Supplementary Information

In-situ Observation of Hierarchical Self-Assembly Driven by Bicontinuous Gelation in Mixed Nanodisc Dispersions

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Supplementary Movies:

Movie S1: This movie shows the z-stack of bicontinuous gel for $r = 5$, shows the porous structure with well-defined network made of two nanoclays.

Characterization of nanoplatelets

Chemical structure of Laponite $Na^{+0.7}[(Si_8\,Mg_{5.5}\,Li_{0.3})\,O_{20}\,(OH)_4]^{-0.7}$

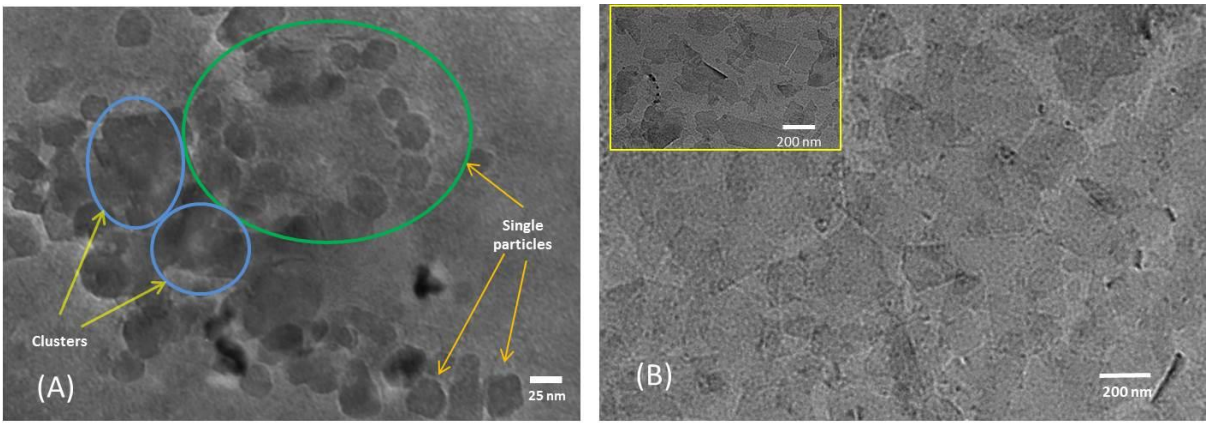


Figure S1 TEM images of Laponite (A) and Na-Montmorillonite (B). The individual particles and clusters are indicated as shown in figure. Laponite clay is monodisperse compared to Montmorillonite.

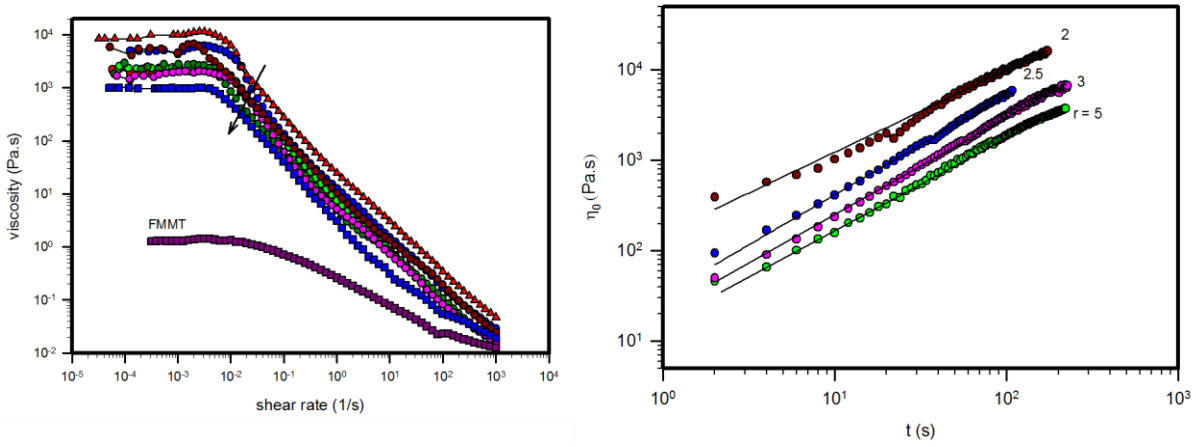


Figure S2 Flow curves and the zero-shear viscosity of the bicontinuous gels for different values of *r*.

