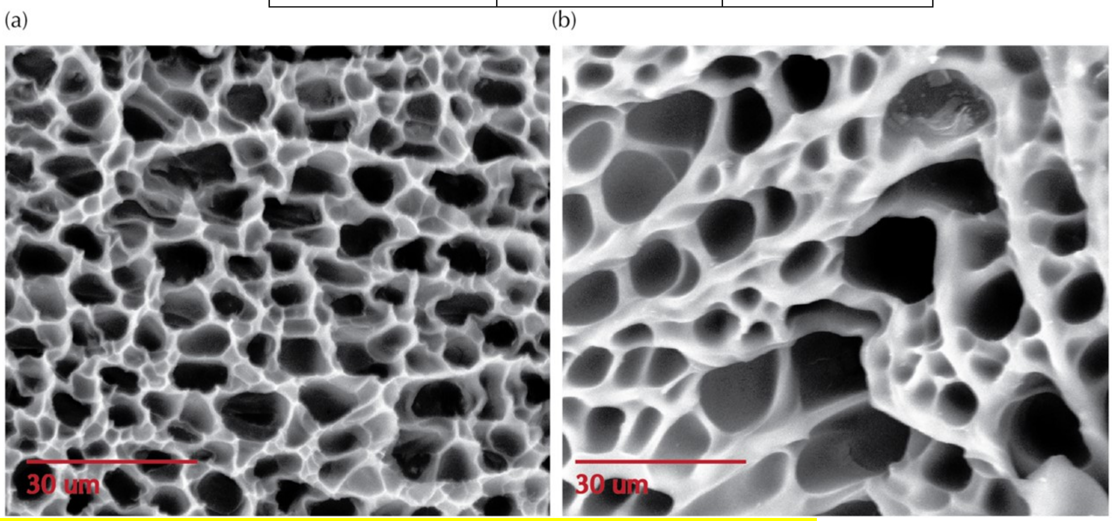

Figure S 1. SEM images of a) MAA and b) DMAPS hydrogels.

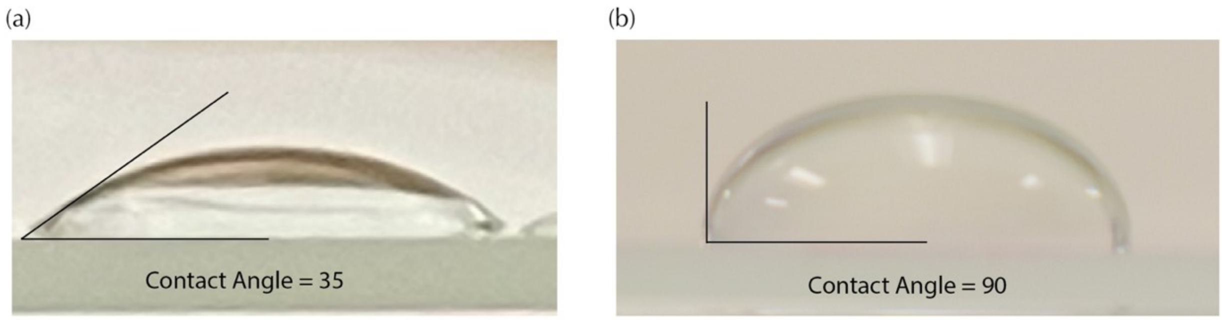

Figure S 2. Contact angle measurements on a) hydrophilic glass slide modified with ozone-plasma treatment and b) hydrophobic glass slide modified with liquid phase Silanization.

