Supporting Information

Investigation of the dynamical slowing down process in soft glassy colloidal suspensions: comparisons with supercooled liquids

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The measure of concentration (% w/v) used in the manuscript refers to the weight of Laponite in grams that is mixed in 100 ml of deionized and distilled water. We must note here that % w/v is almost equal to wt %. For example,

2.0% w/v is equivalent to 1.96 wt %

2.5% w/v is equivalent to 2.44 wt %

3.0% w/v is equivalent to 2.91 wt %

3.5% w/v is equivalent to 3.38 wt %

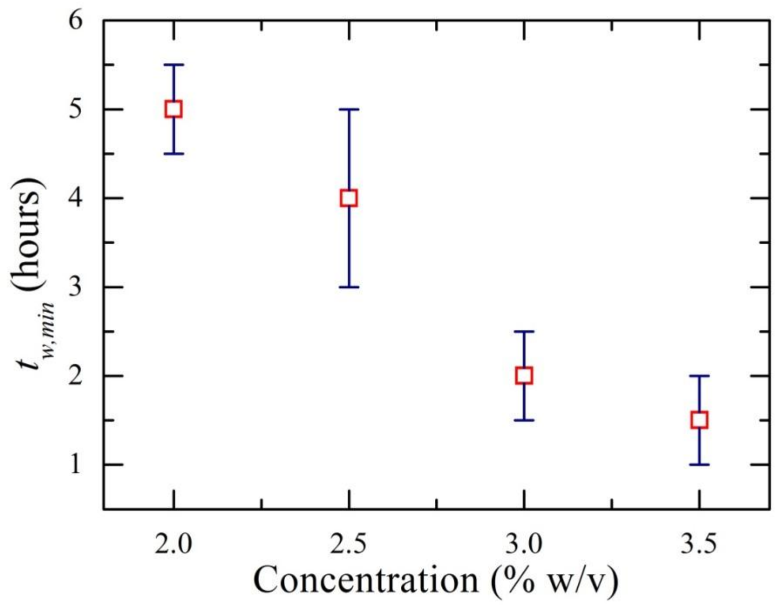

Figure S1: The waiting times associated with the minima in τ_1 ($t_{w,min}$) *vs.* concentration of Laponite.

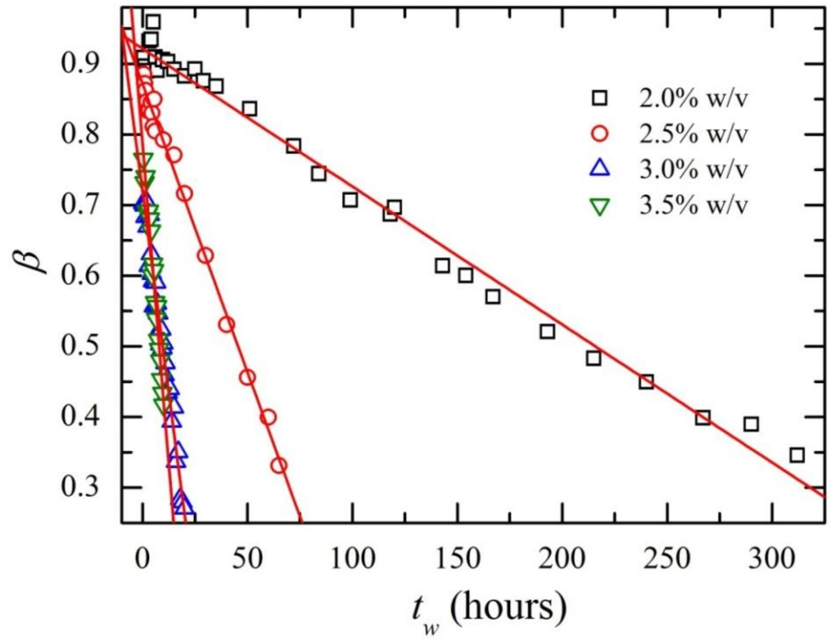

Figure S2: The stretching exponent, *β*, *vs.* waiting time, t_w, for 2.0% w/v (□), 2.5% w/v (○), 3.0% w/v (Δ) and 3.5% w/v (∇) Laponite suspensions. The solid lines are linear fits.