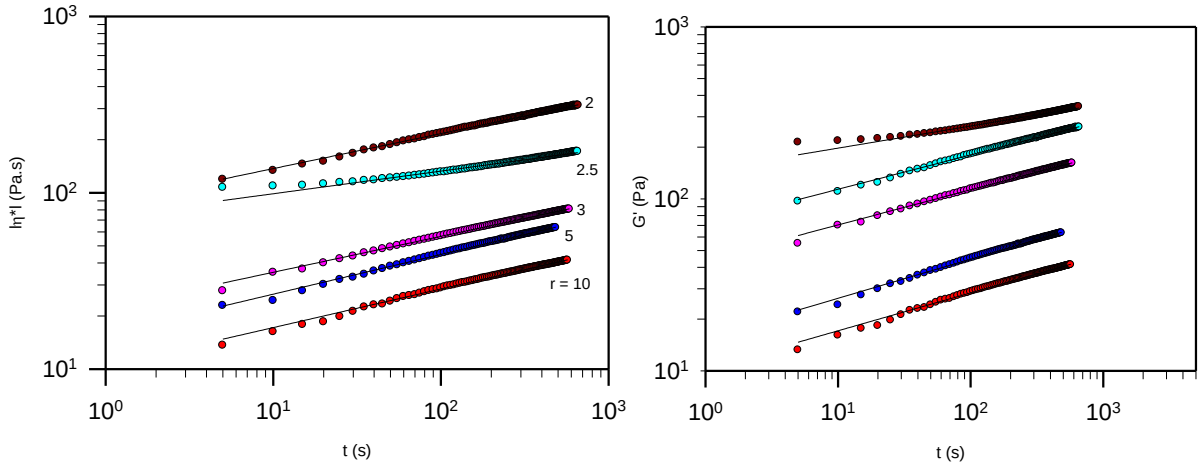
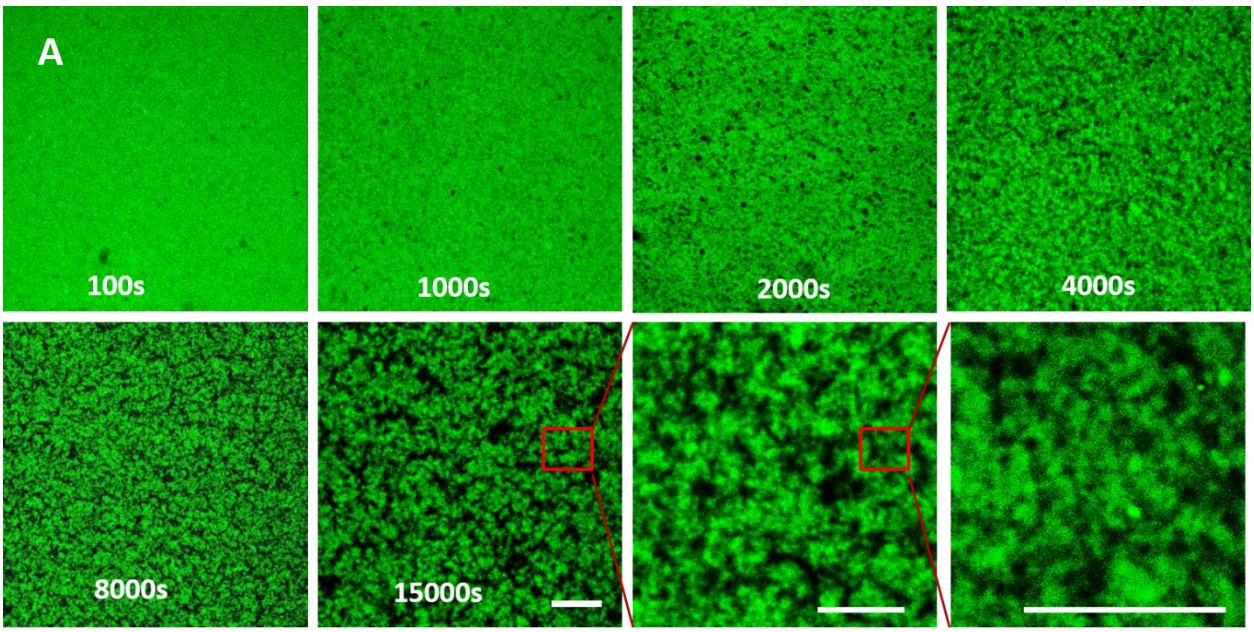


Figure S3. Growth of complex viscosity and elastic modulus with the waiting time at different values of *r*.



