Additional Results

- *Long-time evolution of the static structure factor at low Q*

The long time evolution of the static structure factor at low Q gives a further proof of equilibrium gelation for concentrations $C_w > 1.0$ %. To this purpose we report in Fig. S1 the time dependence of S(Q) for a fixed low Q value for both experiments and simulations. Comparing the two regimes for $C_w = 0.8$ % (circles) and $C_w = 1.2$ % (diamonds), respectively inside and outside the phase separation region, it is evident that while the phase separating sample shows a continuous growth with time, the S(Q) for the higher concentration sample approaches a plateau at long times, indicating that the structure stops evolving when the system is close to a fully bonded configuration (equilibrium gel). Simulation data, taken respectively at $\rho^* = 0.08$ and $\rho^* = 0.16$, display the same behavior.
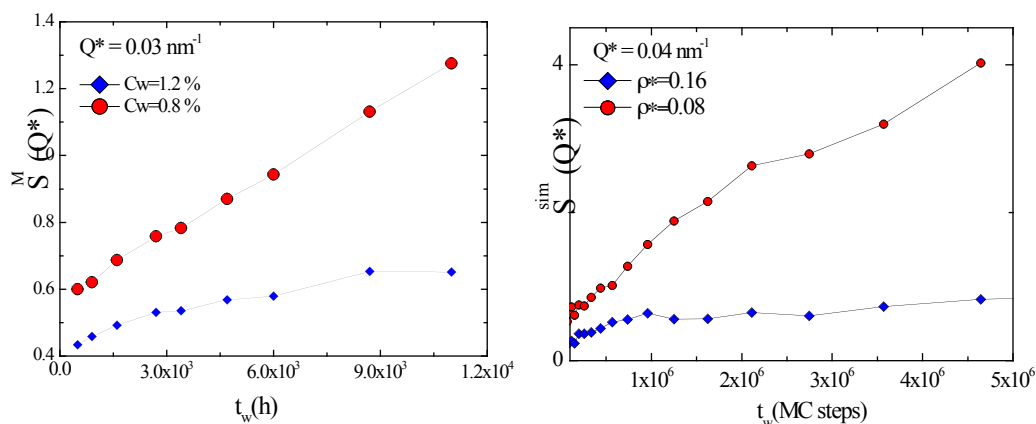

Figure S1: Comparison between S(Q) values for a fixed low Q* as a function of waiting time for phase separating samples (circles) and equilibrium gels (diamonds), respectively from experiments (left panel) and simulations (right panel).

● *Remarks on the additional repulsive electrostatic interactions*

Our model is a "zeroth order model" that neglects the overall repulsive electrostatic interactions. This affects the timescale of the aggregation kinetics but also determines some differences in the shape of S(Q) at short times and in the absolute values of S(Q) at small Q. The focus of our manuscript is to study the behavior of low concentration Laponite samples at large waiting times, when aggregation sets in and where the behavior of experimental and numerical S(Q) are in good agreement. However, we show in this section the comparison between experimental, theoretical and numerical S(Q) at short times obtained considering a repulsive electrostatic interaction. Following our previous work for high Laponite concentrations where the system is — even at long times — repulsive-dominated[1], we have calculated S(Q) for platelets interacting only via screened electrostatic interactions (Yukawa-type) within both integral equation theory[2] and simulations. The simulations are performed using 1000 hard platelets with 19 discrete sites, without attractive patches, but with a Yukawa site-site repulsion[3,4]. In this way, we can accurately reproduce the experimental data for short waiting times, as shown in Fig. S2. The parameters we have used are close to those selected in Ref. 1 for larger clay concentrations.
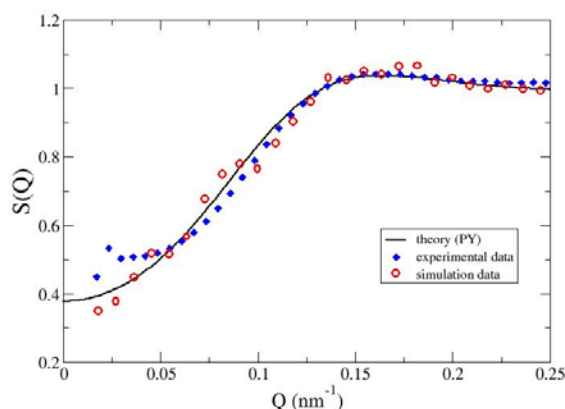

Figure S2: Comparison between experimental data at short times (t_w=500 h) for C_w=1.2 % (full symbols) and a purely repulsive (Yukawa) theoretical (line) and numerical S(Q) (open symbols).

To describe the behavior of Laponite at long waiting times a full model should consider the attractive patches on top of the estimated repulsive interactions. Since the electrostatic barrier estimated from the fits is very large (~ 38 K_BT), the time necessary to form attractive bonds becomes extremely long. Therefore neglecting this barrier within our zero-th order model artificially speeds up the aggregation kinetics. To show that this does not affect the final structure of the system, we have inserted the final configurations from the attractive-only runs reported in the manuscript in a new simulation containing both attractive and repulsive interactions. We have monitored (for a limited time $\sim 10^5$ MC steps) that practically all bonds remain intact and that the final S(Q) changes very slightly, as shown in Fig. S3.
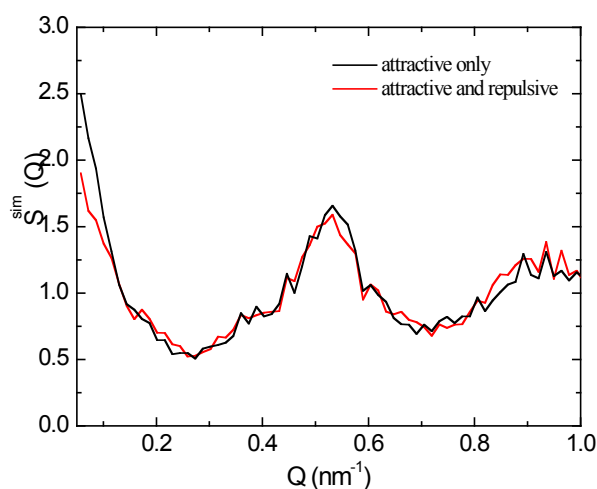

Figure S3: Comparison of numerical S(Q) obtained for ρ^*=0.12 (equilibrium gel) at very long times (t_w=1.1 x 10^8 MC steps) after a quench performed with our zero-th order model (attraction only) and that of the same configuration which has run for additional 10^5 MC steps in the presence of additional electrostatic repulsion.

References

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