Supporting Information for

Photodegradable polyacrylamide tanglemers enable spatiotemporal control over chain lengthening in high-strength and low-hysteresis hydrogels

Joshua S. Lee[1,*], Bruce E. Kirkpatrick[1,2,3,*], Abhishek P. Dhand[4], Lea Pearl Hibbard[1], Benjamin R. Nelson[1,2], Nathaniel P. Skillin[1,2,3], Makayla C. Johnson[1], Dilara Batan[2,5], Benjamin D. Fairbanks[1], Timothy J. White[1,6], Christopher N. Bowman[1,6], Jason A. Burdick[1,2,6], Kristi S. Anseth[1,2,6,#]

*equal contribution

[#]corresponding author, kristi.anseth@colorado.edu

[1]Department of Chemical and Biological Engineering, University of Colorado Boulder

[2]BioFrontiers Institute, University of Colorado Boulder

[3]Medical Scientist Training Program, University of Colorado Anschutz Medical Campus

[4]Department of Bioengineering, University of Pennsylvania

[5]Department of Biochemistry, University of Colorado Boulder

[6]Materials Science and Engineering Program, University of Colorado Boulder

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Supplementary Figures

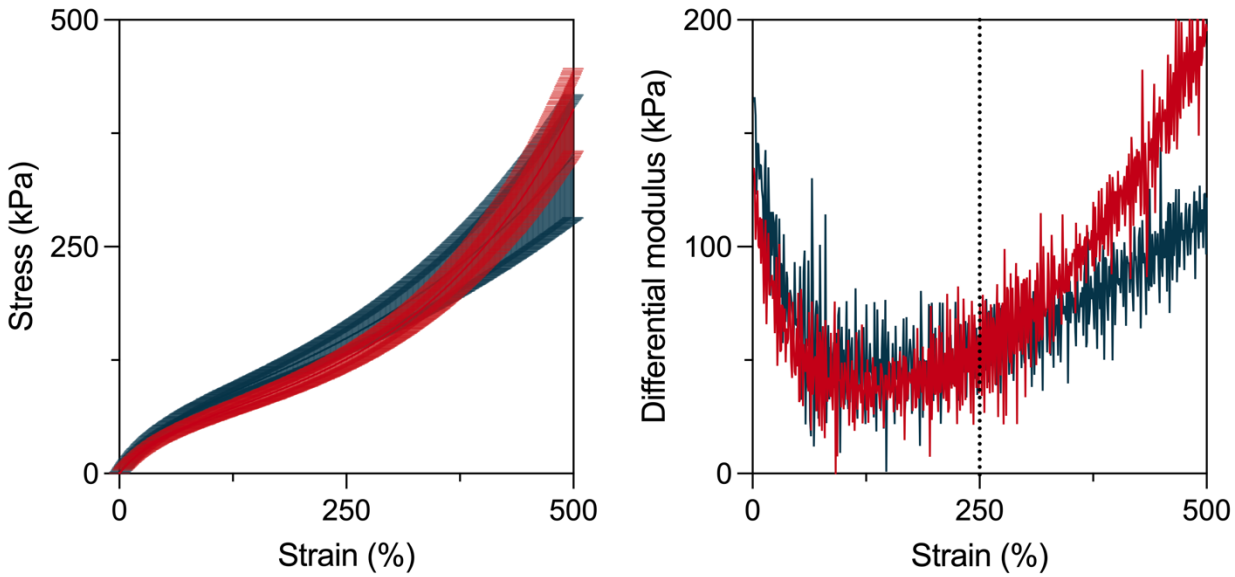

Fig. S1 | The first derivative of the stress-strain curve (differential modulus) illustrates strain-stiffening behavior that occurs after 250% strain, marking the limit of extensibility after which physical entanglements begin to accumulate at covalent crosslinkers.

