Electronic Supplementary Material (ESI) for Soft Matter.

Supporting Information

Coupling of gelation and glass transition in a biphasic colloidal mixture

—from gel-to-defective gel-to-glass

Di Jia,[1,2] Javoris V. Hollingsworth,[1] Zhi Zhou,[1,2] He Cheng[3,4,*] and Charles C. Han[1,*]

1. Supplementary Video Files S1 and S2.

Video title: biphasic mixture formation process

Binary gel formation process (sample CS94) and biphasic mixture formation process (sample CS60) are observed by confocal microscopy at 35 °C and their final morphologies exhibit in the main text (Fig. 8).

S1: video of binary gel formation process of sample CS94 with Φ_{total} =0.1 and its final morphology is in the main text (sample named "CS94" in Fig. 8)

S2: video of biphasic mixture formation process of sample CS60 with Φ_{total} =0.1 and its final morphology is in the main text (sample named "CS60" in Fig. 8)

Keywords of the video: binary gel formation process; network; cluster

2. Dynamic strain sweeps in the glassy state

In order to support the conclusion that when Φ_{PSS} is below 50% (Φ_{total} =32% is fixed), the system goes into glassy state, dynamic strain sweeps for sample CS50, CS25 and CS0 were studied.

As shown in Fig. 1s, all the three samples show characteristics of glass: G' is almost independent of frequency and G'' shows a minimum, which indicates the characteristic time for a particle to explore its cage. When Φ_{PSS} increases from CS50 to CS0, the G' values continue increasing. Besieds, the minimum of G'' moves to higher frequencies as Φ_{PSS} increases, which indicates smaller cages and larger real volume fractions. These results reconfirm the conclusion that the real volume fraction increases as Φ_{PSS} increases (Φ_{total} is fixed).

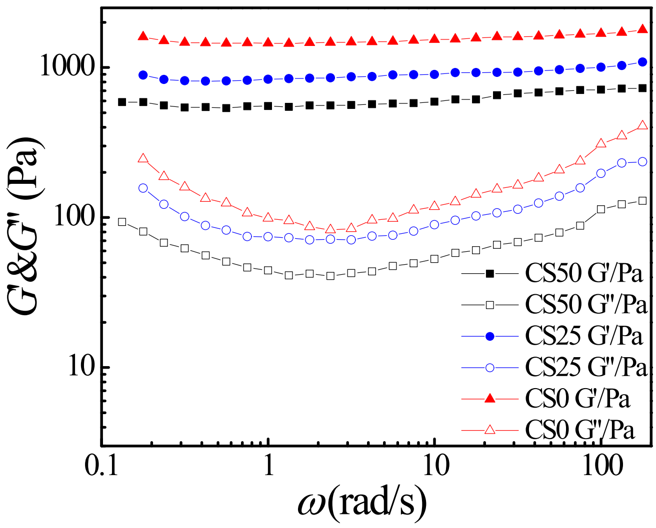

Fig. 1s Dynamic frequency sweeps for sample CS50 (black), CS25 (blue), CS0 (red), respectively at 35 °C, γ=1%. Closed (open) symbols represent storage (loss) moduli.

3. Preshear procedure

First, the sample was presheared at 25°C, the preshear (γ =1%, ω =1 rad/s) was used to make the sample uniform for 180s. Then, the temperature was increased to 35°C and equilibrium for 10 min. Finally, another preshear (γ =1%, ω =1 rad/s) was conducted until the modulus was independent of time.

4. Frequency dependence of yielding behavior

The dynamic strain sweeps at different frequencies for sample CS84 and CS75 are shown in Fig. 2s. For CS84 and CS75, with increasing frequency both *G*′ and *G*″ increase slightly in the linear regime. For CS84, two-step yielding behavior still exists at different frequencies and the yield strains seem to remain unchanged. For CS75, with increasing frequency, both *G*′, *G*″ and the yield strain increase slightly, but there is still one-step yielding at different frequencies. Therefore, there seems to be no transition between two-step yielding and one-step yielding at different frequencies in this system.

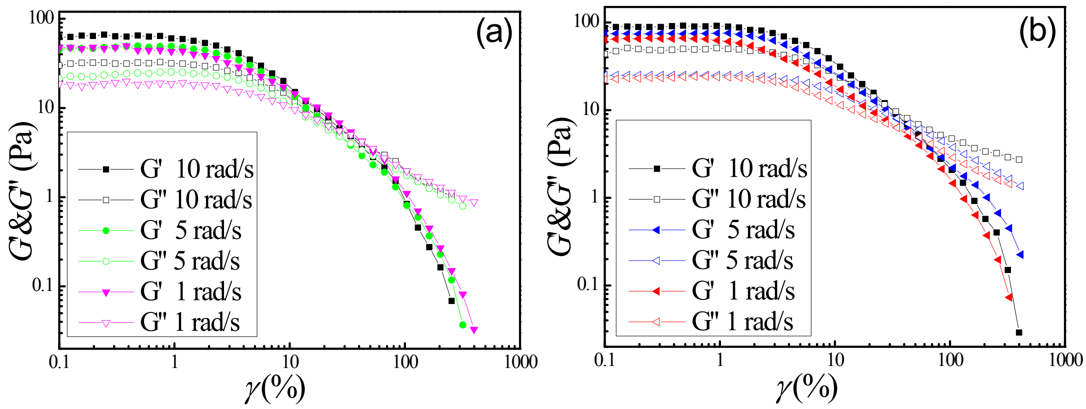

Fig. 2s Dynamic strain sweeps at different frequencies for CS84 (a) and CS75 (b) at 35^{o}C. The closed symbols represent *G*′ and open symbols represent *G*″.