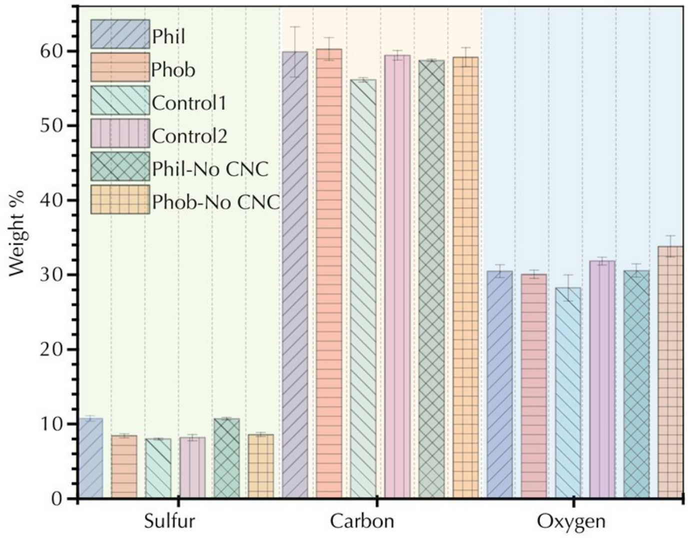

Figure S 3. Elemental analysis of the opposite sides of the gradgel, control hydrogel, and the gradgel with no added CNC. Average values are reported from at least three samples.

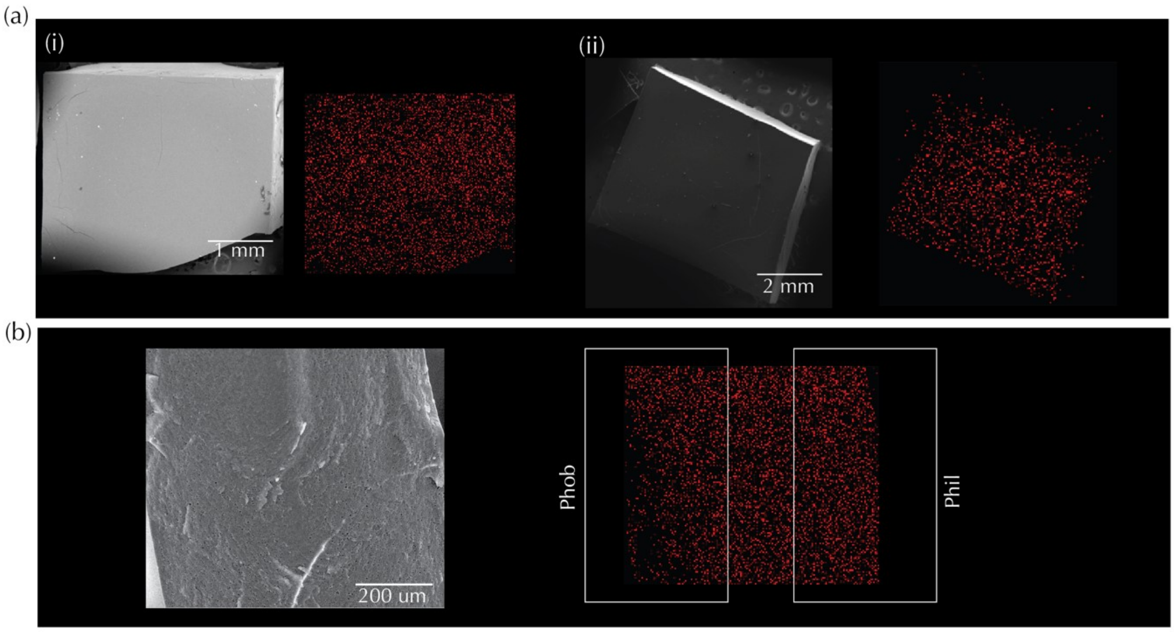

Figure S 4. Elemental mapping for Sulfur from EDX measurements on a) the opposite sides of the gradgel. (i) is the side in proximity to the hydrophilic glass slide and (ii) is the side in proximity to the hydrophobic glass slide. b) along the cross section of the gradgel.

