

**Supporting Information for Kinetics-dominated structure and
stimuli-responsiveness in the assembly of colloidal nanotubes**

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Table S1: Thermodynamic interaction parameters correlated to total molecular interaction that shows the viscosity change (K_d), adhesion of DA to IG (K_{0d}), and hydrocluster formation (u_d) estimated from the change of damping ratio vs. reaction time.

Species of DA	K_d [10^2]	K_{0d}	u_d [10^2]
MA	2.4	0.55	4.4
FA	6.7	1.3	5.3
CTA	2.0	0.54	3.7
MSA	4.5	0.95	4.8

Table S2: Thermodynamic interaction parameters correlated to total molecular interaction that shows the change in elasticity (K_p), interaction between hydroclusters (K_{0p}), and network formation (u_p) estimated from the change in period vs. reaction time.

Species of DA	K_p [10^2]	K_{0p}	u_p [10^3]
MA	1.7	14	1.3
FA	5.0	9.1	5.5
CTA	1.7	8.8	2.0
MSA	2.9	18	1.6

Table S3: Δu of various IG–DA mixtures

Species of DA	MA	FA	CTA	MSA
Δu	3.1	-0.20	1.7	3.2

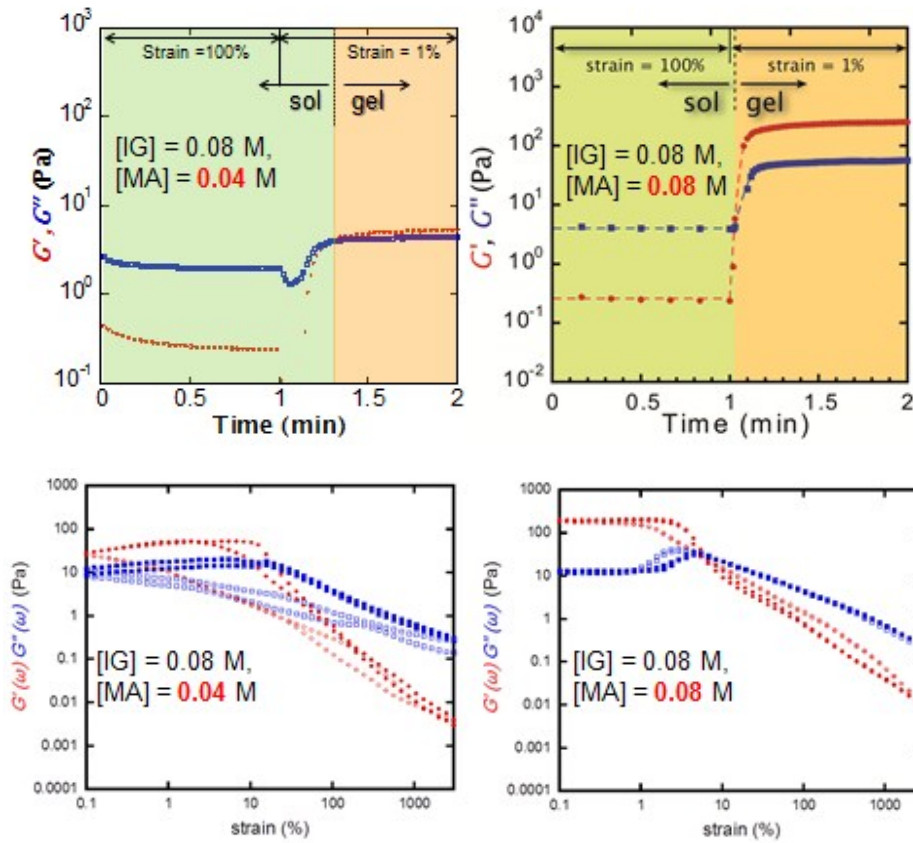


Figure S1. Typical response of storage modulus G' (red circles) and loss modulus G'' (blue squares) of IG–MA mixture at various component concentration (upper) in strain (ε) step change measurements from 100% to 1%, recorded at oscillatory angular frequency of 1 Hz; and (below) in ε sweep measurements by changing ε upward from 0.1% to 3000% (closed symbols) and downward from 3000% to 0.1% (open symbols). $[-\text{Al}(\text{OH})_2]$ of IG and $[\text{MA}]$ are described in each graphs. The solid/liquid (gel/sol) transition was confirmed from the crossover point between the storage modulus (G') and the loss modulus (G''); that is, $G' > G''$ when the sample is in the solid state (gel), and $G' < G''$ when the sample turns to the liquid state (sol). When the strain was maintained at 