Electronic Supplementary Information (ESI)

Thermo-Reversible, Ergodicity and Surface Charge-Temperature Dependent Phase Diagram of Anionic, Cationic and Neutral Co-Gels of Gelatin-BSA Complexes

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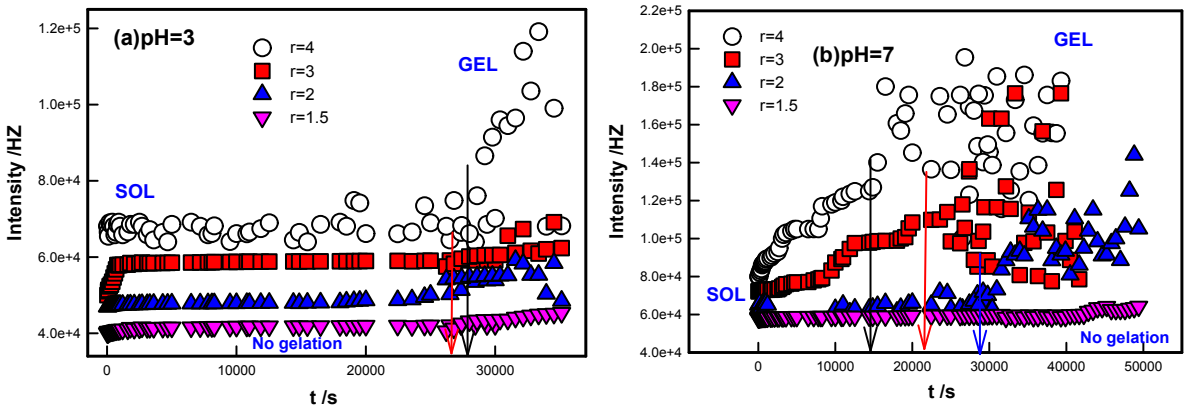


Figure S1: Temporal variation of the light scattered intensity of the gelling solutions shown as function of time during cross linking process of the (BSA-GB) systems for (a) cationic (pH =3) and (b) anionic (pH =7)at mixing ratio (r=1.5, 2, 3, 4) scattering angle of $\theta=90^0$ at 20^0 C. Note the rampant fluctuations in intensity close to onset of gelation. Arrow indicates the sol−gel transition. Note the faster gelation for anionic gel than cationic gels at all mixing ratios.

