

Supporting Information

Viscoelastic Hydrogel Microfibers Exploiting Cucurbit[8]uril Host-Guest Chemistry and Microfluidics

Zhi-Jun Meng,^{†,‡} Ji Liu,^{,⊥} Ziyi Yu,[‡] Hantao Zhou,[#] Xu Deng,^{*,†} Chris Abell^{*,‡} and*

Oren A. Scherman^{,§}*

[†] Institute of Fundamental and Frontier Sciences, University of Electronic Science and Technology of China, Chengdu, 610054, China.

[‡] Department of Chemistry, University of Cambridge, Lensfield Road, Cambridge, CB2 1EW, UK.

[⊥] Department of Mechanical and Energy Engineering, Southern University of Science and Technology, Shenzhen 518055, P. R. China.

[#] College of Chemistry and Molecular Engineering, Peking University, Beijing 100871, P. R. China.

[§] Melville Laboratory for Polymer Synthesis, Department of Chemistry, University of Cambridge, Lensfield Road, Cambridge, CB2 1EW, UK.

Part I. Supplementary experimental details

Part II. Supplementary Results

Part III. Supplementary Movies

Part I. Supplementary experimental details

Fabrication of microcapillary microfluidic device

The coaxial microcapillary microfluidic devices are fabricated following previous protocol.¹ Briefly, a cylinder capillary (inner diameter 700 μm , outer diameter 1 mm) was inserted into a square capillary (inner diameter 1mm, length 5 cm), before bonded onto a glass slide. The two ends of the cylinder microcapillary were connected with a steel tube to protect the microcapillary. Dispensing needles as inlet/outlet for the ice-cold water were then connected at the junctions between capillaries using Epoxy resin.

Preparation of the monomer precursor solution for the hydrogel microfibres

To fabricate the cucurbit[8]uril (CB[8])-contained DN hydrogel microfibre, agar (20 mg, congealing temperature 30-35 $^{\circ}\text{C}$, A7921 Sigma), Acrylamide (AAM, 250 mg), 2-hydroxy-1-[4-(2-hydroxyethoxy)phenyl]-1-methyl-1-propane (0.0059 g, Irgacure®2959, BASF, 1 mol% of AAM), CB[8] (0.014 g), 1-benzyl-3-vinylimidazolium bromide (BVIm, 0.0067 g) and milli-Q H_2O (1 mL) were added into a vial to form a homogeneous solution under a heating condition. Controls of the hydrogel microfibres were fabricated following the same procedure.

Part II. Supplementary results

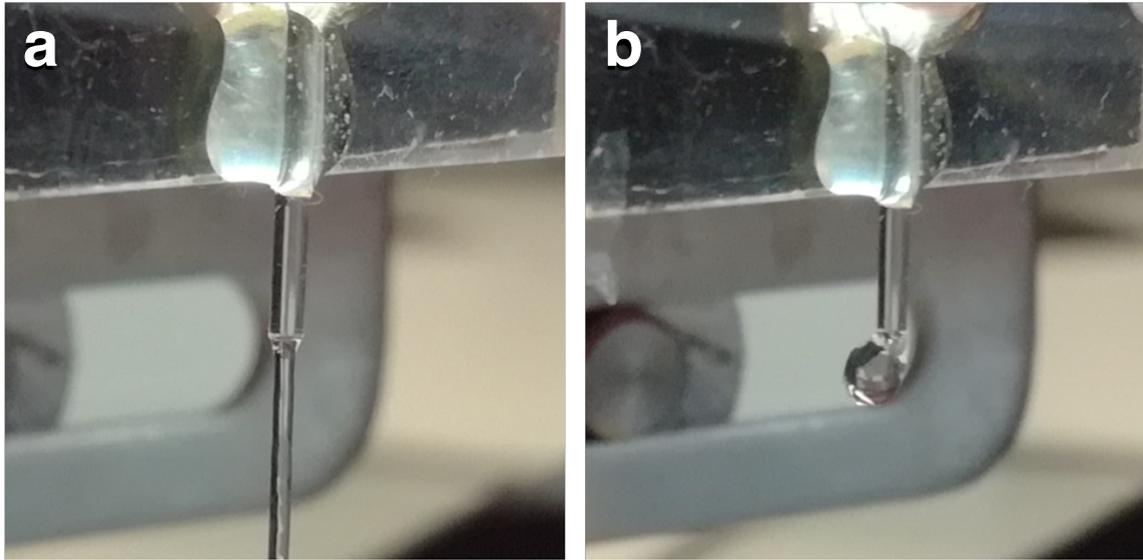


Figure S1. Photographs showing the microfiber fabrication with (a) or without (b) the heat-exchanging process. The monomer precursor solution can be immediately cooled down, inducing gelation of the first agar network.

