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Supporting Information

Unique Crack Propagation of Double Network Hydrogels under High Stretch

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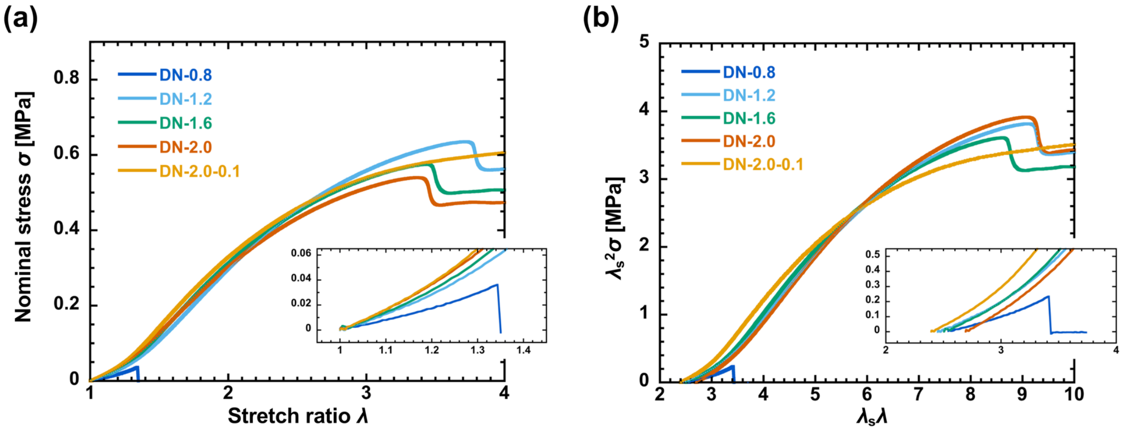

Fig. S1. (a) Uniaxial tensile stress–stretch ratio curves of all DN gels in relatively small stretching regimes. (b) Curves normalized by the pre-stretch ratio of the first network λ_s. The inset shows a magnified image of the curves of brittle DN-0.8.

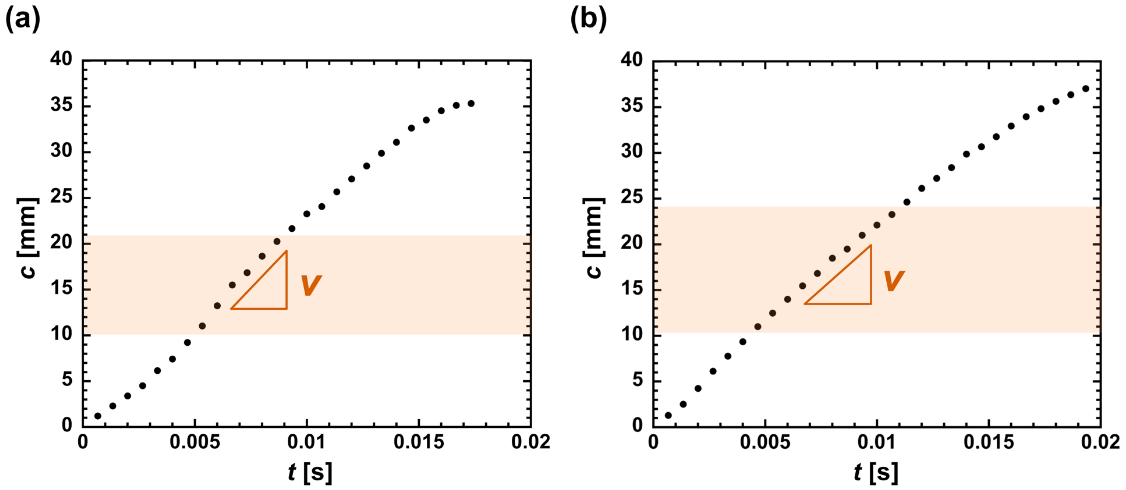

Fig. S2. Representative relationship between time t and cut length c during crack propagation in the case of (a) the fast-mode crack propagation of SN-2.0 ($G = 5.23 \times 10^2$ J/m^2, $v = 2.7$ m/s) and (b) the fast-mode crack propagation of DN-2.0 ($G = 1.63 \times 10^3$ J/m^2, $v = 2.0$ m/s). The crack growth velocity v was estimated from the region highlighted in orange where the crack propagated at a constant speed.

