Supporting Information for

Highly stretchable DNA/clay hydrogels with self-healing ability

Ahmet T. Uzumcu, Orhan Guney,[*] and Oguz Okay[*]

Istanbul Technical University, Departments of Chemistry and Polymer Science & Technology, 34469 Maslak, Istanbul, Turkey

Corresponding Authors:

*(O.O.) E-mail: okayo@itu.edu.tr

*(O.G.) E-mail: oguney@itu.edu.tr

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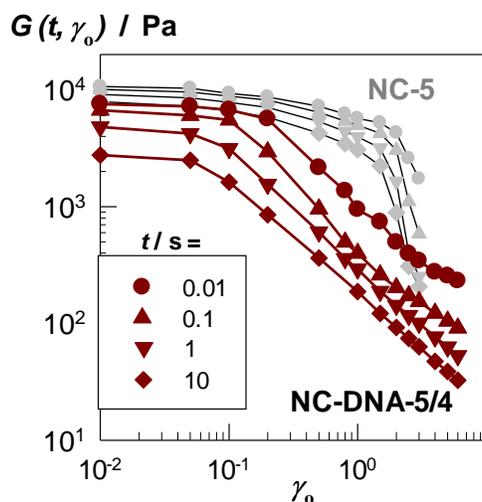**Figure S1.** Relaxation moduli $G(t, \gamma_o)$ of NC-DNA-5/4 (dark red symbols) and NC-5 hydrogels (gray symbols) shown as a function of strain γ_o at different time scales t.[a] Temperature = 25 °C.

[a] The tests were carried out by application of a shear deformation on the gel samples of predetermined strain amplitude γ_o and monitoring the resulting stress $\sigma(t, \gamma_o)$ as a function of time. Here, we present the relaxation modulus $G(t, \gamma_o)$ as a function of γ_o at various relaxation times t. The experiments were conducted with increasing γ_o from 0.01 to 10.
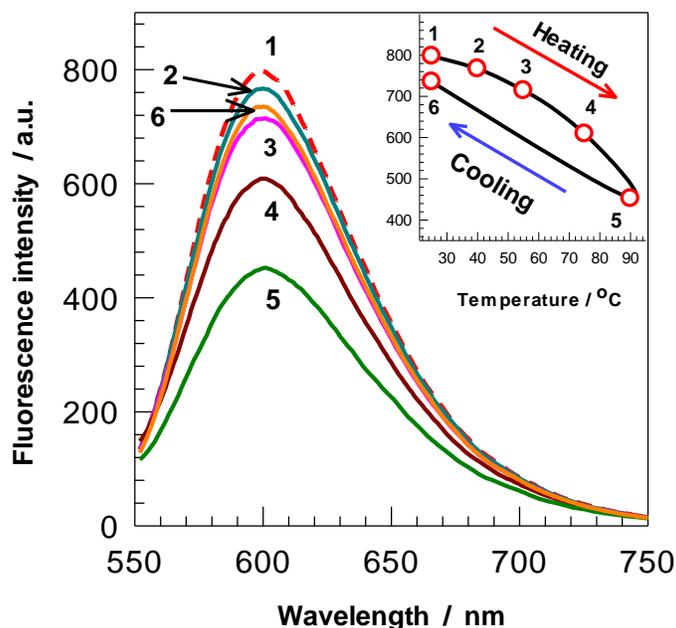

Figure S2. Fluorescence spectra of EtBr in NC-DNA-5/4 hydrogel prepared without XLG at various temperatures. The inset shows emission intensity of EtBr at 600 nm plotted against the temperature. The numbers in the inset correspond to those given on the spectra.

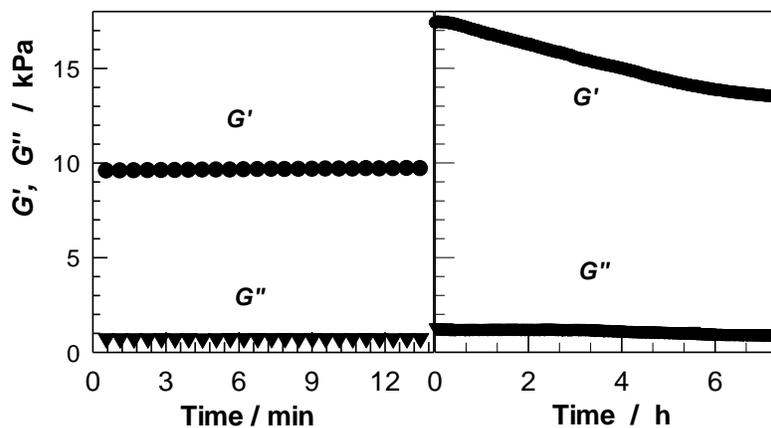**Figure S3**. G' (circles) and G'' (triangles) for NC-5 hydrogel before (left) and after the heating-cooling cycle (right) shown as a function of time. Temperature = 25 °C. $\omega = 6.3$ rad·s^{-1}. $\gamma_0 = 0.01$.