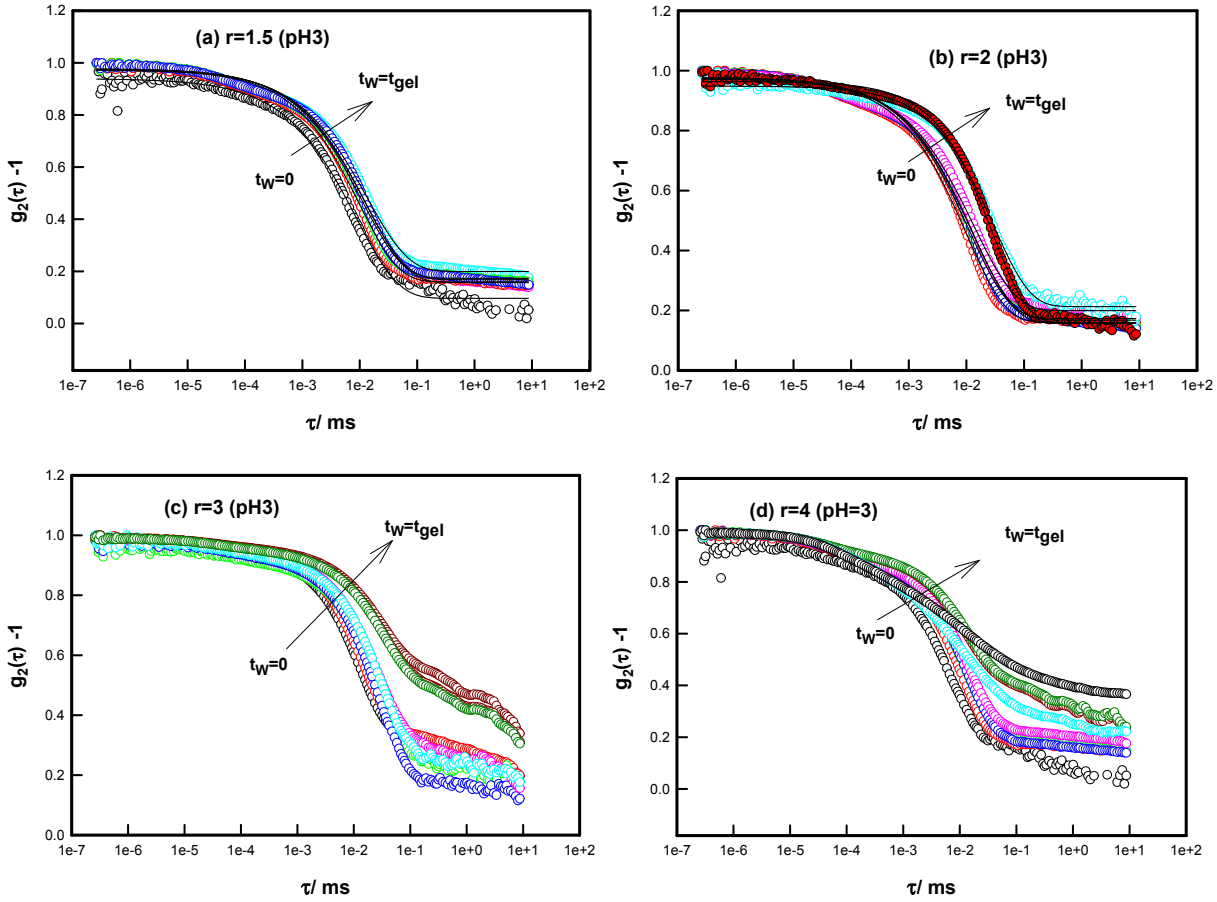
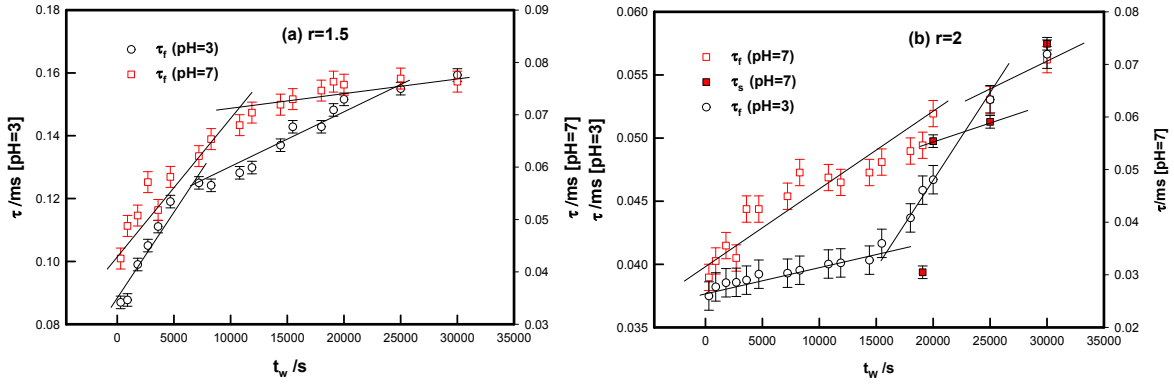


Figure S2: The evolution of dynamic structure factor of thecationic samples at pH=3 with (a) r=1.5, (b)r=2, (c) r=3 and (d) r=4.0. The arrow indicates the evolution of structure factor starting t_w=0h to t_w=t_{gel}h at pH=3.