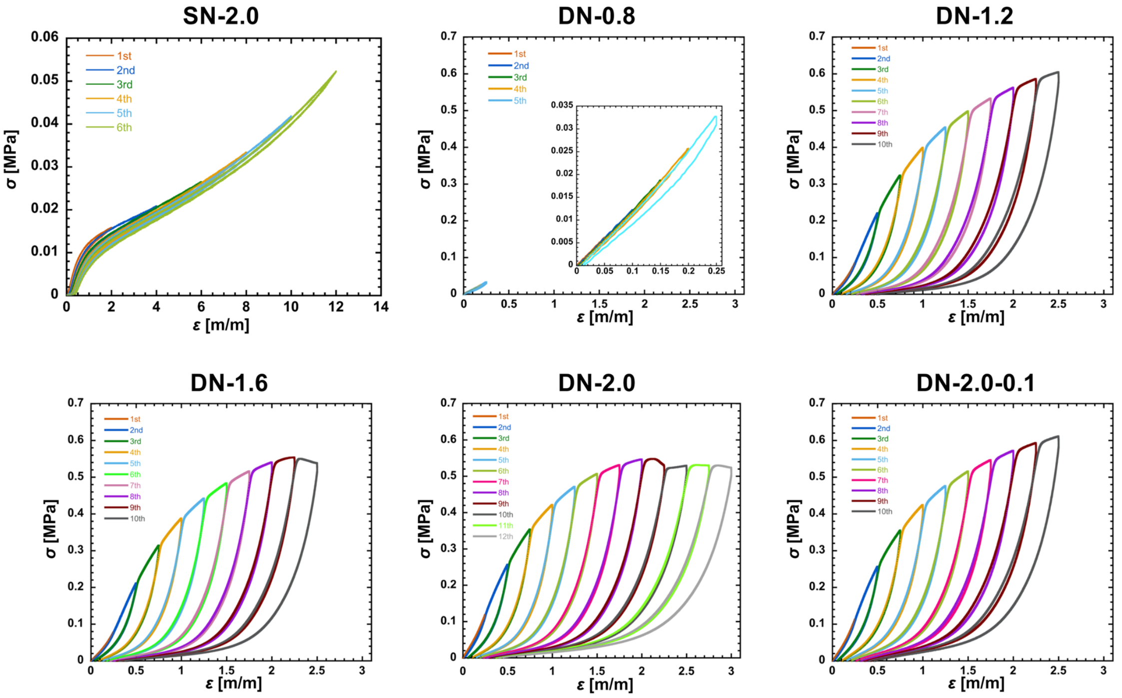

Fig. S3. Cyclic tensile stress–strain curves of the unnotched samples measured in pure shear geometries, as shown in Fig. 1a. The strain ε ($= \varepsilon_{\mathrm{max},n} = n\Delta\varepsilon$) was increased stepwise with an increment ($\Delta\varepsilon$) from $n = 1$ to n^{th} cycle. $\Delta\varepsilon = 2.0$, $n = 6$ for SN-2.0, $\Delta\varepsilon = 0.05$, $n = 5$ for brittle DN-0.8, $\Delta\varepsilon = 0.25$, $n = 12$ for necking DN-2.0, and $\Delta\varepsilon = 0.25$, $n = 10$ for other DN gels. The inset in the graph of brittle DN-0.8 shows magnified plots of the curves.