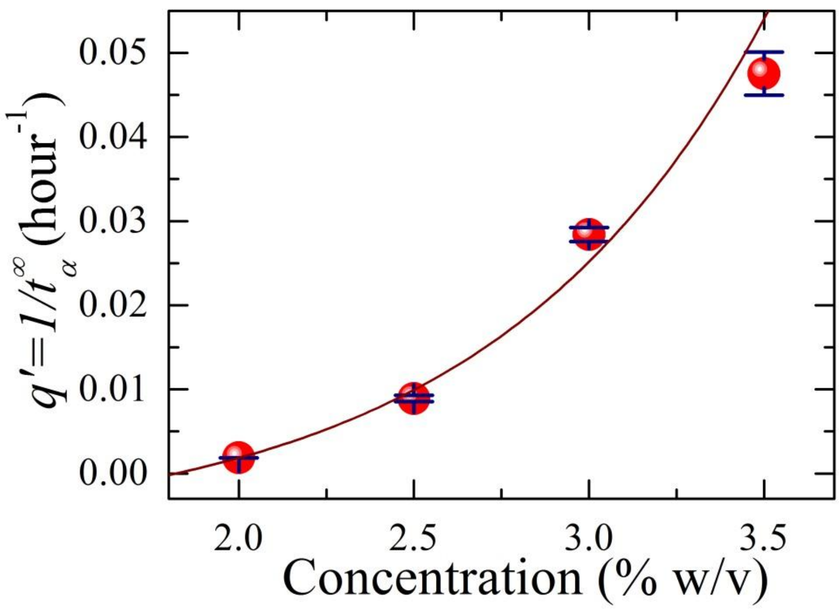

Figure S3: The rate $q'(=1/t_\alpha^\infty)$ at which the system approaches the glass transition is plotted *vs.* Laponite concentration. The solid line is an exponential fit.

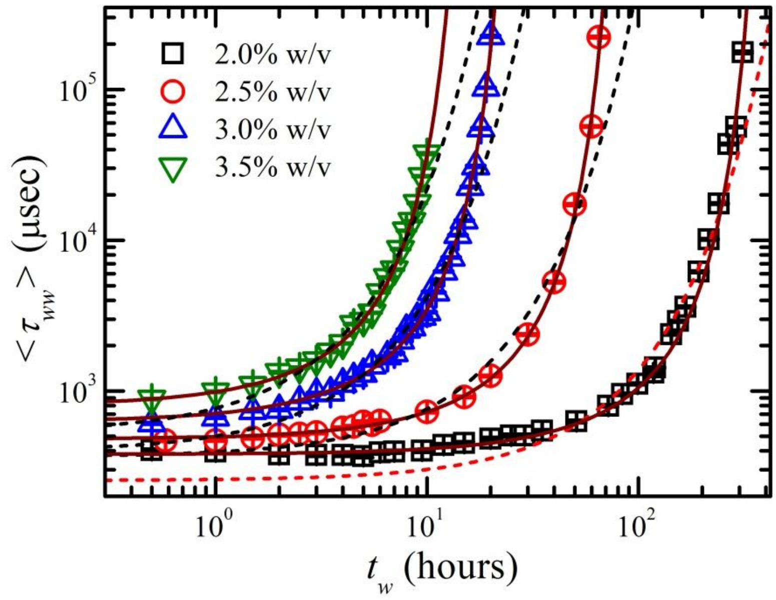

Figure S4: The mean slow relaxation times, $<\tau_{ww}>$, *vs.* waiting time, t_w are plotted for four different concentrations of Laponite The solid lines show fits to the modified VFT functions, $<\tau_{ww}>=<\tau_{ww}>^0 \exp\left(\frac{Dt_w}{t_\alpha^\infty - t_w}\right)$ (equation 3) and the dashed lines show fits to the exponential function $<\tau_{ww}>=<\tau_{ww}>^0 \exp(At_w)$.