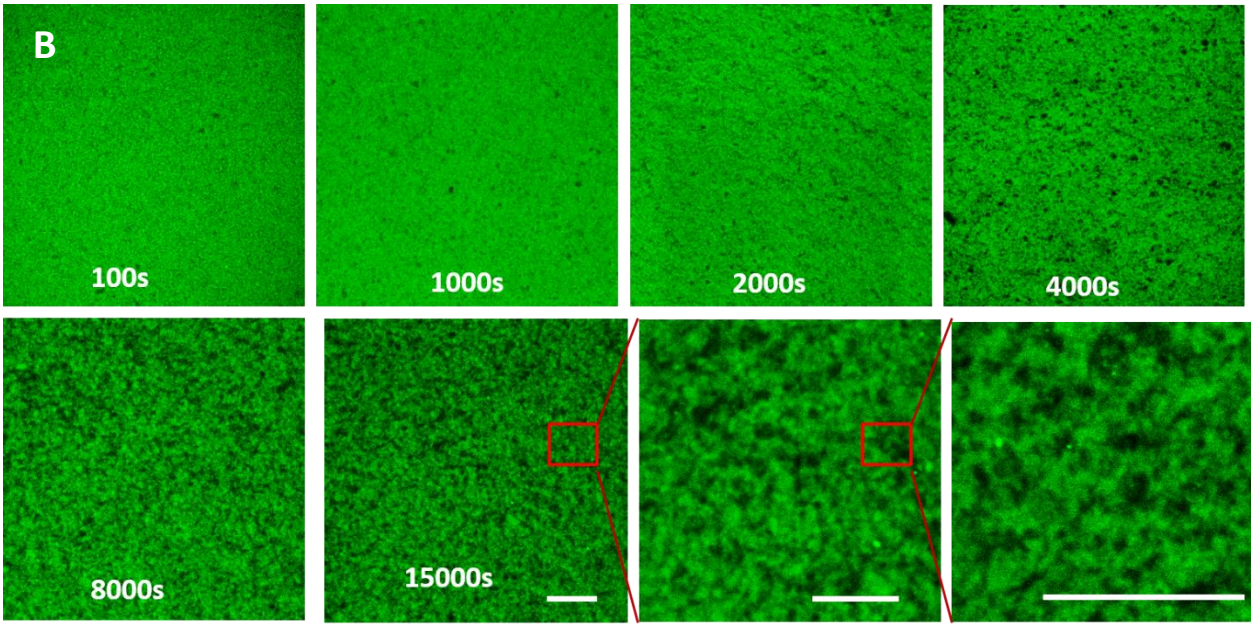


Figure S4 Evolution of gel structure with aging for *r =3* (A) and 2.5 (B). Scare bar is 20 μm.