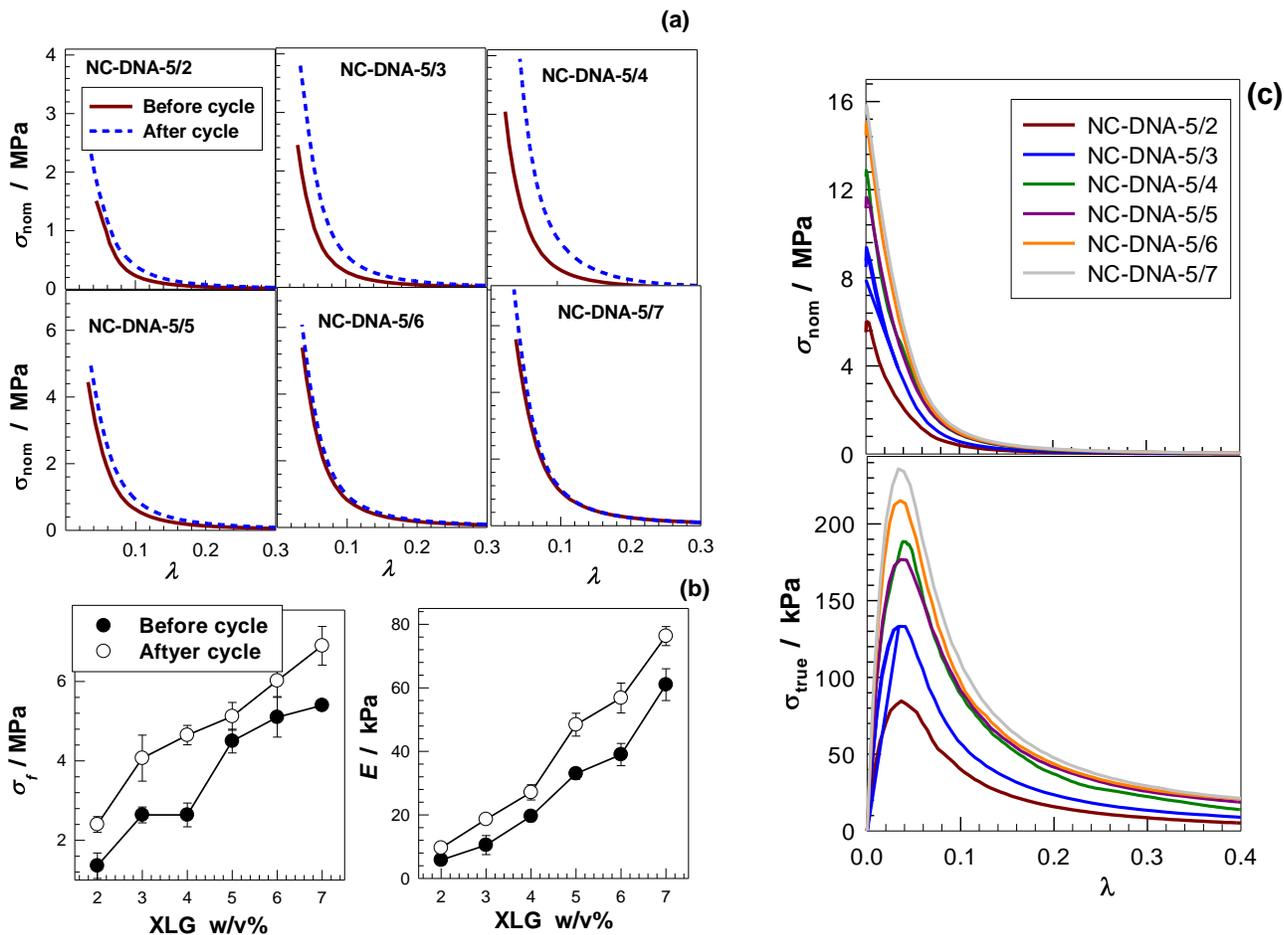**Figure S4.** (**a**): Compressive nominal stress σ_{nom} plotted against the strain λ (deformed sample length / initial sample length) for NC-DNA hydrogels before and after heating/cooling cycle.[a]

The data are shown up to the fracture point.[b] (**b**): Compressive fracture stress σ_f and Young's modulus E of NC-DNA hydrogels containing 2 w/v% DNA plotted against their XLG contents.[b]

[a] Compression measurements were performed on hydrogel samples in cylindrical shape of about 5 ± 0.2 mm in diameter and 3.5 ± 0.5 mm in length. Compressive stress was presented by its nominal σ_{nom} and true values σ_{true}, where the latter is the force per cross-sectional area of the deformed gel specimen and, assuming isotropic deformation during compression, it is given by $\sigma_{true} = \lambda \, \sigma_{nom}$.