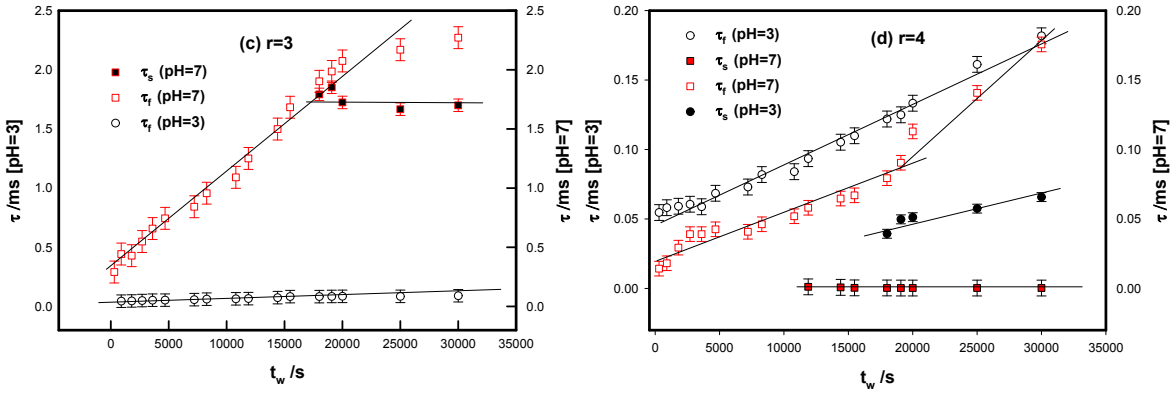
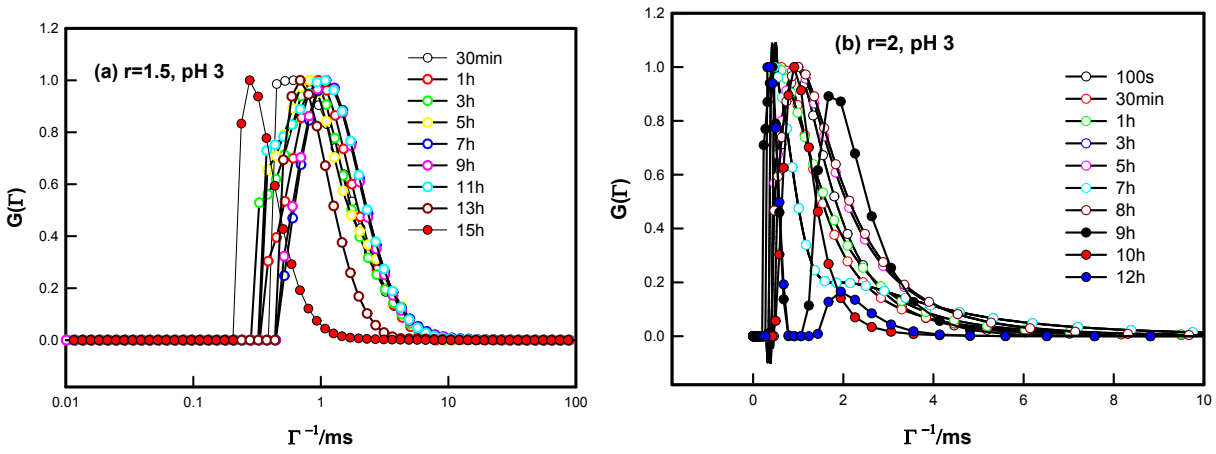


Figure S3: Variation of fast (τ_f) and slow (τ_s) reaction time samples for pH=3 and pH=7as a function of reaction time in equation (2) and (3). Open symbols indicate fast relaxation time while filled symbols indicate slow relaxation time.