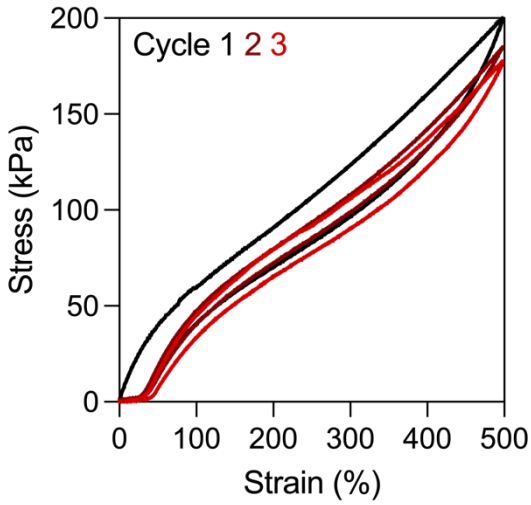

Fig. S2 | Cyclic loading to 500% strain and unloading of diPDA-crosslinked tanglemers identifies increased hysteresis (10-20%) compared to lower strains.

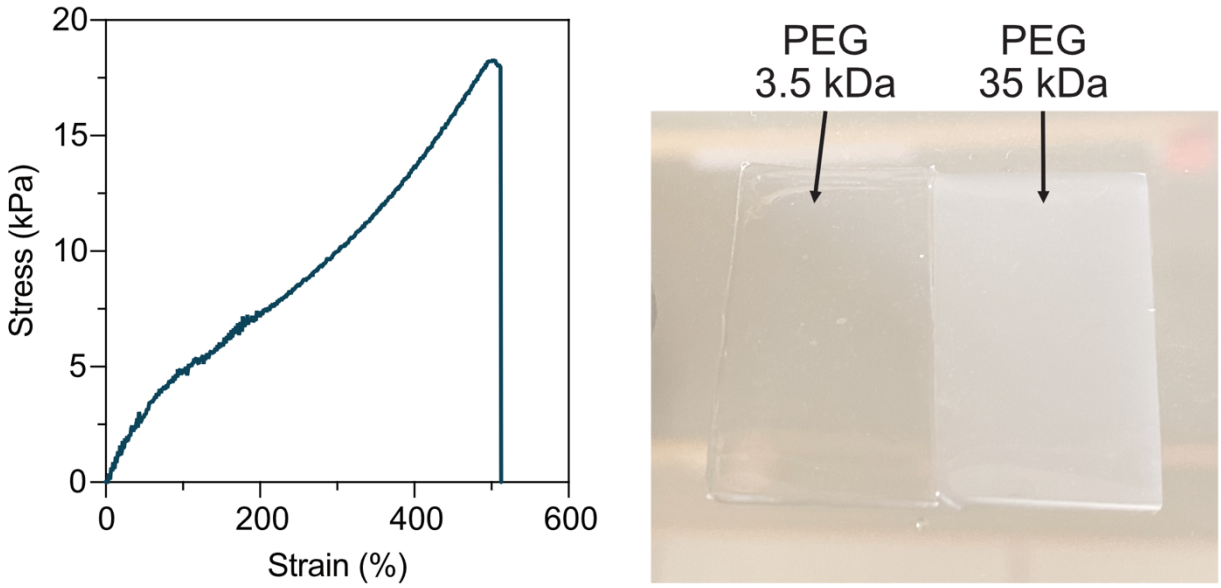

Fig. S3 | Tanglemers prepared from PEG 35 kDa diacrylate are soft, have low fracture strength, and show visible phase separation (indicated by gel opacity).