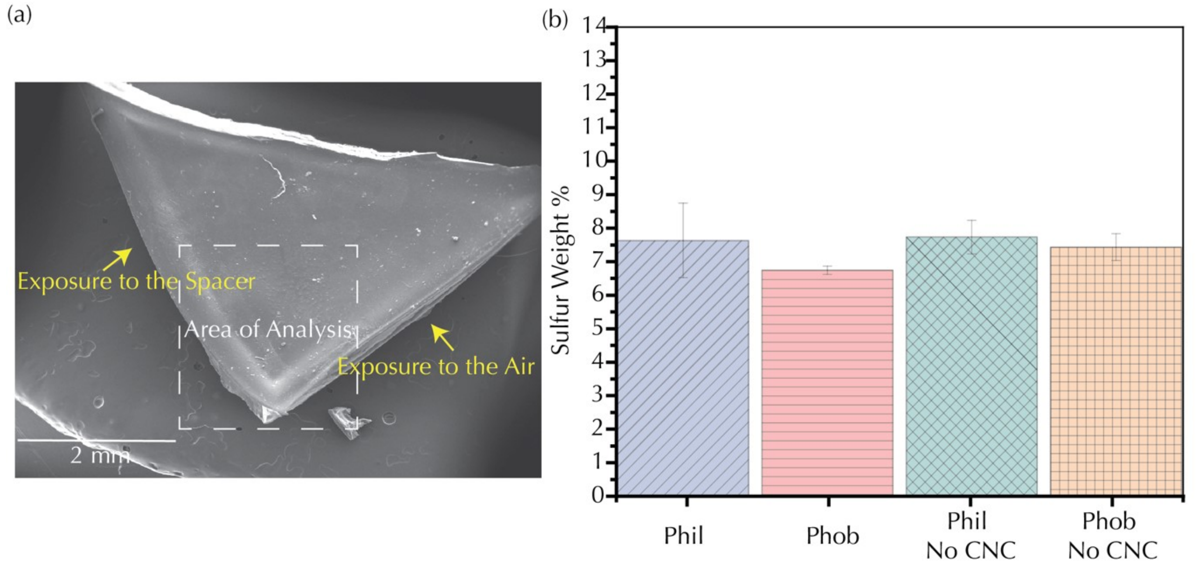

Figure S 5. a) The region of interest for elemental analysis for Sulfur from EDX measurements on the edge of the sample exposed to air and spacer. b) Elemental analysis of the opposite sides of the regular gradgel and the gradgel with no added CNC on the edges. Average values are reported from at least three samples.