100% and abruptly decreased to 1%, as shown in upper graphs, the magnitudes of G' and G'' of the IG–MA mixture were again inverted within several s after the strain change (This is defined as solid/liquid (gel/sol) transition time (T_{trans})) due to the liquid/solid (sol/gel) transition. Because the ε sweep measurements of below graphs exhibited a crossover point at about $\varepsilon = \text{several}\%$, the solid/liquid transition of the IG–MA mixture was induced rapidly at the initial stage of strain increase.

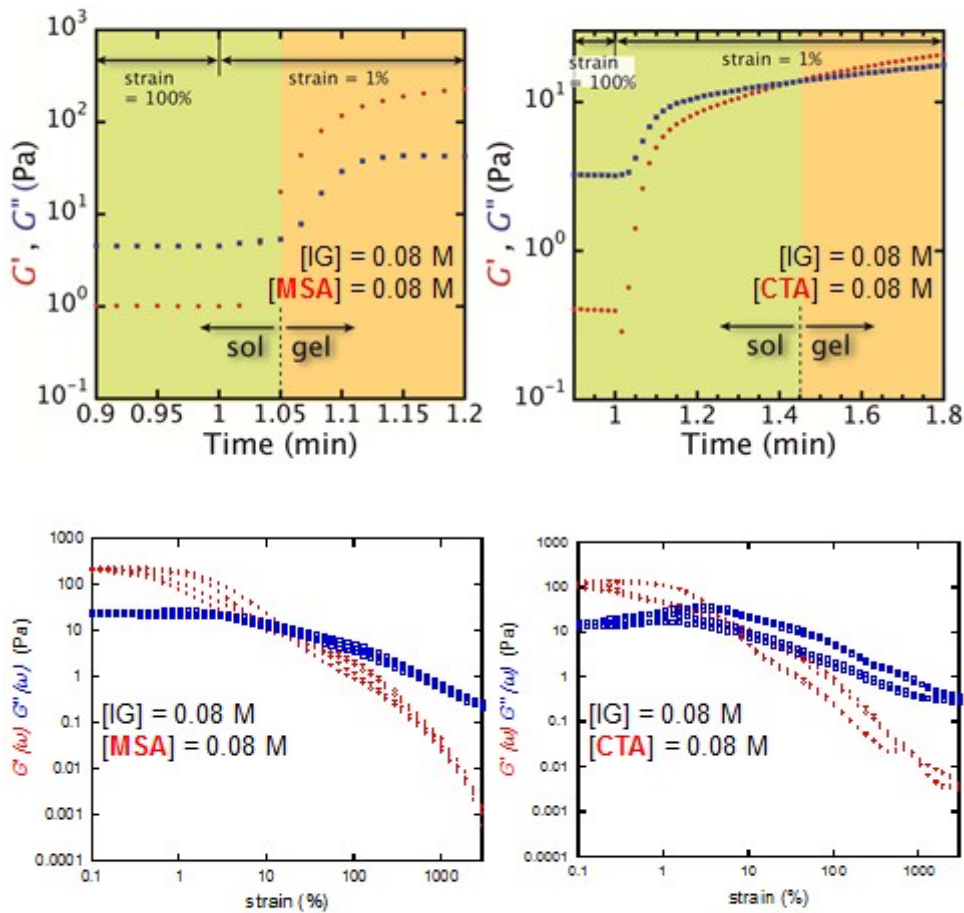


Figure S2. Typical response of storage modulus G' (red circles) and loss modulus G'' (blue squares) of IG–DA mixture (upper) in strain (ϵ) step change measurements from 100% to 1%, recorded at oscillatory angular frequency of 1 Hz; and (below) in ϵ sweep measurements by changing ϵ upward from 0.1% to 3000% (closed symbols) and downward from 3000% to 0.1% (open symbols). $[-\text{Al}(\text{OH})_2 \text{ of IG}] = [\text{DA}] = 0.08 \text{ mol L}^{-1}$. The solid/liquid (gel/sol) transition was confirmed from the crossover point between the storage modulus (G') and the loss modulus (G''); that is, $G' > G''$ when the sample is in the solid state (gel), and $G' < G''$ when the sample turns to the liquid state (sol). When the strain was maintained at 100% and abruptly decreased to 1%, as shown in upper graphs, the magnitudes of G' and G'' of the IG–DA mixture were again inverted within several s after the strain change (This is defined as solid/liquid (gel/sol) transition time (T_{trans})) due to the liquid/solid (sol/gel) transition. Because the ϵ sweep measurements of below graphs exhibited a crossover point at about $\epsilon = \text{several}\%$, the solid/liquid transition of the IG–DA mixture was induced rapidly at the initial stage of strain increase.