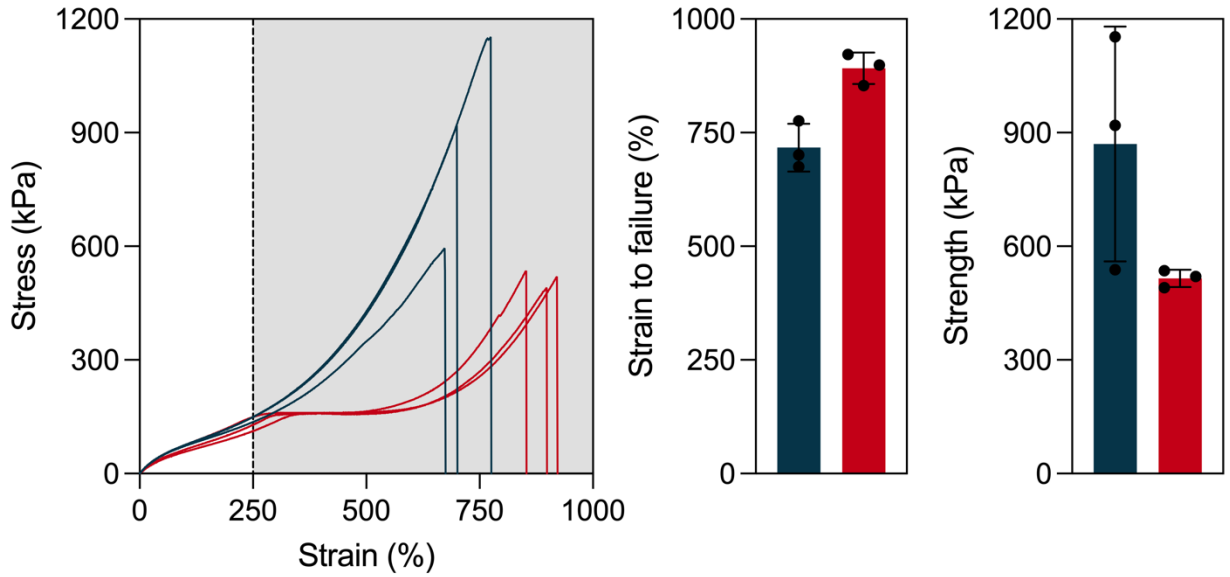

Fig. S4 | *In situ* photodegradation of tanglemers under strain increases extensibility but reduces fracture strength.

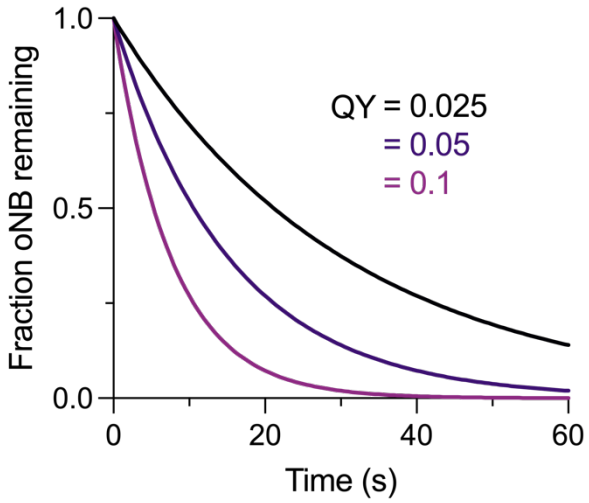

Fig. S5 | Calculated oNB degradation with varied quantum yield (QY), assuming first-order kinetics, a molar extinction coefficient of 4300 M^{-1} cm^{-1} at 365 nm, PEG-diPDA concentration of 0.0125 wt%, and an intensity of 100 mW/cm^2.