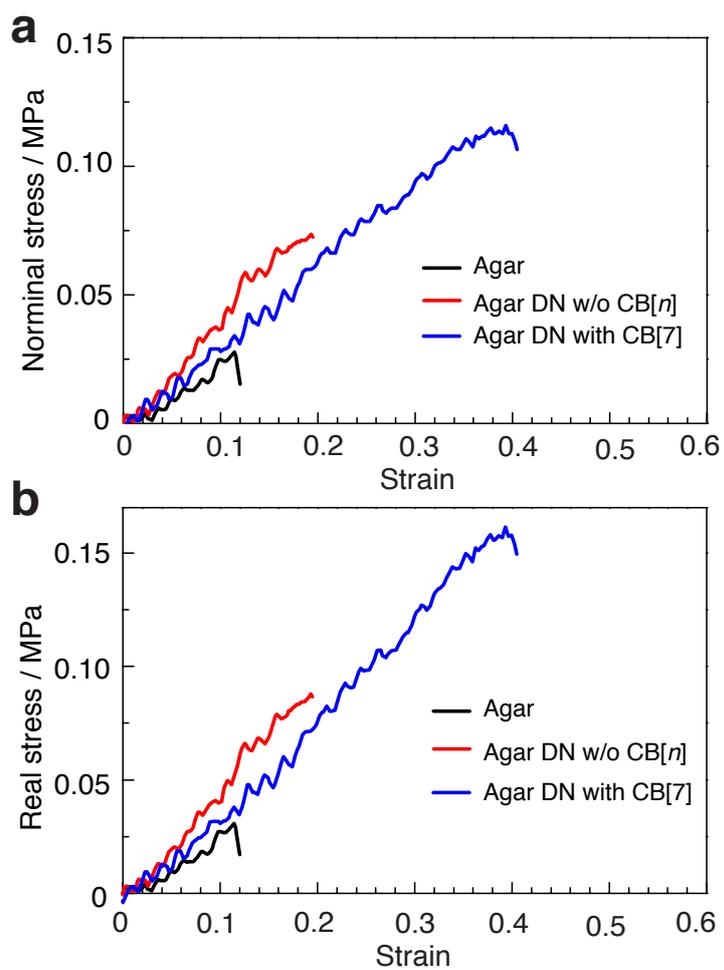


Figure S2. Tensile test curves of the hydrogel microfiber controls: (a) nominal stress-strain curve and (b) true stress-strain curve (deformation rate of 100 mm min^{-1}).

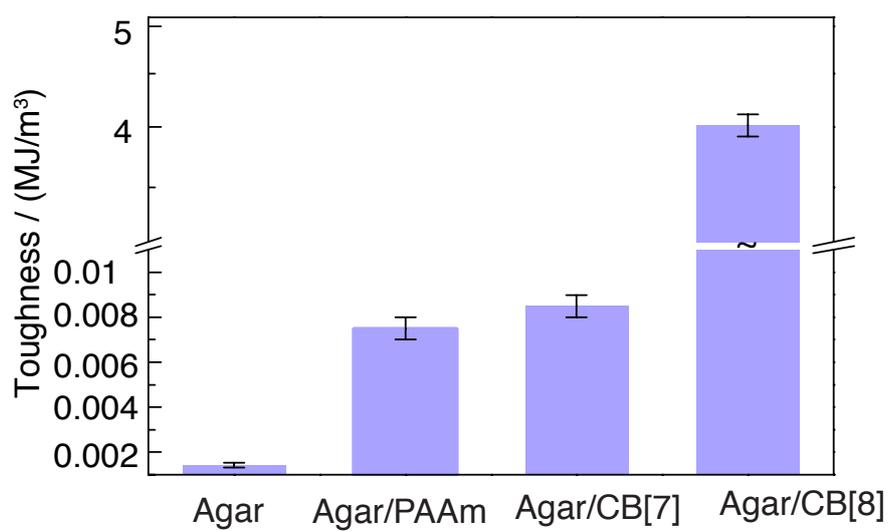


Figure S3. Toughness of the DN hydrogel microfibers including controls without CB[n], with CB[7], as well as a pure agar fiber (deformation rate of 100 mm min^{-1}).