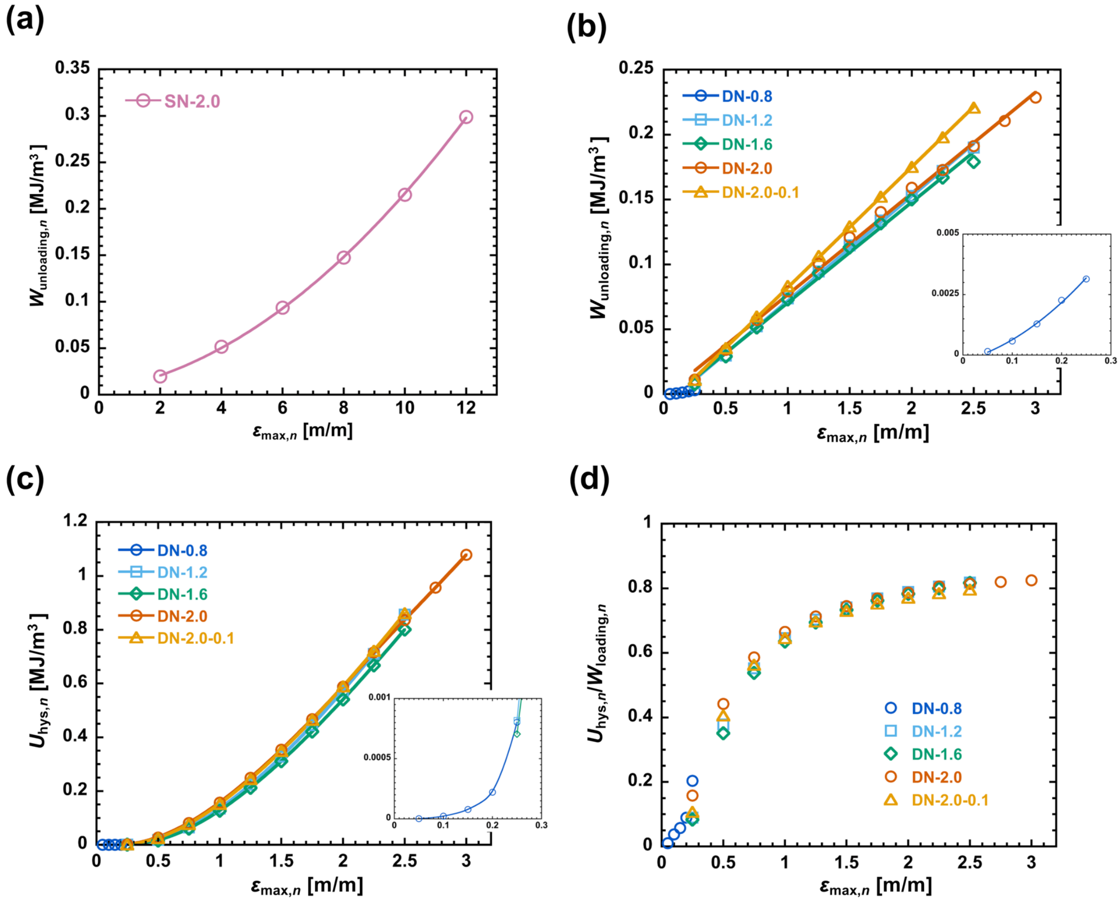

Fig. S4. (a) Dependence of $W_{\mathrm{unloading}}$ on loading strain, $\varepsilon_{\max,n}$, of SN-2.0. As the SN gel was loosely cross-linked, it showed a small amount of hysteresis (~5% to W_{loading}) due to rearrangement of polymer chains during loading. (b) Dependence of $W_{\mathrm{unloading}}$ on $\varepsilon_{\max,n}$ of all DN samples. The inset shows magnified plots of brittle DN-0.8. Brittle DN-0.8 that broke at a very small strain (~ 0.25) demonstrated a very weak mechanical hysteresis. In contrast, the $W_{\mathrm{unloading},n}$–$\varepsilon_{\max,n}$ relations of necking and unnecking DN gels could be fitted by linear regression. (c) Dependence of dissipated energy, $U_{\mathrm{hys},n}$, on $\varepsilon_{\max,n}$ of all DN samples. The inset shows magnified plots of brittle DN-0.8. (d) Dependence of ratio of the dissipated energy to the loading work, $U_{\mathrm{hys},n}/W_{\mathrm{loading},n}$, on $\varepsilon_{\max,n}$ of all DN samples. Here, $U_{\mathrm{hys},n}$ and the loading work $W_{\mathrm{loading},n} = W_{\mathrm{unloading},n} + U_{\mathrm{hys},n}$ were calculated from Fig.S3 in the same way as our previous work [1].

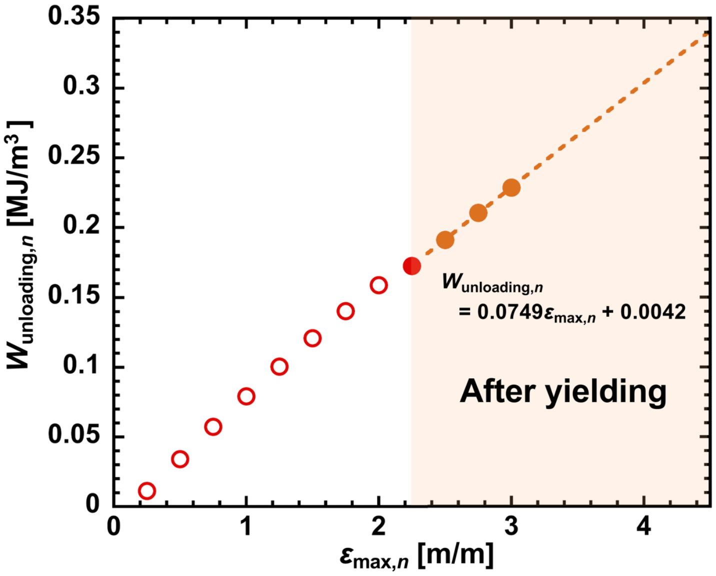

Fig. S5. Extrapolation of the $W_{\mathrm{unloading},n}$–$\varepsilon_{\max,n}$ relation of DN-2.0 for the calculation of the G of samples with smaller l_0 under large preset strain ε (> 3) in Fig. 5. Our previous work demonstrated that, for a typical DN hydrogel, $U_{\mathrm{hys},n}/W_{\mathrm{loading},n}$ keeps constant after yielding point during cyclic tensile test [1]. If we assumed that $W_{\mathrm{loading},n}$ linearly increases with $\varepsilon_{\max,n}$ after yielding, where the stress–strain curve shows plateau region, $W_{\mathrm{unloading},n} = W_{\mathrm{loading},n} - U_{\mathrm{hys}} = W_{\mathrm{loading},n}(1 - U_{\mathrm{hys},n}/W_{\mathrm{loading},n})$ also should linearly increase with $\varepsilon_{\max,n}$. Therefore, $W_{\mathrm{unloading}}(\varepsilon)$ at large ε could be obtained by extrapolating the $W_{\mathrm{unloading},n}$–$\varepsilon_{\max,n}$ relation in the after-yielding region ($\varepsilon_y \sim 2.25$ as shown in Fig. S3).

Reference

[1] T. Nakajima, T. Kurokawa, S. Ahmed, W. Wu, J. P. Gong, Characterization of Internal Fracture Process of Double Network Hydrogels under Uniaxial Elongation, Soft Matter 9 (2013) 1955–1966. https://doi.org/10.1039/C2SM27232F.