Supplementary Information

Characterization of Internal Fracture Process of Double Network Hydrogels under Uniaxial Elongation

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Confirmation of overlapping of loading and unloading curves

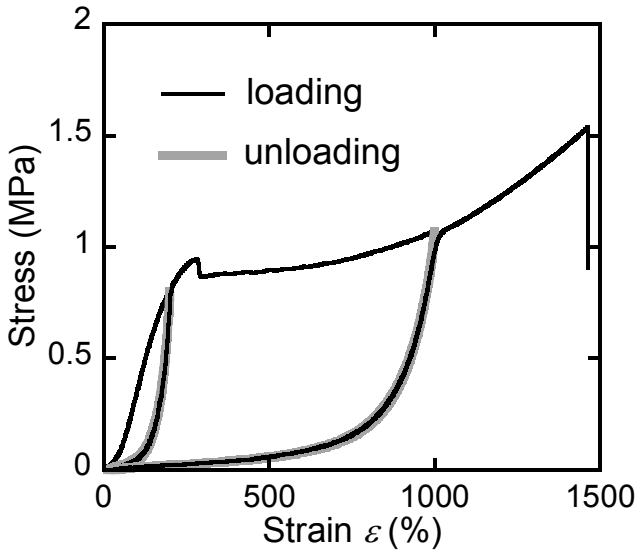

Figure S1 Tensile loading-unloading curves of the DN gel. Strain was calculated from the cross-head displacement, different from the other experiments in main text. The loading and unloading curves were overlapped each other even in large strain. Tensile velocity was fixed at 100 mm/min.

Estimation of chain orientation in PAMPS network

To estimate the chain orientation quantitatively, the chain orientation factor, F_c, of the PAMPS was calculated. Figure S2(a) illustrates the network model that we assumed. Focusing on one polymer chain, which has two end-points, A and B, the F_c of this chain can be expressed as $\cos^2\theta$, where the θ is the angle between the tensile direction and the straight line through A and B. The chain is directed along the tensile direction if $\cos^2\theta$ is equal to 1 and perpendicularly to the tensile direction if $\cos^2\theta$ is equal to 0. Under the assumption of an affine deformation and 0.5 of a Poisson's ratio (constant volume), the length of the segment OA (a) and OB (b) become $a_0\lambda_x$ and $b_0\lambda_x^{-0.5}$ by deformation, where a_0 and b_0 are the values of a and b before deformation, respectively, and λ_x is the deformation ratio defined as ε+1. Thus, $\cos^2\theta$ after stretching with a strain ε can be calculated as;

$$F_c = \cos^2\theta = \left(\frac{a}{\sqrt{a^2+b^2}}\right)^2 = \frac{a_0^2\lambda_x^2}{a_0^2\lambda_x^2+b_0^2\lambda_x^{-1}}$$

The dependence of F_c on the strain, ε, was calculated for three chains with different initial chain orientations. The first chain initially orients almost in perpendicular to the tensile direction and has an initial F_c of 0.2. The second chain orients at 45° and has an initial F_c of 0.5. The third chain orients almost in parallel to the tensile direction and has an initial F_c of 0.8. Figure S2(b) shows the results. In the pre-necking region ($\varepsilon = 2$), although the value of F_c significantly increases with ε for all the three initial orientation, larger initial F_c still leads to larger F_c after deformation. This implies that the PAMPS chains which originally orient in the tensile direction breaks preferentially in this region. In the hardening region ($\varepsilon = 10$), however, all of the F_c values increase significantly and show similar values of approximately 1. In other words, virtually all chains are oriented along the tensile direction, regardless their initial orientation. This implies that the fracture of the PAMPS chains occurs evenly for all the chains regardless of their original orientations in this region.

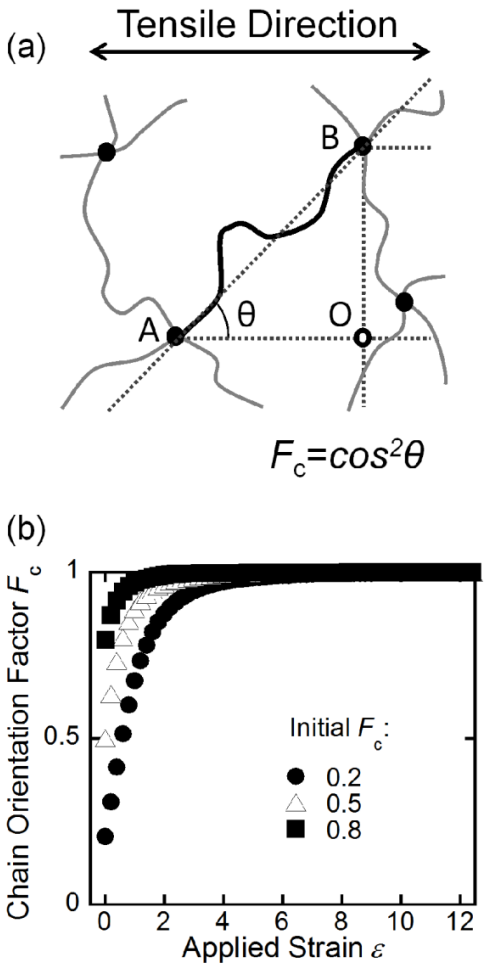

Figure S2 (a) A network model for calculating the chain orientation factor F_c. The factor F_c of the chain AB (heavy line) is defined by $F_c = \cos^2\theta$. (b) Dependence of F_c of three different chains with different initial F_c on the strain ε