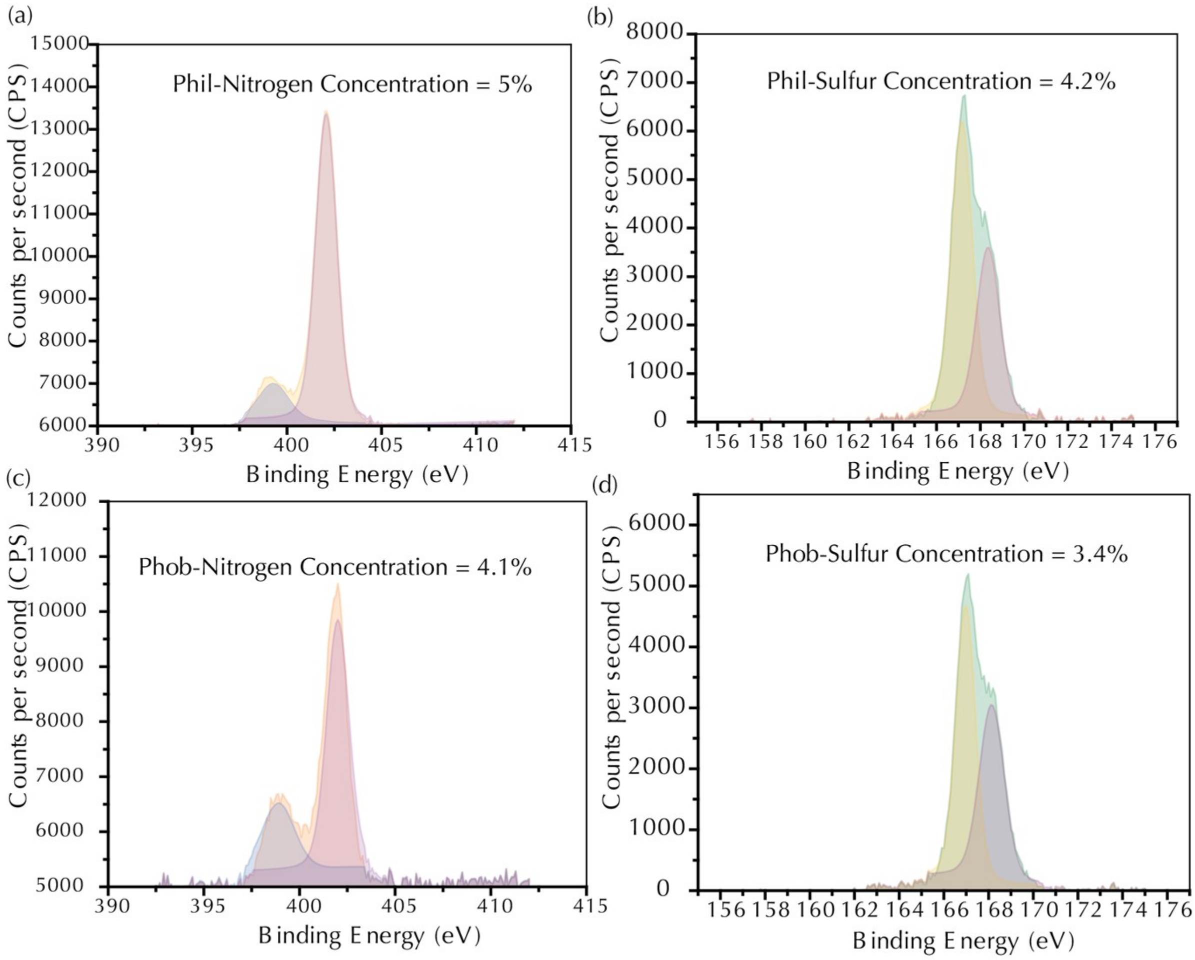

Figure S 6. X-ray photoelectron spectroscopy (XPS) analysis of the opposite sides of the gradgel.