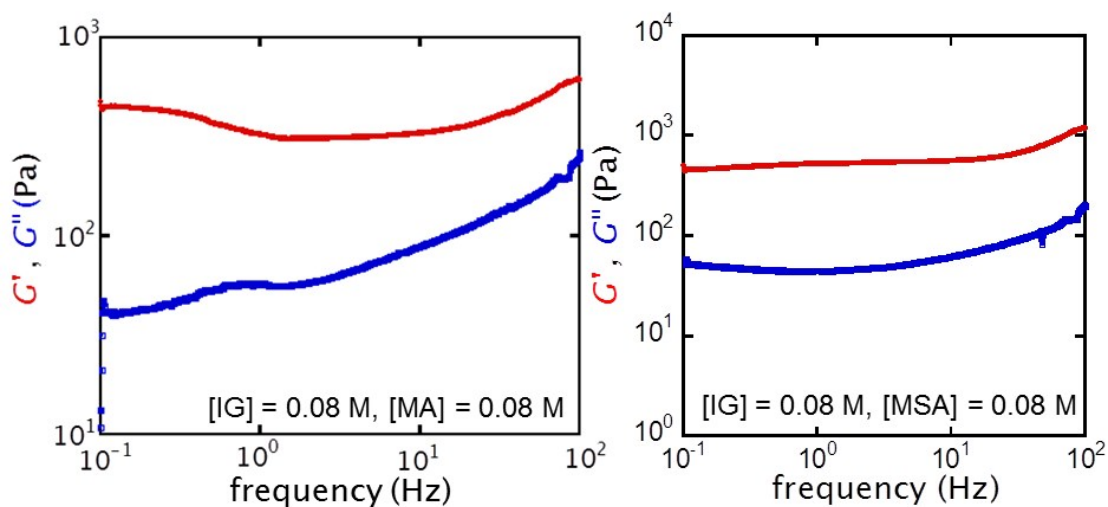


Figure S3. Response of storage modulus G' (red circles) and loss modulus G'' (blue squares) of IG–MA mixture (left) and IG–MSA mixture (right) in strain frequency sweep measurements by changing frequency upward from 0.1% to 10 Hz [$-\text{Al}(\text{OH})_2$ of IG] = [DA] = 0.08 mol L⁻¹. As shown in these responses, the IG–DA mixtures which show thixotropic behavior keep its solid (gel)-state at frequency = 0.1 ~ 100 Hz. These bulk mechanical responses of the IG–DA mixtures were measured with a stress-controlled rheometer (AR-G2, TA Instruments, New Castle, DE). Parallel plates (titanium), 60 mm in diameter, were used for the measurement. The plates were placed in a gap of 400 μm , and the temperature was maintained at r.t.