[b] We observed that the compressive fracture stress σ_f and strain λ_f obtained from σ_{nom} vs λ curves do not match with those obtained from σ_{true} vs λ curves. For instance, Figures S4c shows compressive stress-strain curves of NC-DNA hydrogel samples after heating/cooling cycle as the dependences of σ_{nom} and σ_{true} on λ. The apparent fracture strains obtained from σ_{nom} vs λ curves are at above 98% strains while the corresponding σ_{true} - λ plots pass through maxima much below these strain values. This behavior is likely a result of the hydrogel samples with microscopic cracks still supporting the stress and/or non-isotropic deformation of gel samples under large strain. Therefore, the nominal fracture stress σ_f and strain λ_f were calculated from the maxima in σ_{true} - λ plots.
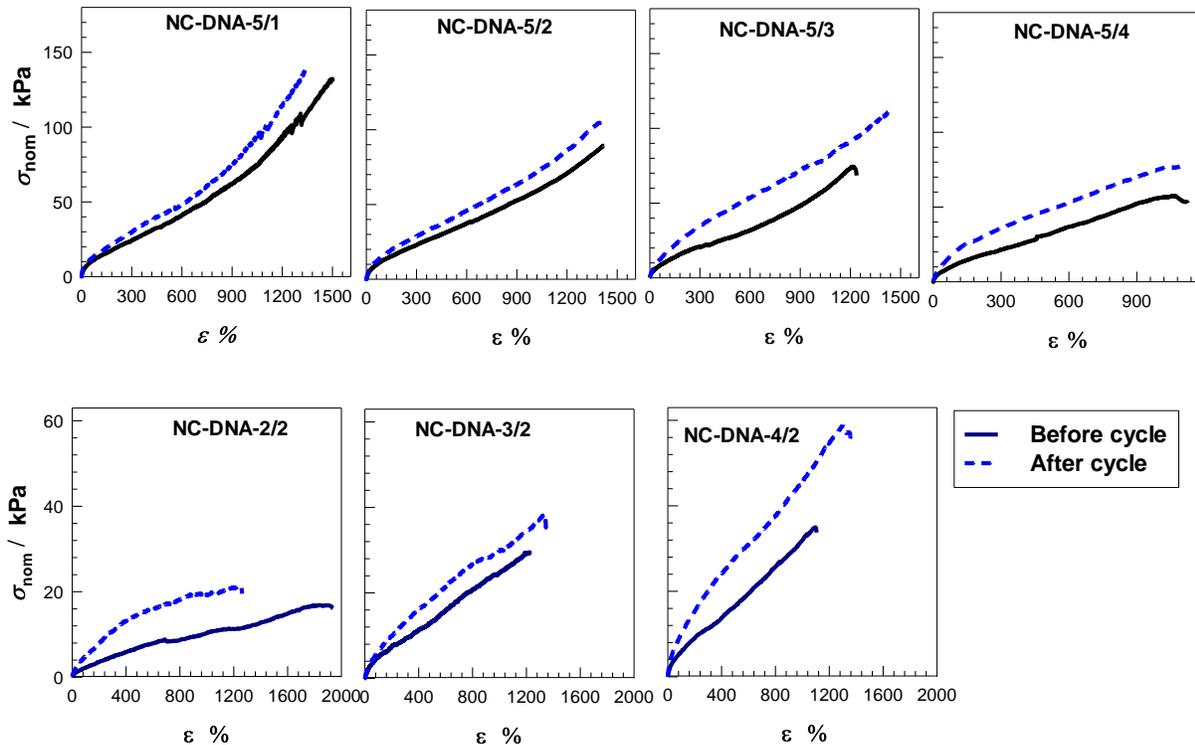