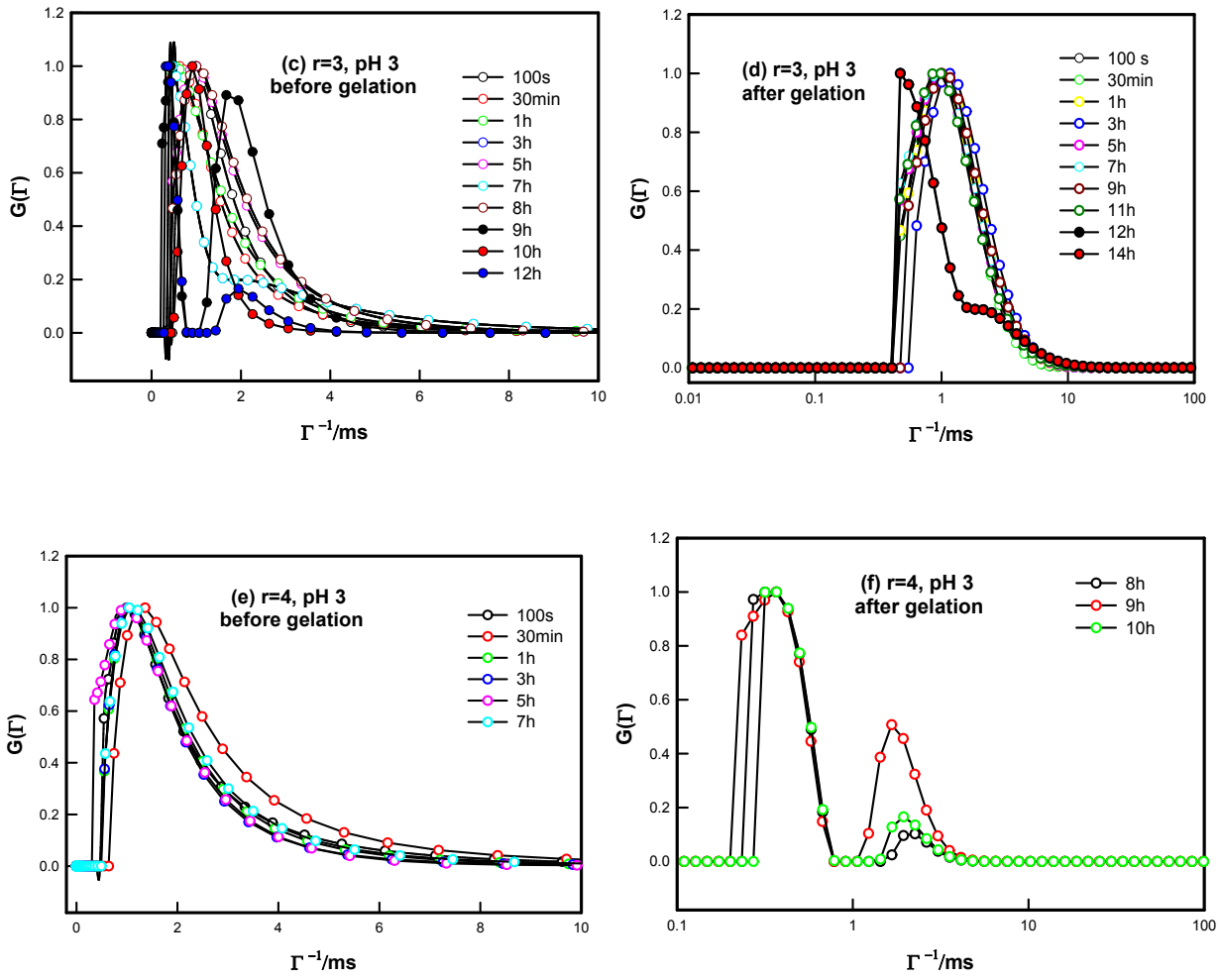


Figure S4: The evolution of decay rate distribution functions obtained for samples with r=1.5, 2.0, 3.0 and 4.0 before and after gelation as a function of time at pH= 3.

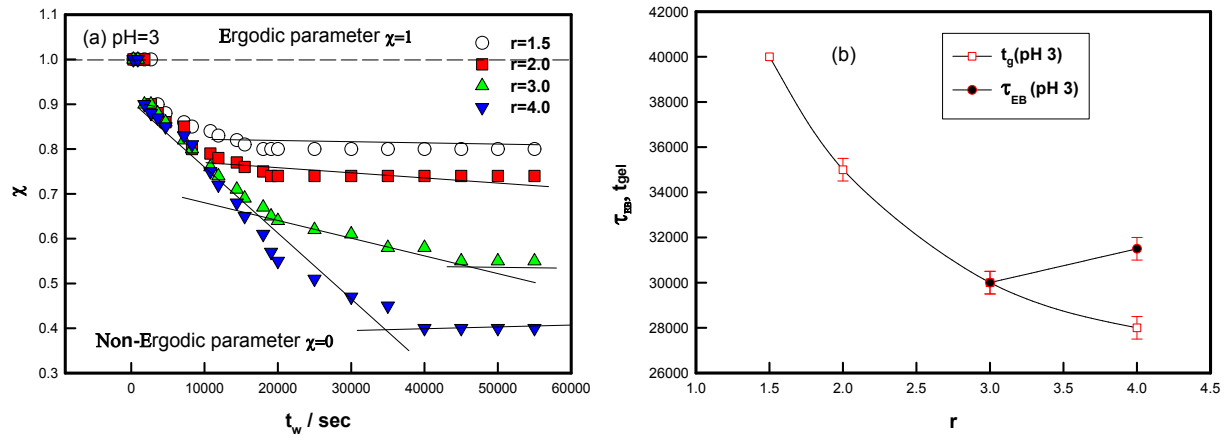


Figure S5: (a) Variation of heterodyne contribution χ at pH 3 shown as a function of waiting time /sec. The point where there is slop changes in the value of χ is defined as aergodicity breaking time τ_{EB}. (b) The ergodicity breaking time (τ_{EB}) and gelation time (t_g) of samples as a function of mixing ratio r at pH 3.