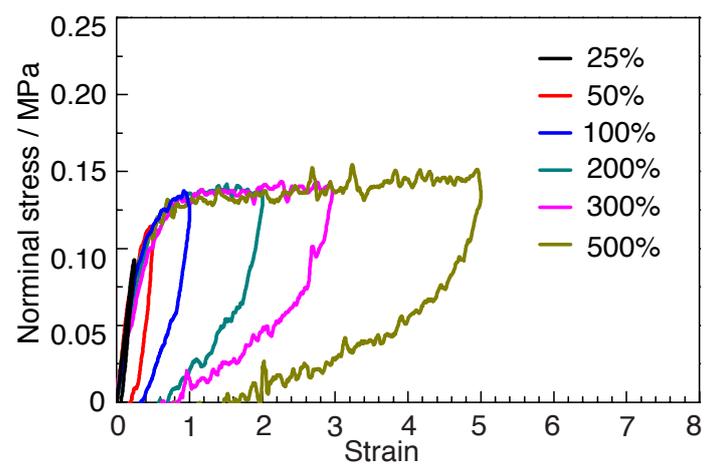


Figure S4. Cyclic tensile tests of the DN hydrogel microfibers at various strains (25-500%, deformation rate of 100 mm min^{-1}).

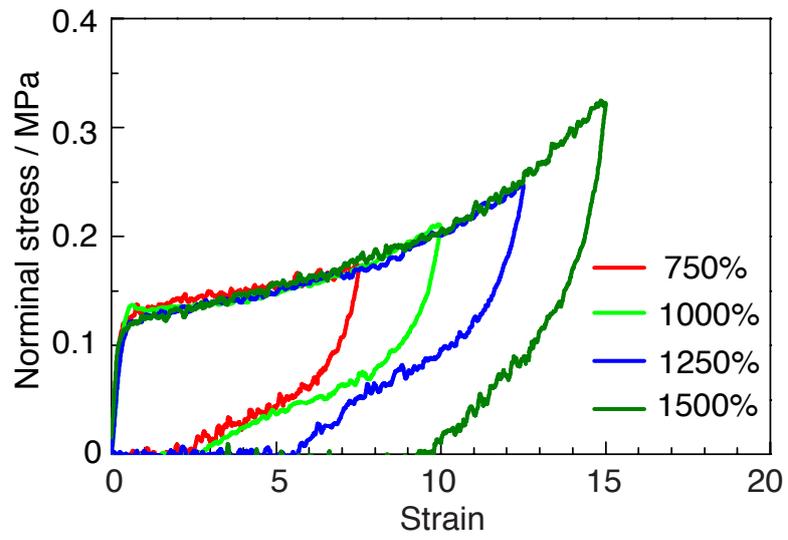


Figure S5. Cyclic tensile tests of the DN hydrogel microfibers at various strains (750-1500%, deformation rate of 100 mm min^{-1}).

