# Brownian simulations for tetra-gel-type phantom networks composed of prepolymers with bidisperse arm length

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## Supporting Information

Figure S1 shows the distribution of bead number per strand $P(n)$ for the Rnd networks, which were formed from single long chains via random crosslinking. As expected, the distribution obeys the form $P(n) = (1/n_0)exp(-n/n_0)$, where $n_0$ is the number averaged bead number and $n_0 = 40$ in this case. The polydispersity index is 2.

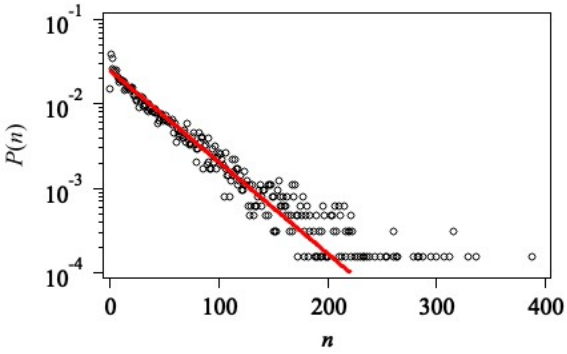


**Figure S1** Distribution of bead number per strand $P(n)$ for Rnd. Red solid line shows $P(n) = (1/n_0)exp(-n/n_0)$ with $n_0 = 40$.

Figure S2 shows the time development of prepolymer fractions with the number of reacted arms of 0 to 4. At the beginning of gelation, all the prepolymers are unreacted, and thus, the fraction of polymers with 0 reacted arms, $\varphi_0$, is unity (see black curves). Immediately after the initiation of the reaction, the fraction of polymers with one arm reacted, $\varphi_1$, grows rapidly (see blue curves). $\varphi_1$ decays after a peak because the reaction occurs for the rest of the arms, and the polymer fractions

with 2 or 3 arms are reacted, $\varphi_2$ and $\varphi_3$, grow in turn (see red and green curves). Finally, $\varphi_4$ (brown curves) saturates around unity, reflecting that almost all the arms are reacted.

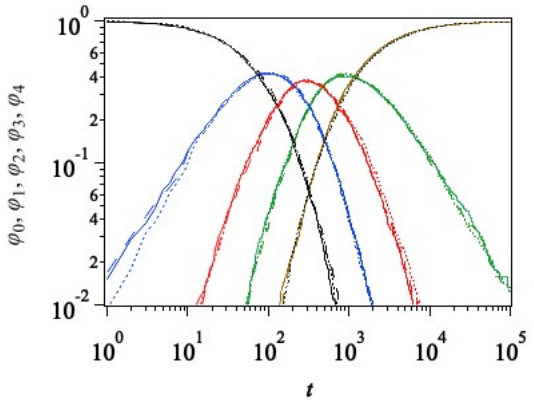


**Figure S2** Time development of prepolymer fractions having number of reacted arms of 0 (black), 1 (blue), 2 (red), 3 (green), and 4 (brown), for 2a2-3802 (broken curve), s2020 (solid curve), and 1a3-0525 (dotted curve).

Figure S3 shows the stress-stretch ($\sigma - \lambda$) relation of a s2020 network with various integration step size, $\Delta t$. We have confirmed that the result is insensitive to $\Delta t$ if we chose the value as $\Delta t \leq 0.002$.

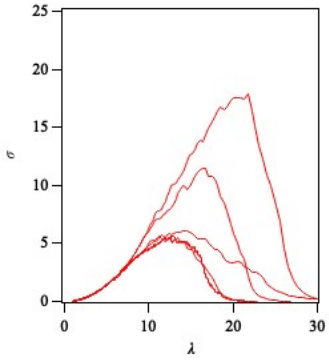


**Figure S3** $\sigma - \lambda$ relation for a s2020 network with various integration step size, $\Delta t$, at 0.02, 0.01, 0.005, 0.002, and 0.001, from top to bottom. The strain rate is $2 \times 10^{-5}$.

Figure S4 shows the $\sigma - \lambda$ relation of a s2020 network with various strain rates. This result clearly demonstrates that the fracture behavior strongly depends on the stretch rate due to the structural relaxation induced by bond scissions. We observed that the strain rate dependence became not large in comparison to the difference among independent simulation runs, if the strain rate is lower than $\dot{\varepsilon} = 2 \times 10^{-5}$.

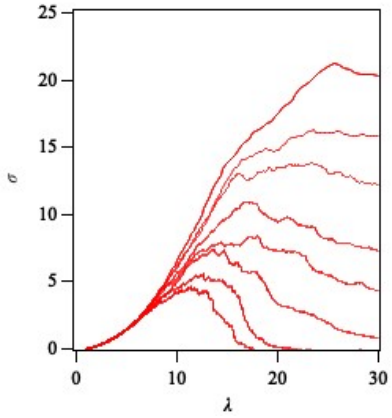


**Figure S4** $\sigma - \lambda$ relation for a s2020 network with the strain rates at $1 \times 10^{-5}$, $2 \times 10^{-5}$, $5 \times 10^{-5}$, $1 \times 10^{-4}$, $2 \times 10^{-4}$, $5 \times 10^{-4}$, $1 \times 10^{-3}$, and $2 \times 10^{-3}$, from bottom to top.