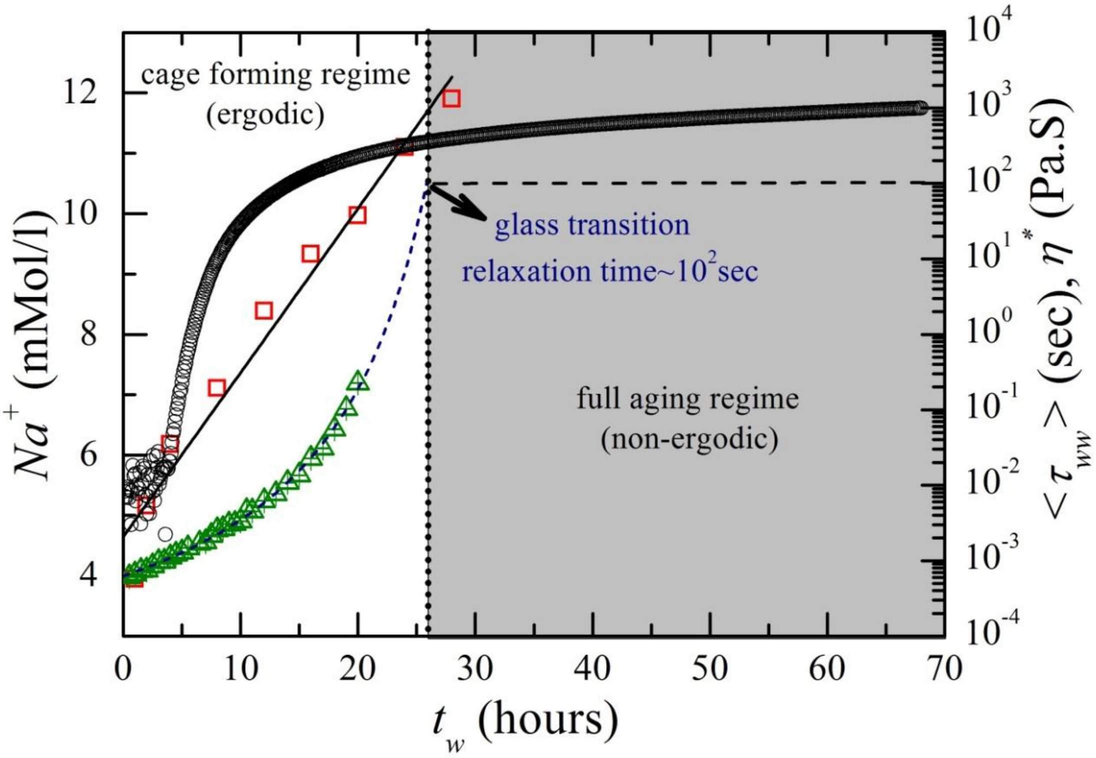

Figure S5: The simultaneous time-evolution of the sodium ion concentration (□), the complex viscosity (o) and the mean slow relaxation time (Δ) of a 3.0% w/v Laponite sample. The vertical dotted line indicates the glass transition as defined by Angell in reference [3] of the manuscript. Sodium ion concentration is measured with waiting time by a Eutech CyberScan 2100 pH/ion meter equipped with a sodium ion selective electrode (ROSS Sure-Flow).

Calculations of activation energies

Derivation of equation 5:

For a supercooled liquid, $\tau_1 \sim \exp(E / k_B T)$ [Ngai, *J. Chem. Phys.*, ***109***, *6982* (1998)].

Comparison with equation 2 of our paper yields: $\dfrac{E}{k_B T} = \dfrac{t_w}{t_\beta^\infty}$

The one-to-one mapping (*1/T* ↔ t_w) demonstrated in this paper (equations 2 and 3 of the manuscript, plotted in figures 2(a) and 2(b)) yields the following expression:

$$E = \frac{k_B c_1}{t_\beta^\infty}$$

E can therefore be written in terms of t_β^∞. This is plotted in the inset of figure S5. The constant c_1 in the above equation has the dimension of [time]×[temperature].