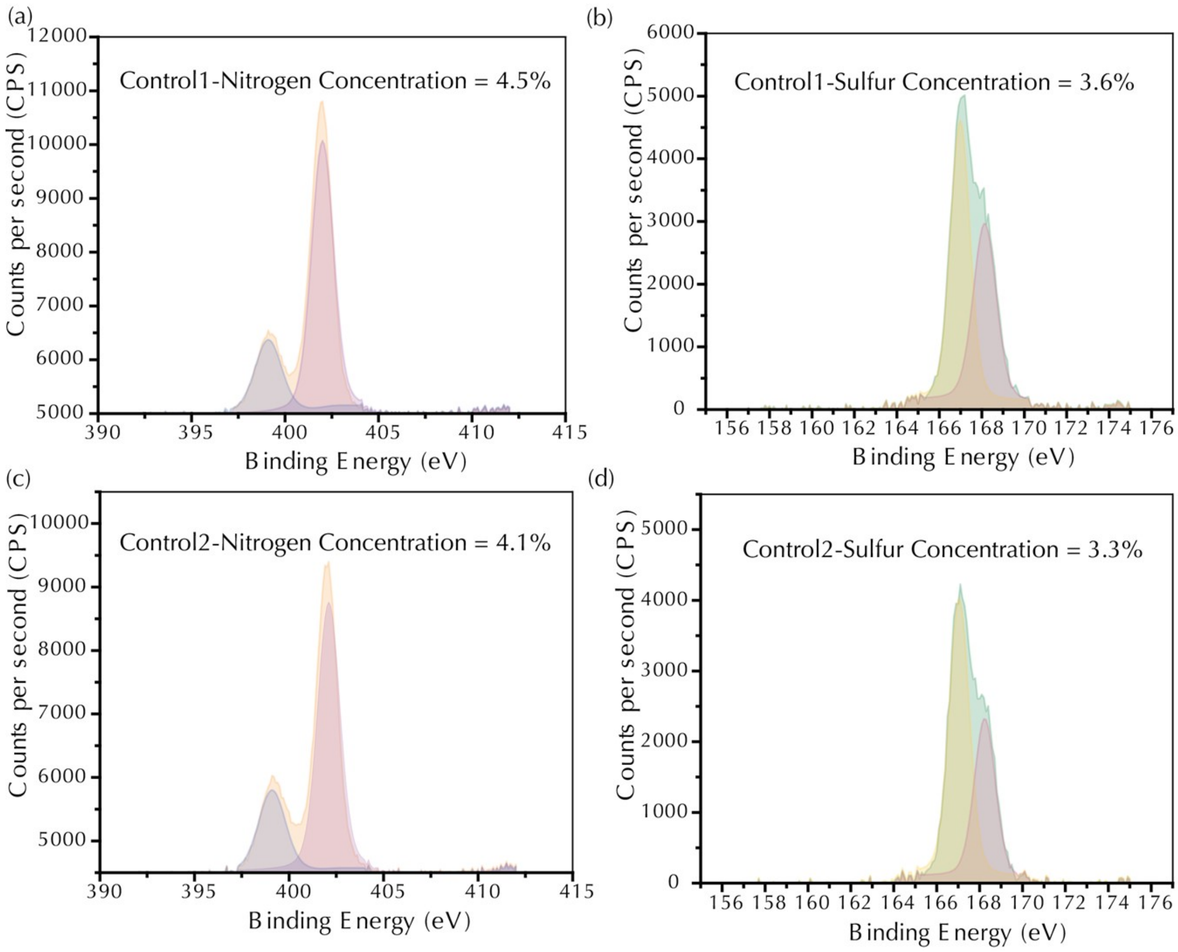

Figure S 7. X-ray photoelectron spectroscopy (XPS) analysis of the opposite sides of the control sample with no gradient.

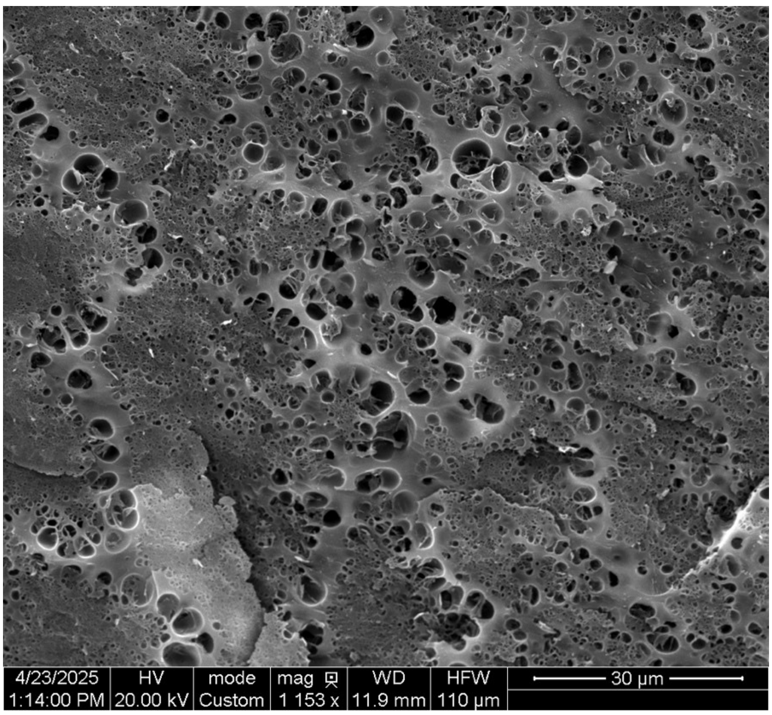

Figure S 8. SEM image from the bulk region of the gradgel far from the edges.

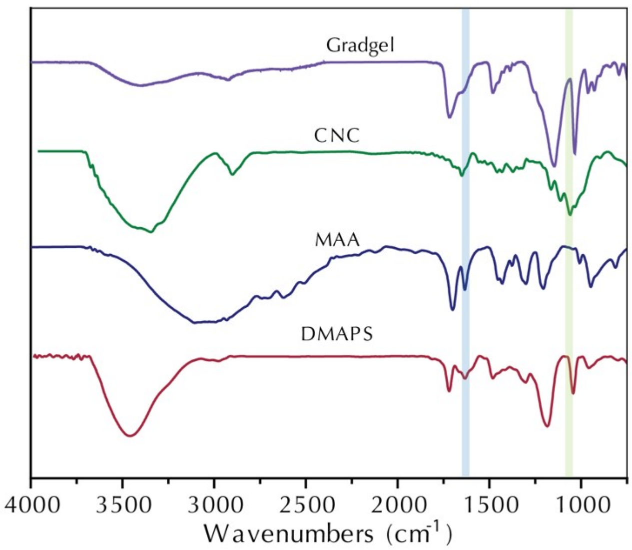

Figure S 9. FTIR spectra of the hydrogels and their components. The C=C stretching and C-O stretching regions are highlighted in blue and green, respectively.