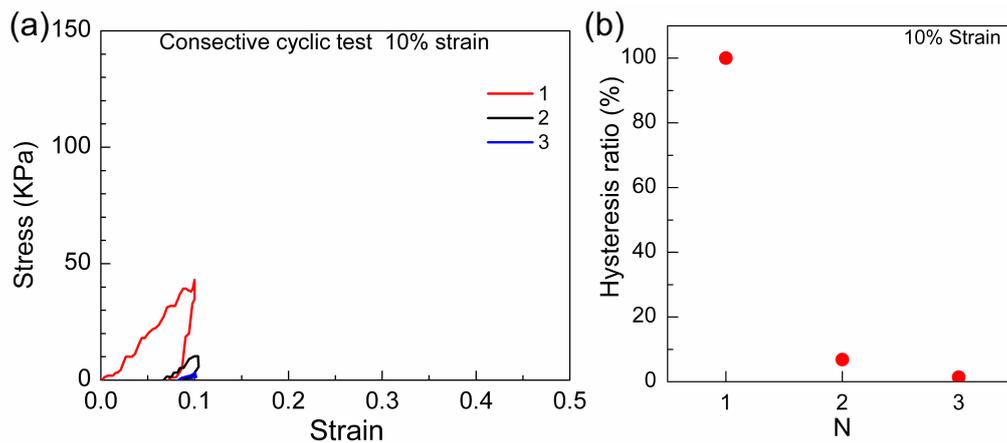


Figure S6. (a) Consecutive cyclic tensile tests of the Agar DN w/o CB[n] hydrogel microfibers at 10% strain. (b) Hysteresis ratio of the Agar DN w/o CB[n] hydrogel microfibers for each cycle during the consecutive cyclic tensile tests at 10% strains. The deformation rates are 100 mm min^{-1} .