Figure S5 exhibits the $\sigma - \lambda$ curves for s2020 systems with various system sizes. The involved prepolymers are 16,400, 32,800, and 65,600, respectively. The scatter among different systems depends on the system size, and it becomes smaller for larger systems. The stress mitigation after the peak does not significantly depend on the system size, and the size effect is concealed in the difference among simulation runs.

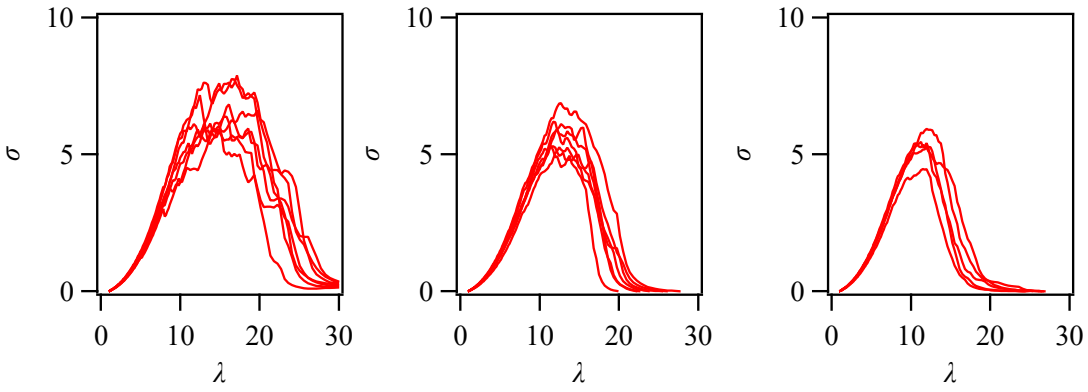


Figure S5 $\sigma - \lambda$ relation for s2020 systems for eight different simulation runs with the number of prepolymers of 16,400, 32,800, and 65,600 from left to right. The strain rate is $2 \times 10^{-5}$.

Figure S6 shows the $\sigma - \lambda$ relations for 1a3 systems. As seen for 2a2 systems in Fig 7 in the main text, the $\sigma - \lambda$ curves for different initial configurations are diverse for the networks made of asymmetric prepolymers. We observe a few bundles of the $\sigma - \lambda$ curves for some cases in which the network breakage is dominated by the scission of network strands with specific lengths.

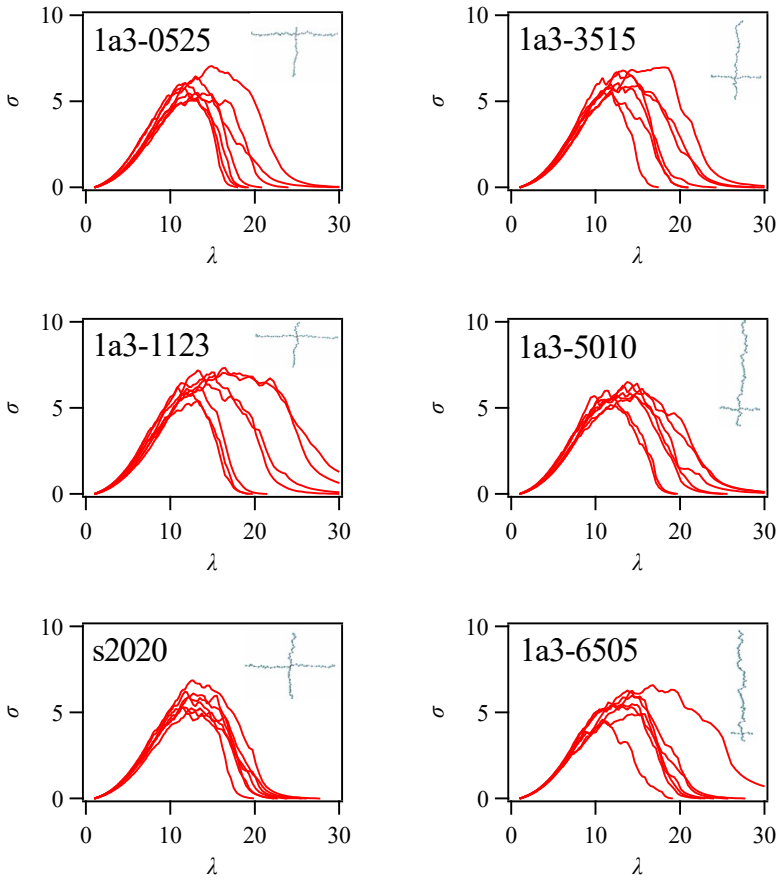


Figure S6 $\sigma - \lambda$ relations 1a3 systems. The results from 8 independent simulation runs are shown.