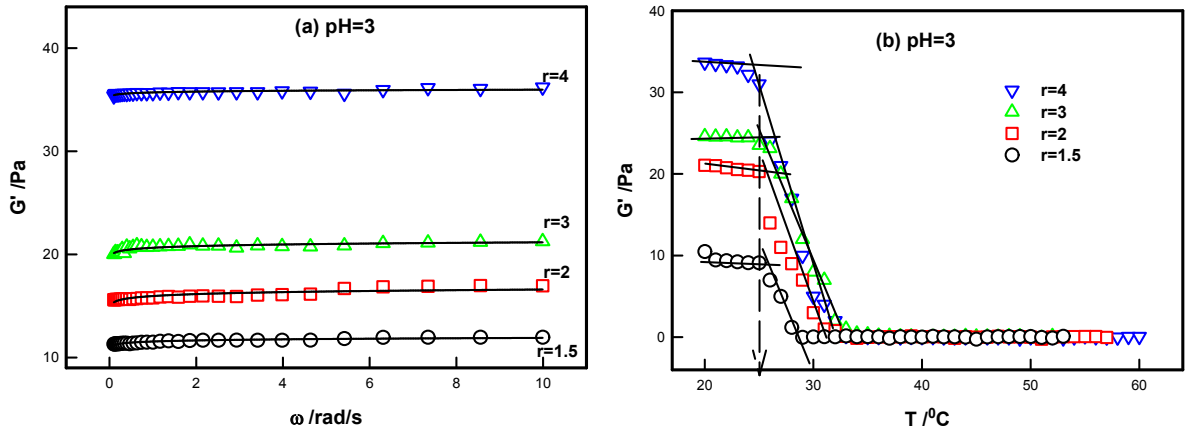
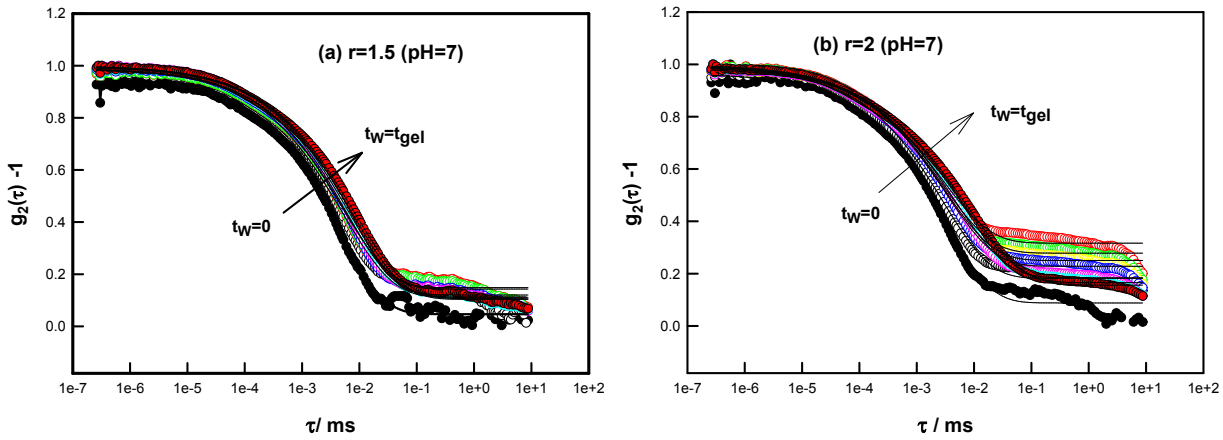


Figure S6: (a) Storage G′(ω) modulus of gel as a function of frequency. (b) Storage modulus G′(ω) of gel as a function of temperature. Sharp drop indicates a melting temperature. The measurements were performed at 25 °C using a constant oscillation stress of 1 Pa. Solid lines is guides to the eye.