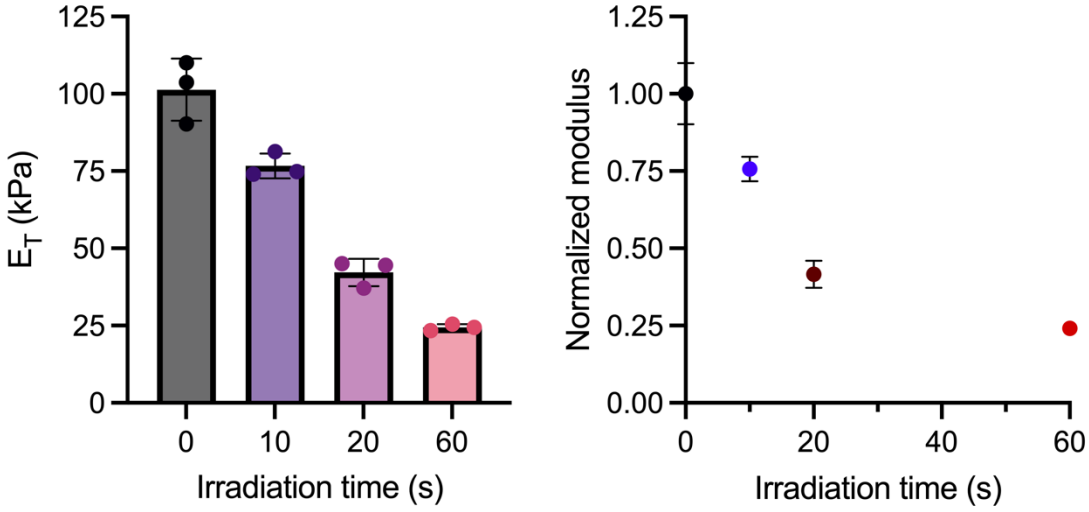

Fig. S6 | Raw data of oNB tanglemer photodegradation (left) and modulus normalized to 0s of irradiation (right).

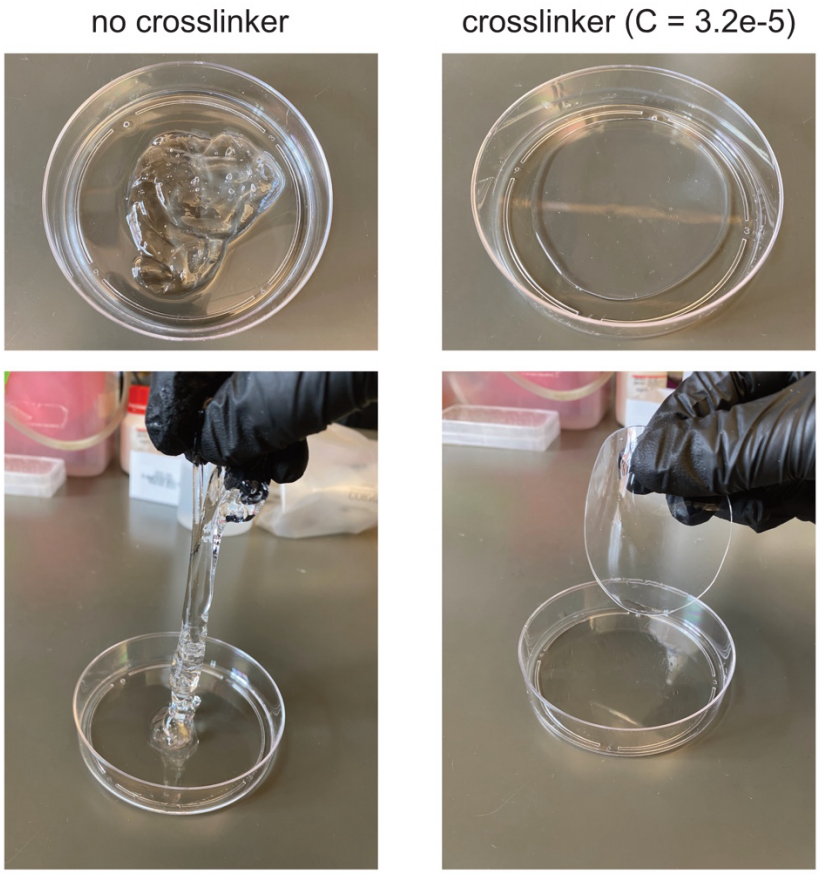

Fig. S7 | Images of tanglemers prepared with and without crosslinker, showing dramatic swelling in samples without entanglement-stabilizing covalent crosslinks.