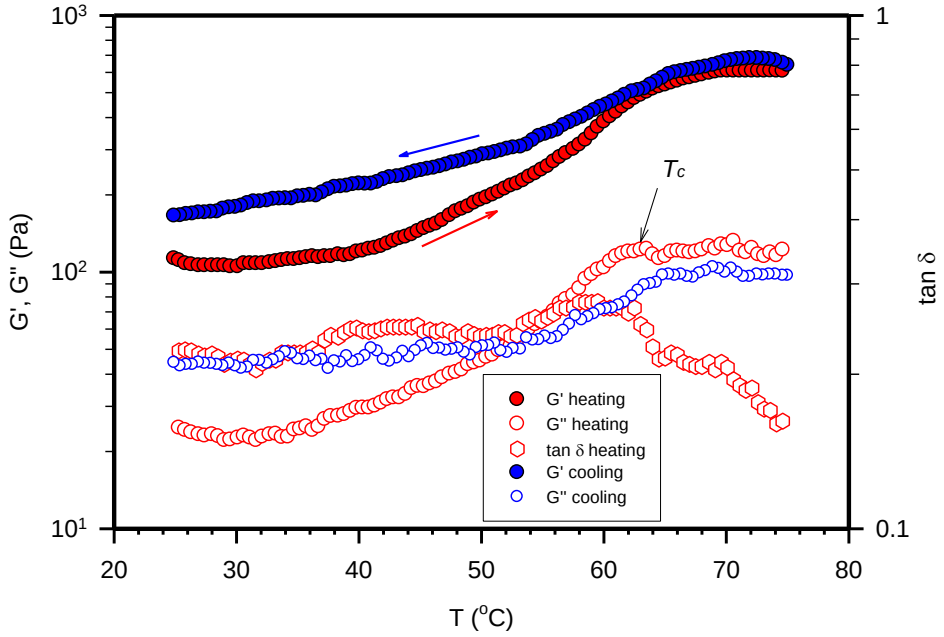


Figure S5. Temperature dependent rheological properties of the bicontinuous gel.

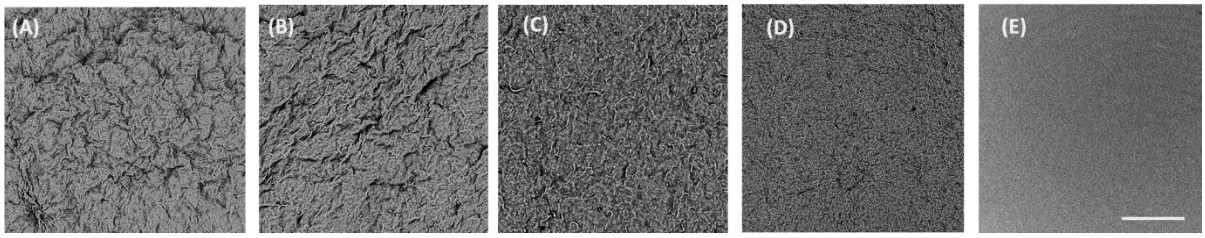


Figure S6 SEM images of dried films of FMMT, bicontinuous gels with ratios r = 8, 5, 3 and Laponite.

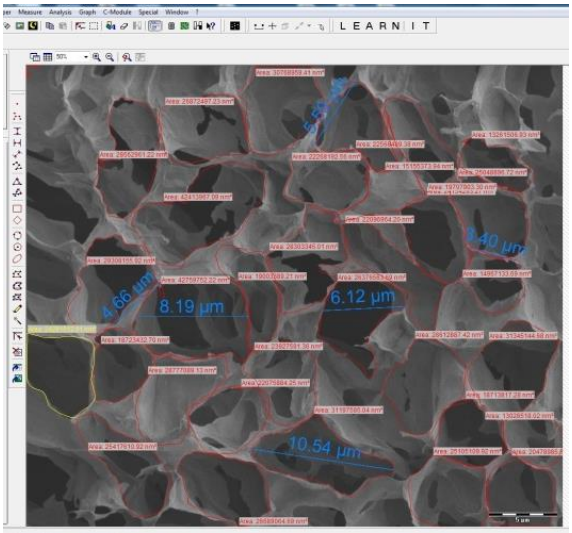


Figure S7 SEM analysis pore size and thickness of the branched of the BCG.

Clay	ρ (g/cc)	Aspect ratio (L/D)	CEC (meq/g)	D_{eff} (nm)	ζ (mV)	Physical state Concentration (c) dependent
Laponite	2.53	25	1.2	25	-40±2	$c < 1$ Phase Separation $1 < c < 2$ Equilibrium Gels (Time Dependent) $2 < c < 3$ Glass (Time Dependent) $c > 3$ Repulsive/Attractive glass Nematic
Na-MMT	2.86	250	0.75	250	-32±2	$c < 2.5$ Phase Separation $2.5 < c < 5$ Soft gels, Nematic $c > 5$ Strong gels, Nematic
L+MMT					Linear combination	$c < 0.3$ Stable sol $0.3 < c < 2.2$ Gel $c > 2.2$ Glass, ordered glass

Table S1: Physio-chemical Properties of clays of Laponite and Na-MMT.

Time\sample	**MMT (5%)**	**Lap (3%)**	**1:0.15**	**1:0.30**	**1:0.60**	**1:1**	**1:1.5**
Day 1	43	40	53	55	57	63	57
Day 5	47	42	56	56	58	65	59
Day 10	49	45	58	58	61	67	60

Table S2: Transition temperatures of bicontinuous gels made from different mixing ratios.