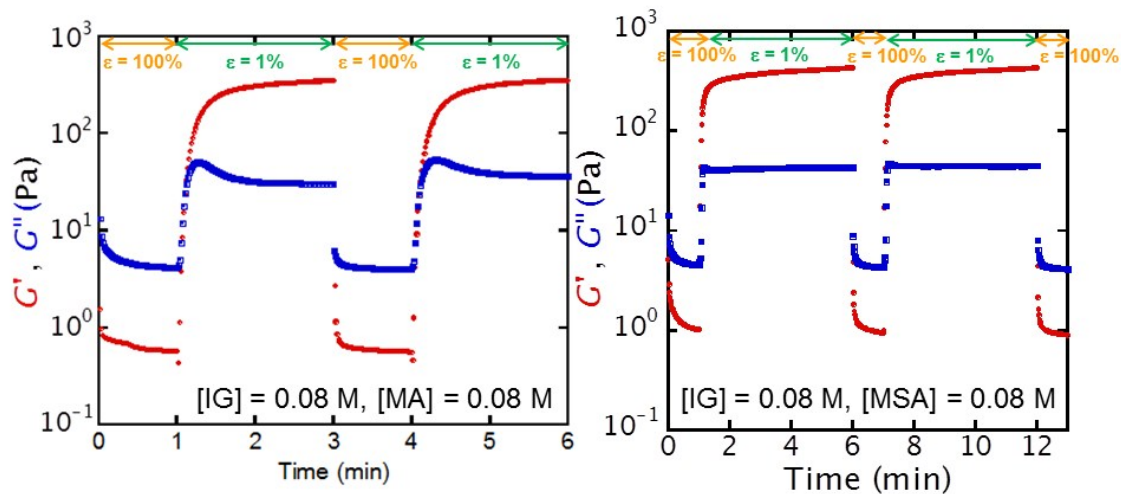


Figure S4. Response of storage modulus G' (red circles) and loss modulus G'' (blue squares) of IG–MA mixture (left) and IG–MSA mixture (right) in ϵ step change measurements from 100% to 1%, recorded at oscillatory angular frequency of 1 Hz. $[-\text{Al}(\text{OH})_2 \text{ of IG}] = [\text{DA}] = 0.08 \text{ mol L}^{-1}$. When the strain was maintained at 100% and abruptly decreased to 1%, as shown in a, the magnitudes of G' and G'' of the IG–MA mixtures were again inverted within several hundred sec after the strain change due to the liquid/solid (sol/gel) transition. Because the ϵ sweep measurements of Figure S1 and S2 exhibited a crossover point at about $\epsilon = \text{several}\%$, the solid/liquid transition of the IG–MA mixture and IG–MSA mixture were induced rapidly at the initial stage of strain increase. As shown in these responses, the fast recovery of the IG–DA mixtures which show thixotropic behavior is not disturbed after shear in the gel continuously for some time and then left at rest to recover.

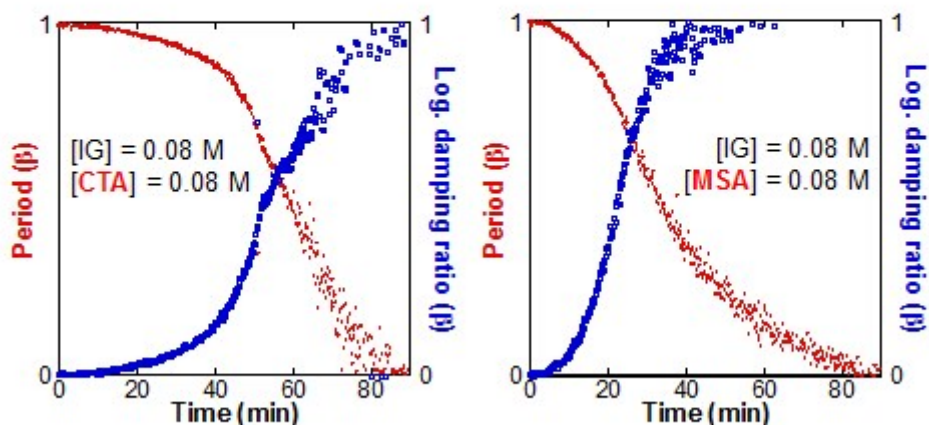


Figure S5. Typical rigid-body free damping oscillation (RFDO) curves of IG–DA mixture at the initial gelation stage. $[-\text{Al}(\text{OH})_2 \text{ of IG}] = [\text{DA}] = 0.08 \text{ mol L}^{-1}$. From the RFDO curves, we can obtain the binding constant (K) as well as the other thermodynamic parameters of the adhesion of DA to IG, hydrocluster formation, interaction between hydroclusters, and network formation by the following equation. $K = K_0 u = 1/(T_s)_{0.5}$, where K_0 is the binding constant of the IG nanotube to DA or of hydrocluster to another isolated hydrocluster (initiation process), $(T_s)_{0.5}$ is the reaction time at $\beta = 0.5$ (β is defined as a ratio of average oscillation period or logarithmic damping ratio to their maximum values), and u is the cooperative parameter which tells the extra interaction energy between the IG nanotubes sheathed DA or between the hydroclusters (propagation process). The value of u can be calculated from the slope of the RFDO curves at the half-value point. $(d\beta/d \ln T_s)_{0.5} = u^{0.5}/4$.