Derivation of equation 6:

According to reference 50:

$$E_{VFT} = k_B \left[\frac{d(\ln \tau)}{d(1/T)} \right]$$

Substituting $\tau = \tau_0 \exp\left(\frac{DT_0}{T - T_0}\right)$ [Reference: C. A. Angell, J. Res. Natl. Inst. Stand. Technol. 102, 171 (1997)] in the above expression for E_{VFT}, we get, $E_{VFT} = k_B\left[\frac{DT_0T^2}{(T-T_0)^2}\right]$

Substituting $c_2/T = t_w$ and $c_2/T_0 = t_\alpha^\infty$, we obtain

$$E_{VFT} = k_B c_2\left[\frac{D}{t_\alpha^\infty\left(1 - t_w/t_\alpha^\infty\right)^2}\right]$$

E_{VFT} can therefore be written in terms of t_w and t_α^∞. This is plotted in the figure S5. The constant c_2 in the above equation has the dimension of [time]×[temperature].

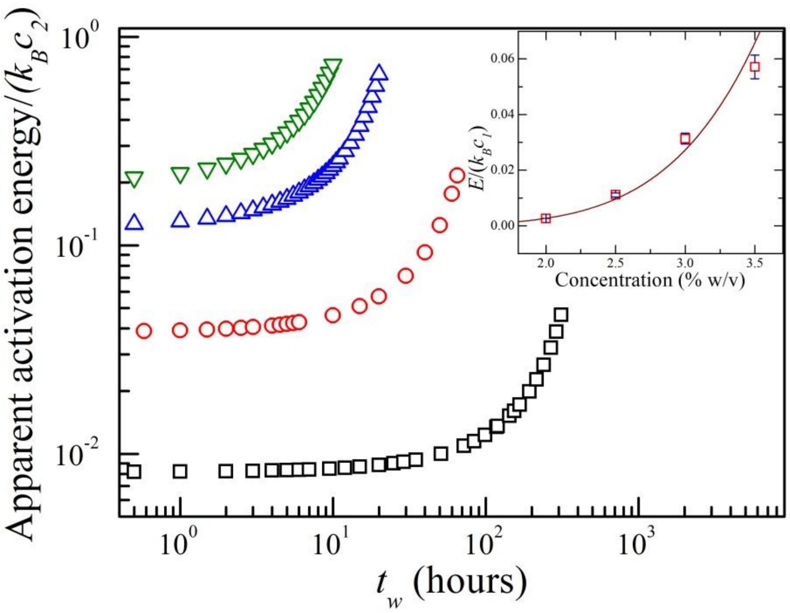

Figure S6: The normalized apparent activation energy (E_{VFT}) associated with the α-relaxation process *vs.* waiting time (t_w) for 2.0% w/v (□), 2.5% w/v (○), 3.0% w/v (Δ) and 3.5% w/v (∇) Laponite suspensions. In the inset, the normalized activation energy (E) associated with the β-relaxation process is plotted *vs.* Laponite concentration c. The solid line is a power law fit ($E \sim c^{5.7\pm0.3}$).

Table T1: Estimates of t_α^∞, t_β^∞ and t_g *vs.* Laponite concentration

Concentration of Laponite (% w/v)	t_α^∞ (hours)	t_β^∞ (hours)	t_g (hours)
2.0	538.0	375.0	397.0
2.5	112.3	89.8	83.1
3.0	35.2	31.9	25.8
3.5	21.0	17.5	15.5

Calculation of the width parameter (α_1) and non-Gaussian parameter (α_2):

Distribution of α-relaxation is given by,

$$\rho_{ww}(\tau) = \frac{\tau_{ww}}{\pi\tau^2}\sum_{k=0}^{\infty}(-1)^k \sin(\pi\beta k)\Gamma(\beta k+1)\left(\frac{\tau}{\tau_{ww}}\right)^{\beta k+1}$$

and the n^{th} moment of the distribution is given by,

$$\left\langle \tau_{ww}^n \right\rangle = \frac{\tau_{ww}^n}{\beta}\frac{\Gamma\left(\frac{n}{\beta}\right)}{\Gamma(n)}$$

The width parameter is given by:

$$\alpha_1 = \frac{\left\langle \tau_{ww}^2 \right\rangle - \left\langle \tau_{ww} \right\rangle^2}{\left\langle \tau_{ww} \right\rangle^2}$$

and the non-Gaussian parameter is given by:

$$\alpha_2 = \frac{3\left\langle \tau_{ww}^4 \right\rangle}{5\left\langle \tau_{ww}^2 \right\rangle^2} - 1$$