Figure S5. Tensile stress – strain curves of NC-DNA hydrogels before (solid curves) and after the heating-cooling cycle (dashed curves). Temperature = 24 ± 1 °C.
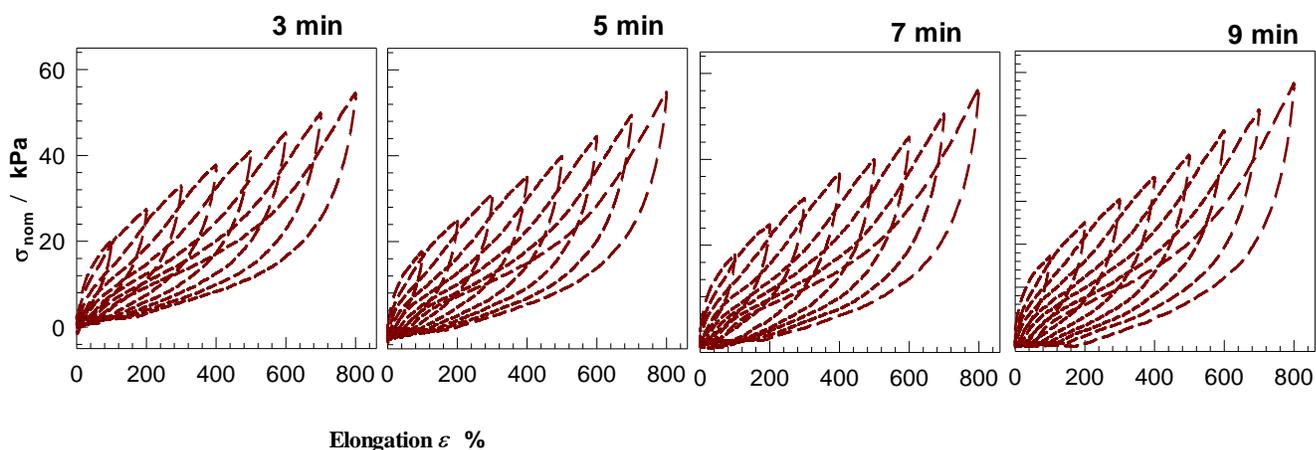
Figure S6. Cyclic elongation test results of NC-DNA-7/2 hydrogels conducted with various waiting times between successive cycles as indicated in the figures.
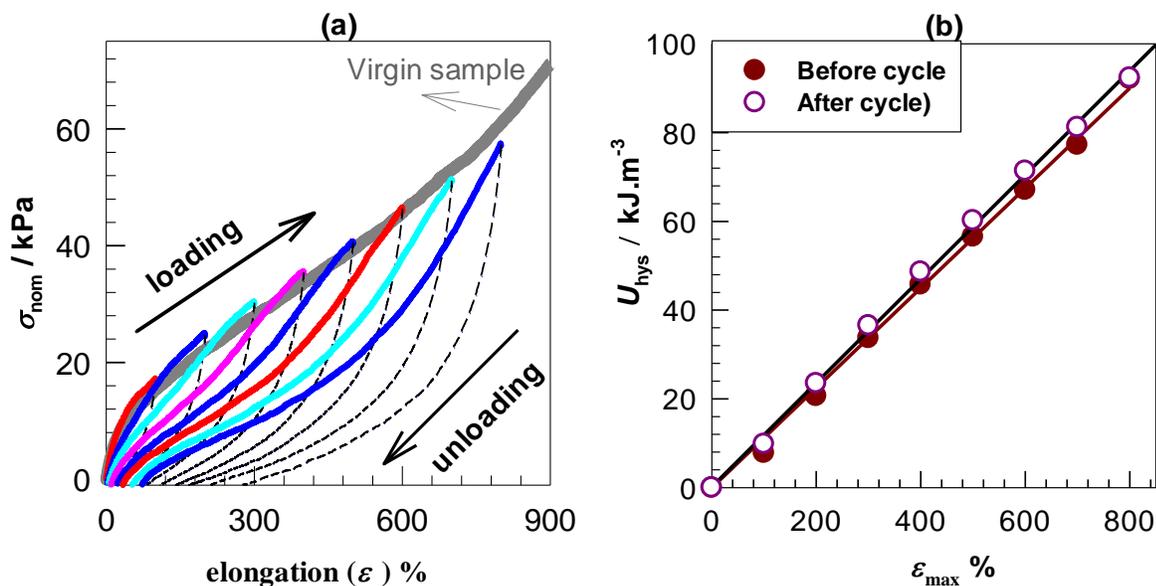

Figure S7. (**a**): Cyclic elongation test results of NC-DNA-7/2 hydrogels conducted with increasing maximum strain ε_{max} with a waiting time of 9 min between cycles. (**b**): Hysteresis energies U_{hys} of NC-DNA-7/2 hydrogel before and after heating/cooling cycle shown as a function of ε_{max}. The lines are best fit to the data. The average dissociation energies U_{xl} of the non-covalent bonds in the hydrogels are 2.9±03 and 3.0±03 kJ.mol^{-1} before and after the cycle.