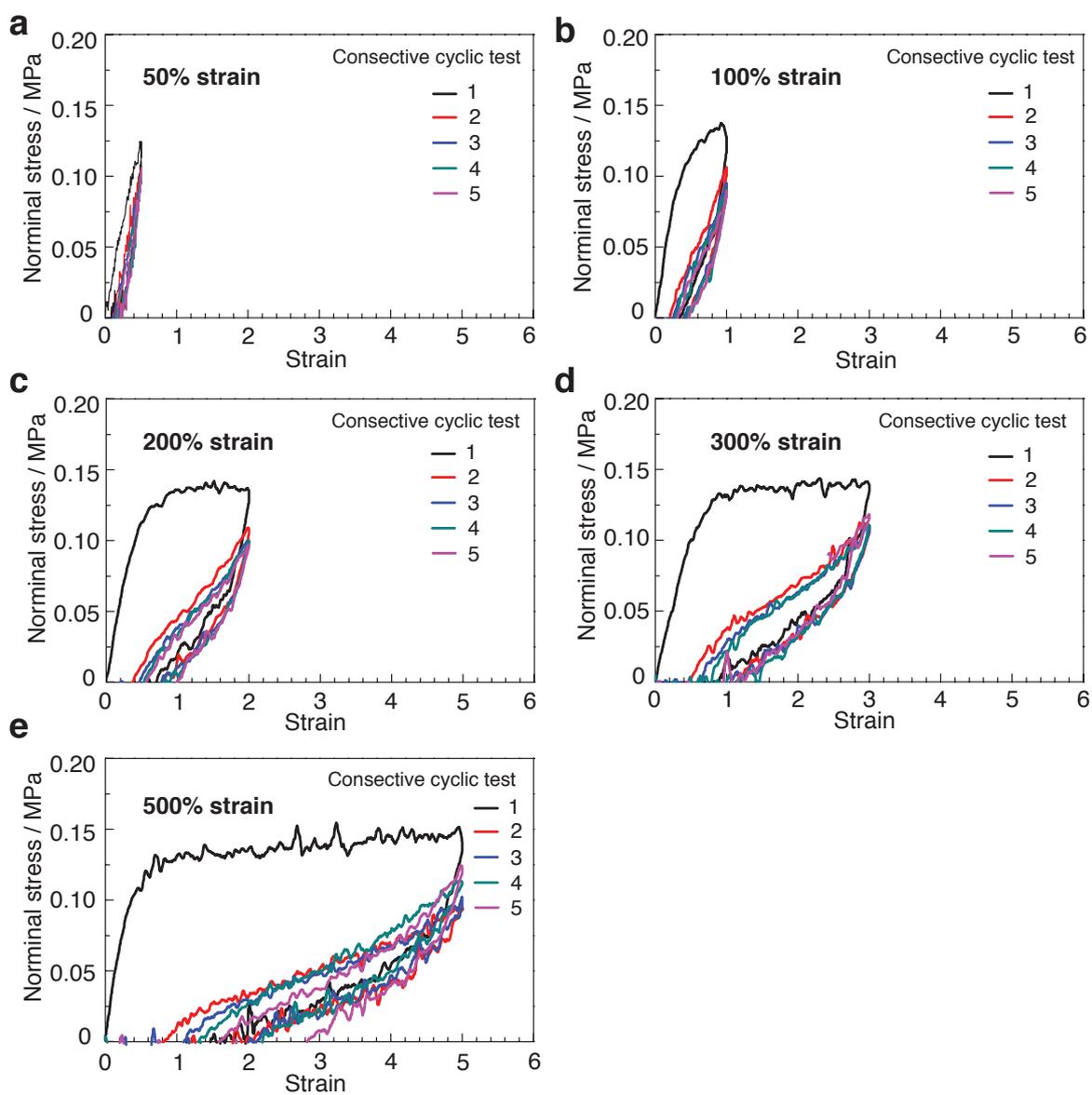


Figure S7. Consecutive cyclic tensile tests of the DN hydrogel microfibers at various strains (25-500%, deformation rate of 100 mm min^{-1}).

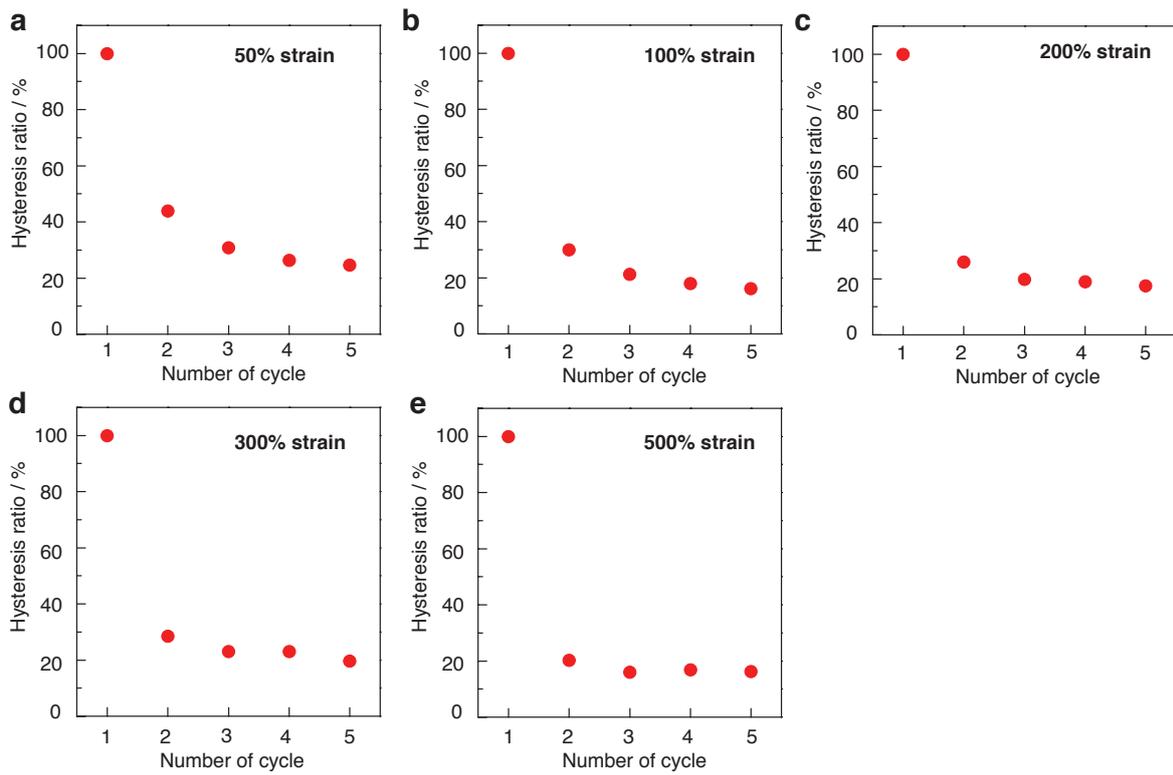


Figure S8. Hysteresis ratio of the DN hydrogel microfibers for each cycle during the consecutive cyclic tensile tests at various strains (25-500%, deformation rate of 100 mm min^{-1}).