Table T2: Calculations of the width parameter (α_1)

t_w (hours) 2.0% w/v	α_1 2.0% w/v	t_w (hours) 2.5% w/v	α_1 2.5% w/v	t_w (hours) 3.0% w/v	α_1 3.0% w/v	t_w (hours) 3.5% w/v	α_1 3.5% w/v
0.5	1.10462	0.58	1.13886	0.5	1.25068	0.5	1.37439
1	1.12202	1	1.15897	1	1.27299	1	1.46605
2	1.13112	1.5	1.17539	1.5	1.29918	1.5	1.44102
3	1.07323	2	1.20227	2	1.27879	2	1.60044
4	1.07089	2.5	1.22575	2.5	1.34573	2.5	1.61353
5	1.04068	3	1.22457	3	1.44056	3	1.59831
6	1.10304	4	1.20479	3.5	1.34722	3.5	1.64067
7	1.12992	4.5	1.232	4	1.4582	4	1.71128
9.5	1.10856	5	1.27024	4.5	1.50602	4.5	2.04392
12	1.11274	5.5	1.19652	5	1.54408	5	1.94416
15	1.1286	6	1.28325	5.5	1.66356	5.5	1.99879
20	1.14278	10	1.30975	6.5	1.64615	6	2.32306
25	1.12814	15	1.36033	7	1.72245	6.5	2.53988
29	1.1536	20	1.51513	7.5	1.80119	7	2.37135
35	1.16563	30	1.87861	8	1.90796	7.5	2.88049
51	1.22121	40	2.62192	8.5	1.95065	8	3.05582
72	1.32969	50	3.76481	9	1.95813	8.5	3.30927
84	1.43005	60	5.45427	9.5	2.02253	9	3.82171
99	1.54471	65	10.26433	10	2.04866	9.5	4.31219
118	1.61584			10.5	2.35735	10	4.81554
120	1.5795			11	2.2524		
143	1.96069			12	2.61647		
154	2.04236			13	2.66661		
167	2.26105			14	3.10576		
193	2.7322			15	3.17541		
215	3.25971			16	4.13807		
240	3.91152			17	4.26916		
267	5.50604			18	5.68258		
290	5.90128			19	6.97967		
312	8.77478			20	8.16226		

Table T3: Calculations of non-Gaussian parameter (α_2)

t_w (hours) 2.0% w/v	α_2 2.0% w/v	t_w (hours) 2.5% w/v	α_2 2.5% w/v	t_w (hours) 3.0% w/v	α_2 3.0% w/v	t_w (hours) 3.5% w/v	α_2 3.5% w/v
0.5	-0.24034	0.58	-0.18569	0.5	0.00769	0.5	0.24693
1	-0.21244	1	-0.15234	1	0.0488	1	0.44208
2	-0.19752	1.5	-0.12475	1.5	0.09815	1.5	0.38702
3	-0.28886	2	-0.0785	2	0.05847	2	0.75497
4	-0.29243	2.5	-0.03697	2.5	0.18898	2.5	0.78711
5	-0.33765	3	-0.03916	3	0.38603	3	0.74938
6	-0.24266	4	-0.07439	3.5	0.19166	3.5	0.85466
7	-0.19965	4.5	-0.02609	4	0.42451	4	1.03724
9.5	-0.23406	5	0.04385	4.5	0.53147	4.5	2.02054
12	-0.22773	5.5	-0.08901	5	0.61953	5	1.70427
15	-0.2023	6	0.06826	5.5	0.91324	5.5	1.8752
20	-0.17934	10	0.11857	6.5	0.86864	6	3.00386
25	-0.20344	15	0.21824	7	1.06717	6.5	3.86843
29	-0.16157	20	0.55228	7.5	1.28284	7	3.18867
35	-0.14165	30	1.50652	8	1.59404	7.5	5.40767
51	-0.04568	40	4.21947	8.5	1.72437	8	6.28734
72	0.15727	50	10.45309	9	1.74562	8.5	7.66252
84	0.36368	60	24.4168	9.5	1.95025	9	10.82908
99	0.62136	65	96.6505	10	2.03587	9.5	14.34769
118	0.79297			10.5	3.1348	10	18.45914
120	0.70384			11	2.74087		
143	1.7556			12	4.19569		
154	2.01552			13	4.41496		
167	2.77215			14	6.54793		
193	4.71058			15	6.92068		
215	7.38437			16	13.04328		
240	11.43881			17	14.01893		
267	24.86205			18	26.74489		
290	28.99567			19	42.04245		
312	69.0407			20	59.04992		