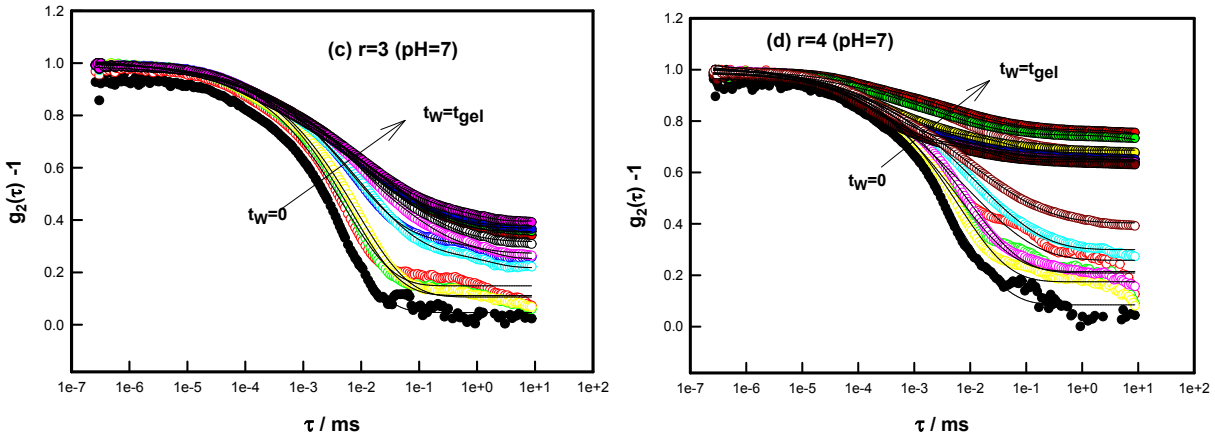


Figure S7: The evolution of dynamic structure factor of the samples with (a) r=1.5, (b) r=2, (c) r=3 and (d) r=4.0 for anionic gels (pH=7). The arrow indicates the evolution of structure factor starting t_w=0 to t_w=t_{gel}h at pH=7.