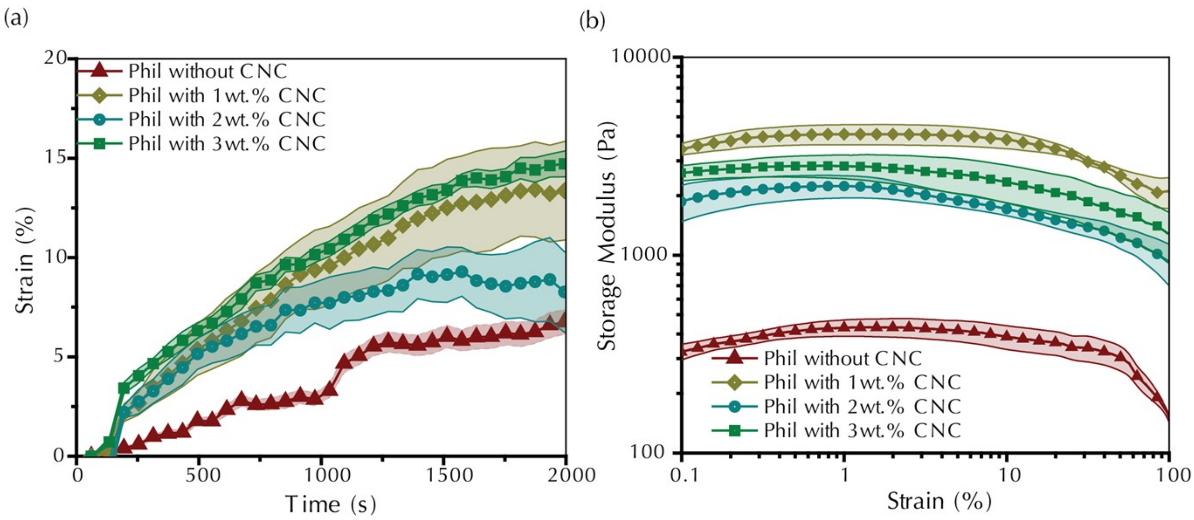

Figure S 10. The effect of CNC concentration on mechanical properties of the DMAPS-rich side of the gradgels.

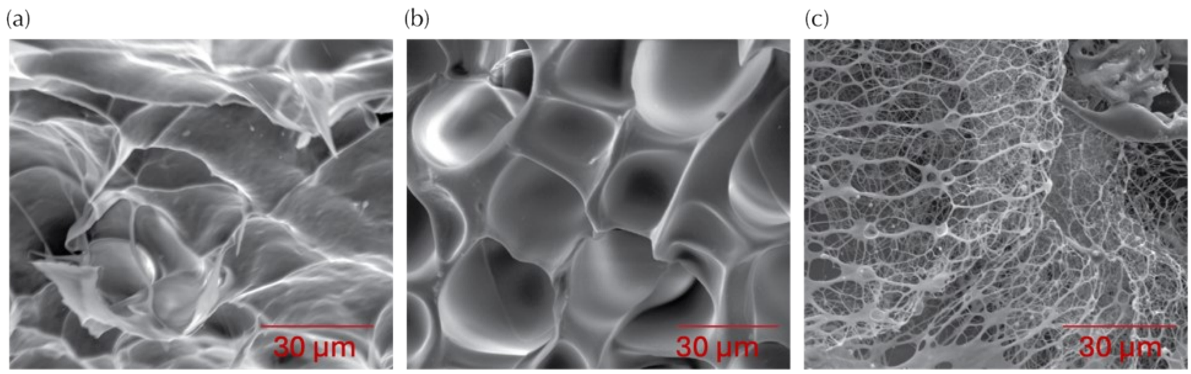

Figure S 11. SEM images of DMAPS-rich side of the gradgels with a) 1 wt.% b) 2 wt.%, and c) 3 wt.% CNC.