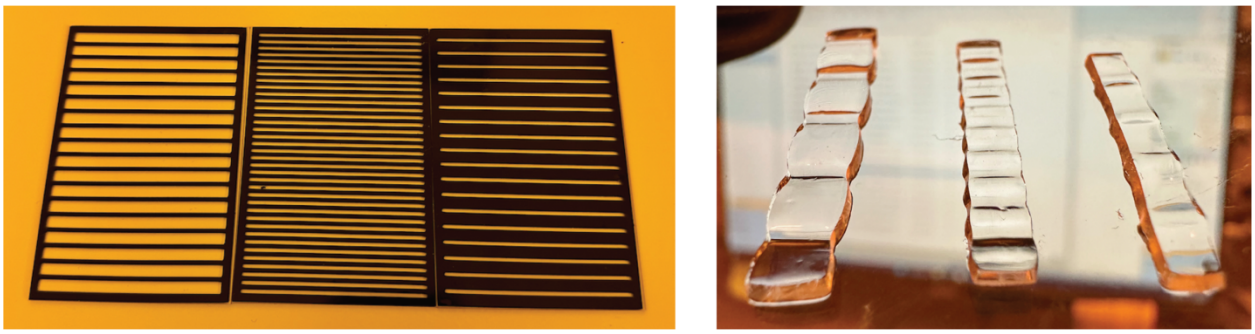

Fig. S8 | Vinyl photomasks affixed to glass slides (left) and tanglemers patterned using these masks and equilibrated following light exposure (right), showing varied swelling depending on spatial exposure.

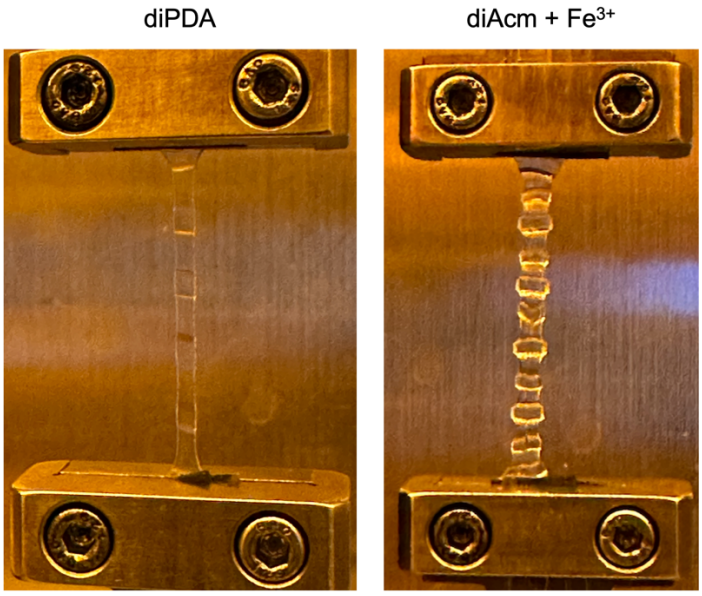

Fig. S9 | Macro image of tanglemers spatially patterned by either oNB cleavage or Fe^{3+} oxidation.

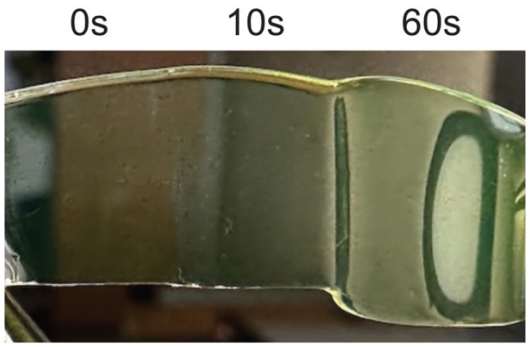

Fig. S10 | Macro image of tanglemer exposed to 365 nm light at 100 mW/cm^2 for varied times (used for FRAP characterization), showing varied swelling depending on temporal exposure.

Movie Captions

Movie S1. | Reduced strain-induced birefringence is observed in diPDA-crosslinked tanglemers following photodegradation.

Movie S2. | Through-thickness photo-oxidation of diAcm-crosslinked tanglemers results in complete degradation of the network.

Movie S3. | Reverse gelation of pre-irradiated diPDA-crosslinked tanglemers via photo-oxidation.