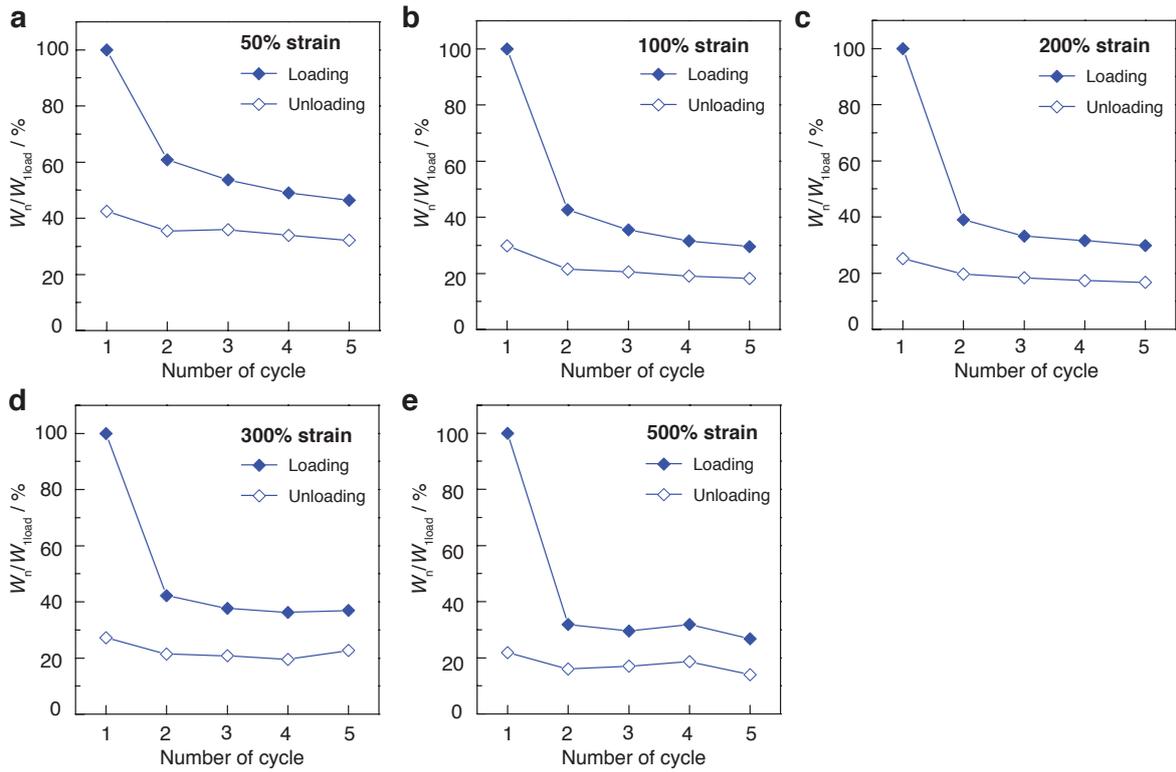


Figure S9. Resilience of the DN hydrogel microfibers for each cycle during the consecutive cyclic tensile tests at various strains (25-500%, deformation rate of 100 mm min^{-1}). Resilience was quantified by the ratio of work (W_N) of subsequent loadings and unloadings to the work of the first loading ($W_{N=1,load}$).