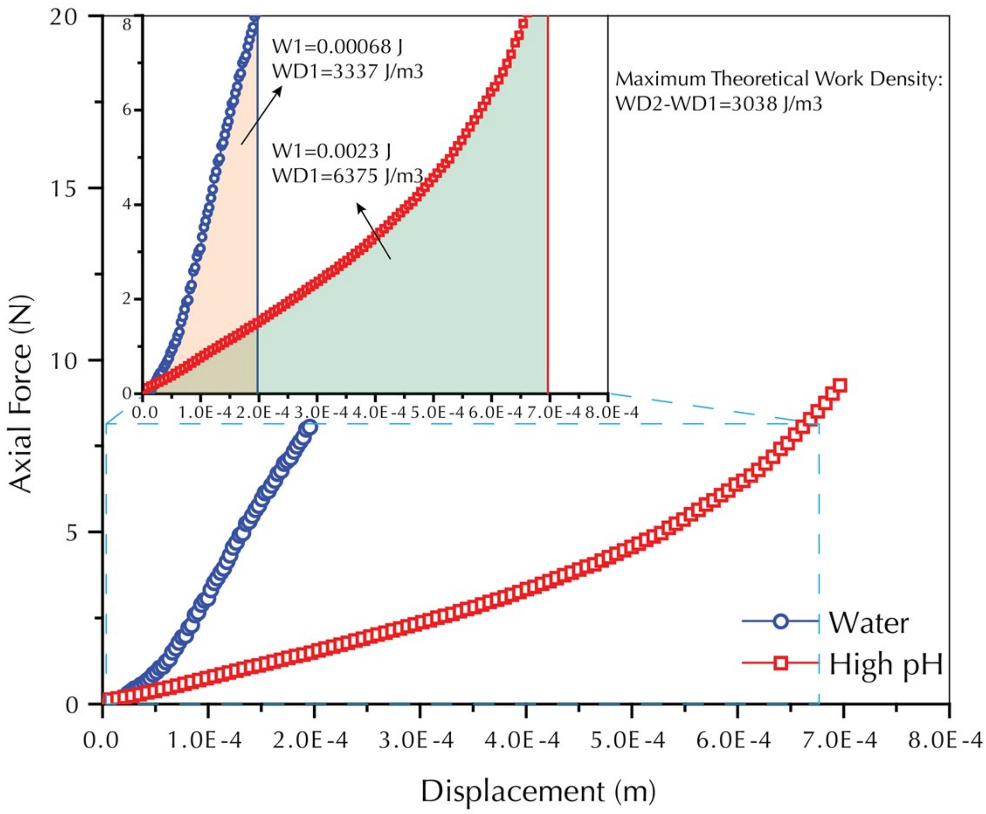

Figure S 12. Work density of the gradgel obtained from the difference between the area under the Force-Displacement curves for the gradgels swelled in water and high pH solution normalized by the initial volume of the samples.

Description of Supplementary Videos

Video S1. The video shows the bending of the gradgel with 3 to 1 weight ratio of DMAPS to MAA.

Video S2. The video shows the bending of the gradgel with 1 to 2 weight ratio of DMAPS to MAA.

Video S3. The video shows the bending of the gradgel with 1 wt.% CNC.

Video S4. The video shows the bending of the gradgel with 3 wt.% CNC.

Video S5. The video shows the bending of the gradgel with the thickness of 125 μm.

Video S6. The video shows the bending of the gradgel with the thickness of 250 μm.

Video S7. The video shows the bending of the gradgel with the thickness of 500 μm.

Video S8. The video shows the deformation mode of gradgels with different aspect ratios.

Video S9. The video shows the deformation of control hydrogels with different aspect ratios.

Video S10. The video shows the cyclic bending and relaxation of a gradgel sample.

Video S11. The video shows the S-shape deformation of a gradgel.

Video S12. The video shows the M-shape deformation of a gradgel.

Video S13. The video shows the two-sides bending deformation of a gradgel.

Video S14. The video shows the rapid forward movement of a hydrogel octopus.