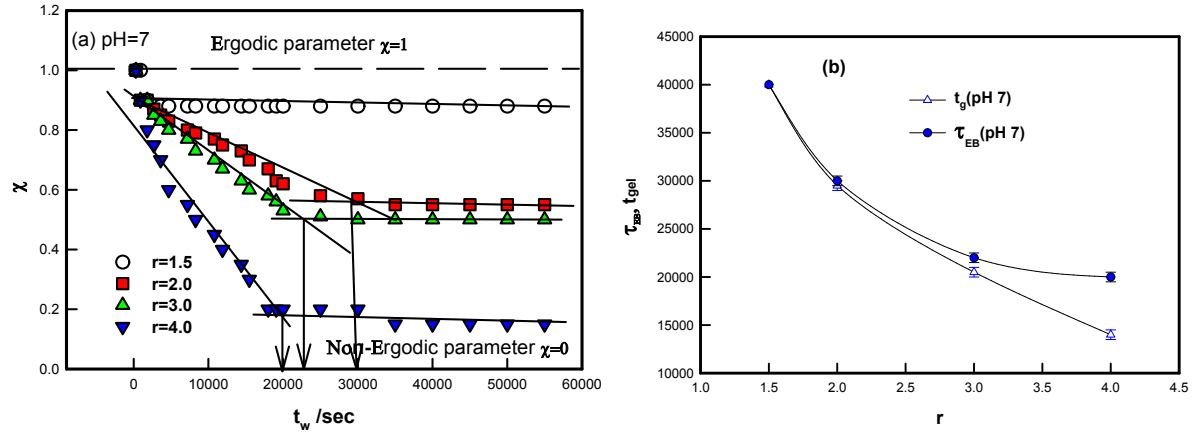
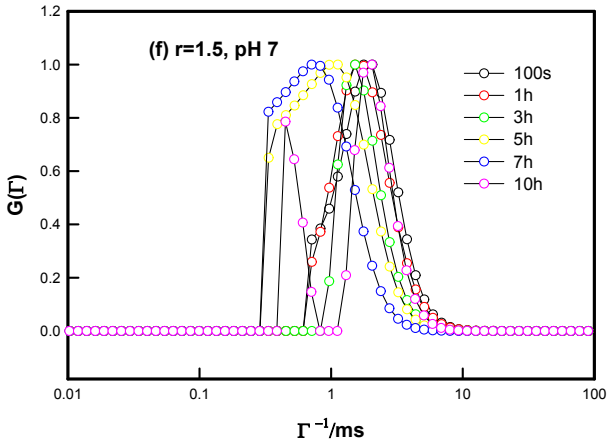
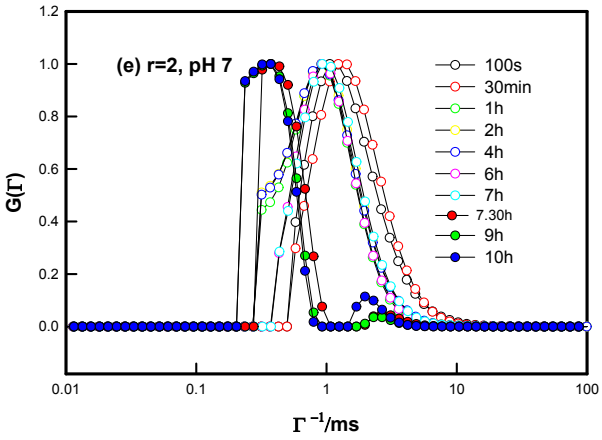
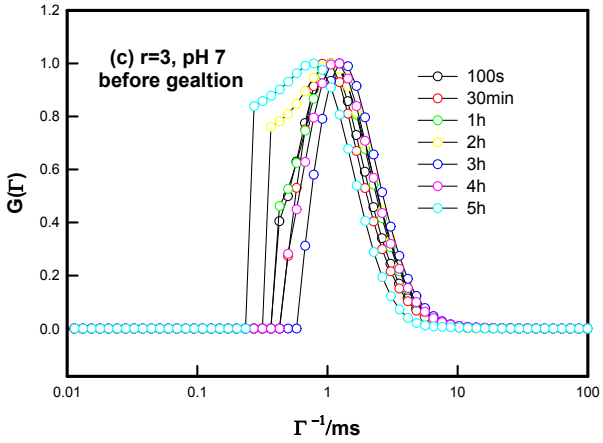
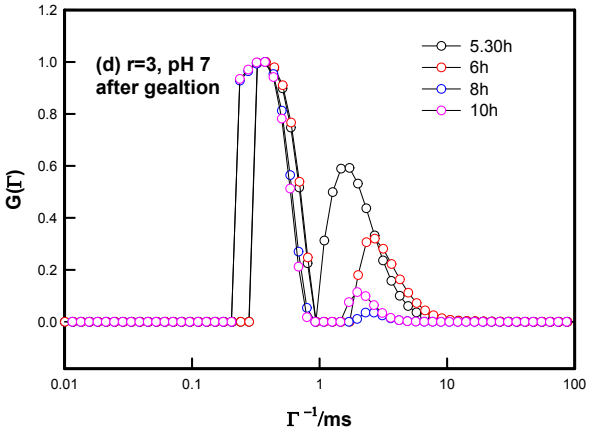


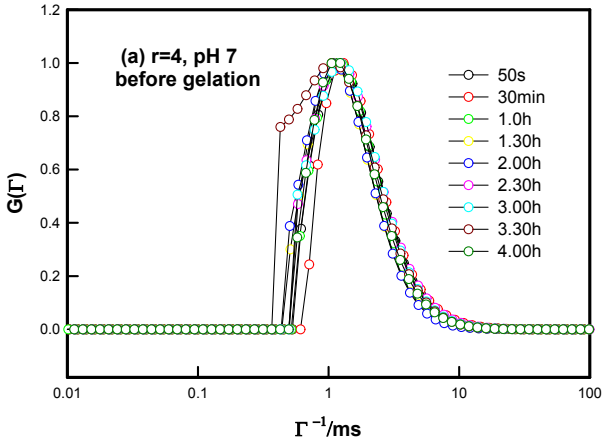
Figure S8: (a) Variation of heterodyne contribution χ at pH 3 shown as a function of waiting time /sec. The point where there is slop changes in the value of χ is defined as aergodicity breaking time τ_{EB}. (b) The ergodicity breaking time (τ_{EB}) and gelation time (t_{gel}) of samples as a function of mixing ratio r at pH 7.