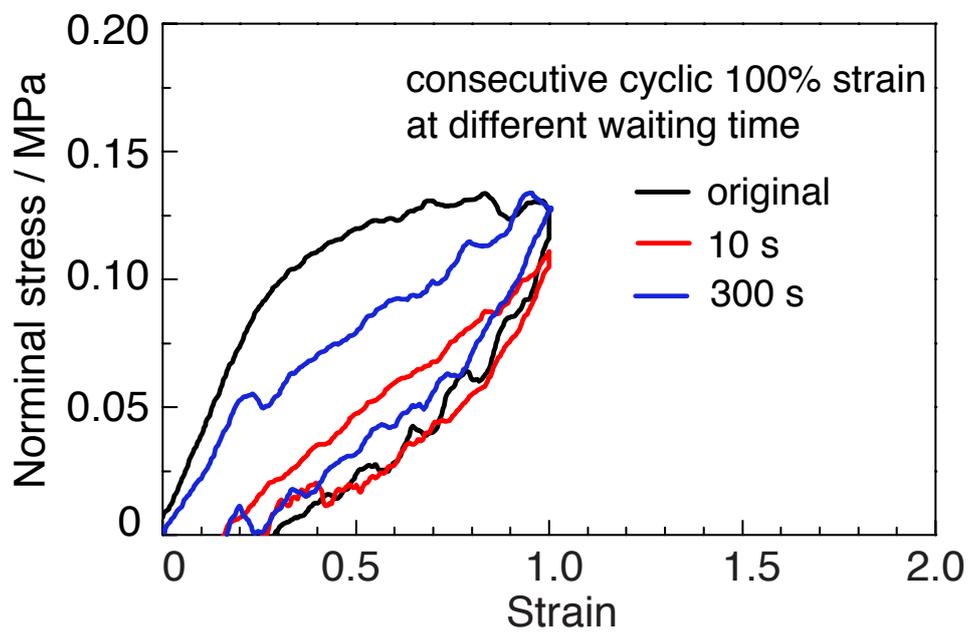


Figure S10. Consecutive cyclic tensile tests with different at different waiting time between two cycles. (strain of 100%, deformation rate of 100 mm min^{-1}).

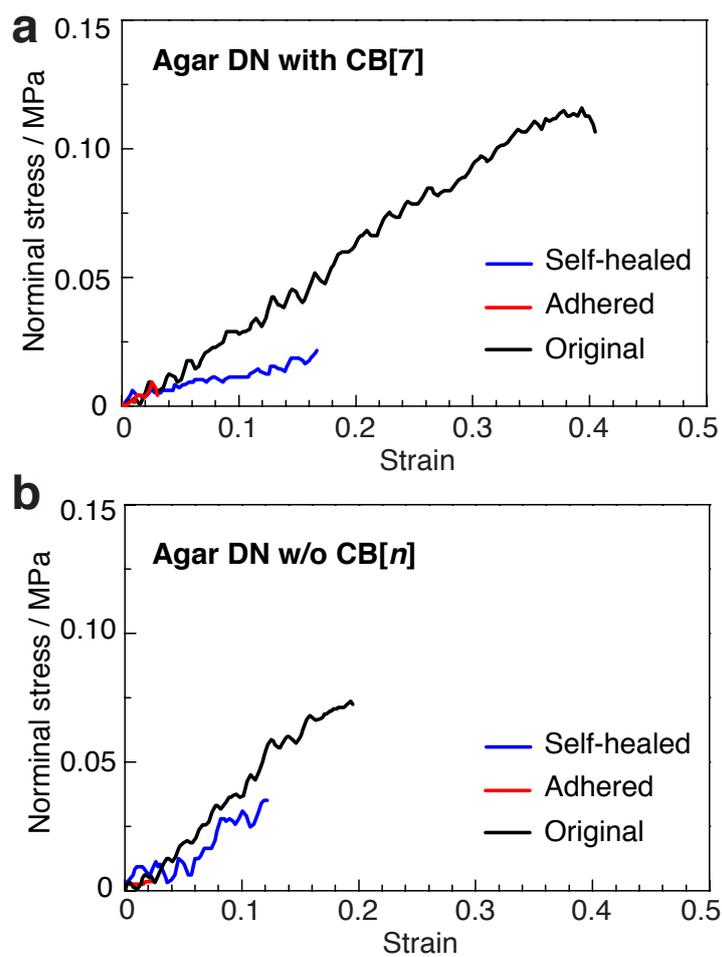


Figure S11. Self-healing performance of the DN hydrogel microfiber controls with CB[7] (**a**) or without CB[n] (**b**).

Part III. Supplementary Movies S1 and S2

Movie 1

Manual tensile test with the DN hydrogel microfibres.

Movie 2

Manual tensile test with the DN hydrogel microfibres control in the presence of CB[7].

References

1. Z.-J. Meng, W. Wang, R. Xie, X.-J. Ju, Z. Liu, L.-Y. Chu, *Lab Chip* **2016**, 16, 2673–2681.
2. M. Y. Guo, L. M. Pitet, H. M. Wyss, M. Vos, P. Y. W. Dankers, E. W. Meijer, *J. Am. Chem. Soc.* 2014, **136**, 6969.