(f) r=1.5, pH 7
100s
1h
3h
5h
7h
10h
G(Γ)
Γ⁻¹/ms


(e) r=2, pH 7
100s
30min
1h
2h
4h
6h
7h
7.30h
9h
10h
G(Γ)
Γ⁻¹/ms


(c) r=3, pH 7
before gealtion
100s
30min
1h
2h
3h
4h
5h
G(Γ)
Γ⁻¹/ms


(d) r=3, pH 7
after gealtion
5.30h
6h
8h
10h
G(Γ)
Γ⁻¹/ms


(a) r=4, pH 7
before gelation
50s
30min
1.0h
1.30h
2.00h
2.30h
3.00h
3.30h
4.00h
G(Γ)
Γ⁻¹/ms

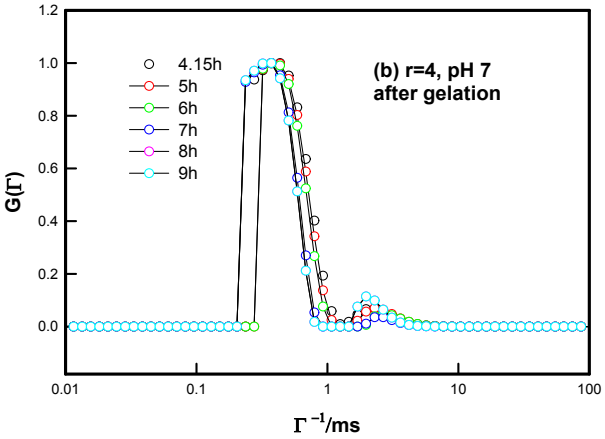

4.15h
5h
6h
7h
8h
9h
(b) r=4, pH 7
after gelation
G(Γ)
Γ⁻¹/ms

Figure S9: The evolution of decay rate distribution functions obtained for samples with r=1.5, 2.0, 3.0 and 4.0 before and after gelation as a function of time at pH= 7.

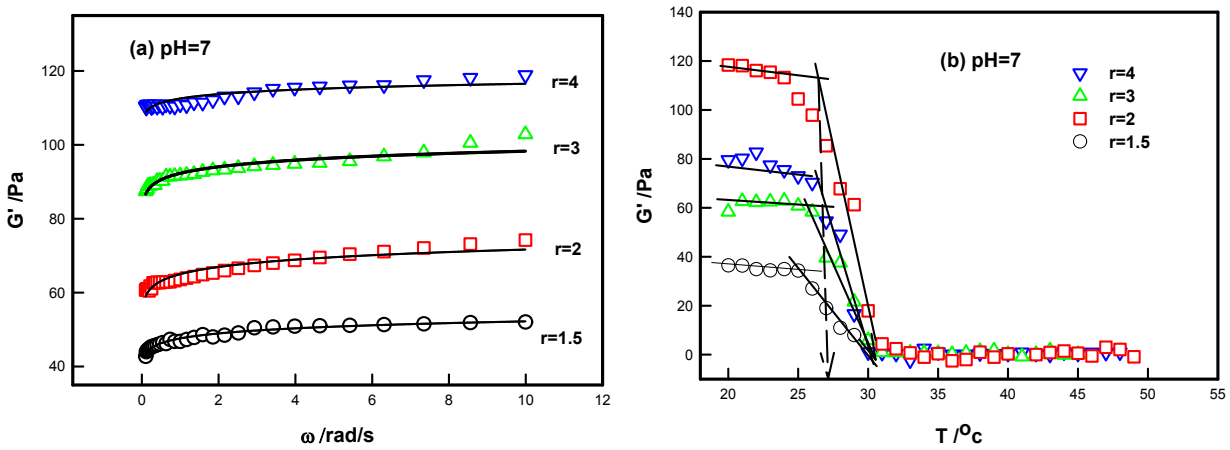


Figure S10: (a) Storage G′(ω) modulus of gel as a function of frequency. (b) Storage modulus G′(ω) of gel as a function of temperature. Sharp drop indicates a melting temperature. The measurements were performed at 25 °C using a constant oscillation stress of 1 Pa. Solid lines is guides to the eye.