Data acquired at a different scattering angle (θ = 60°)

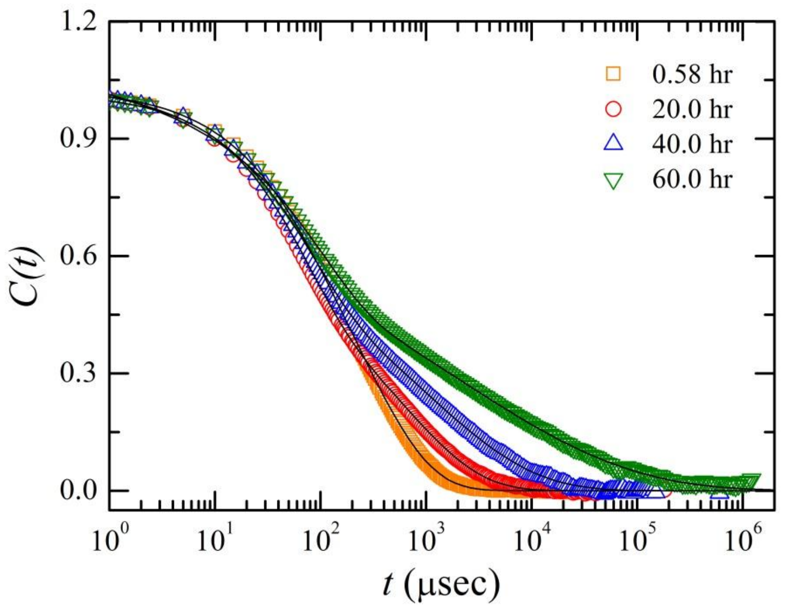

Figure S7: Intensity autocorrelation functions for a 2.5% w/v sample at scattering angle θ = 60° for four different waiting times. The solid lines are fits to equation 1.

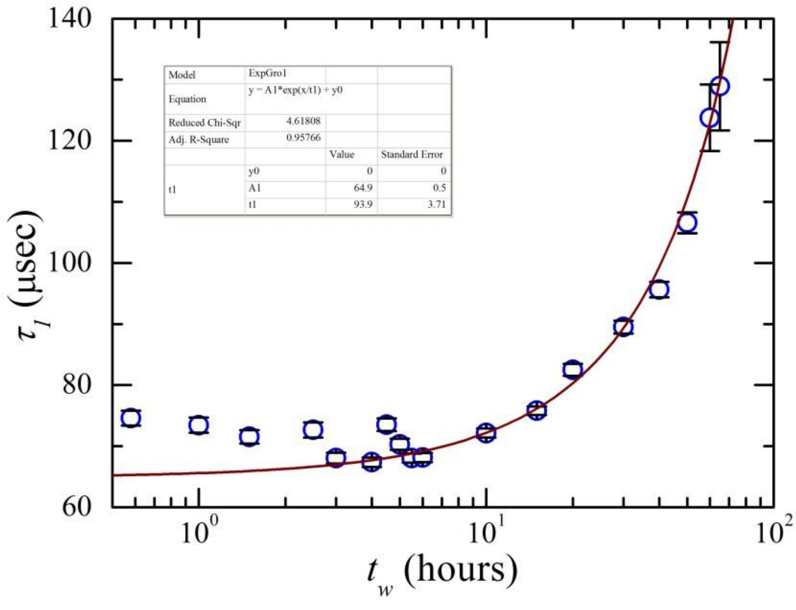

Figure S8: The fast relaxation timescales of a 2.5% w/v Laponite sample, extracted from fits of the data plotted in Figure S5 to equation 1, versus time since preparation of the sample. The solid line is a fit to the modified Arrhenius form (equation 2). A decrease in τ_1 at very early times, followed by an eventual increase, as highlighted in Figure 2(a), is also present.

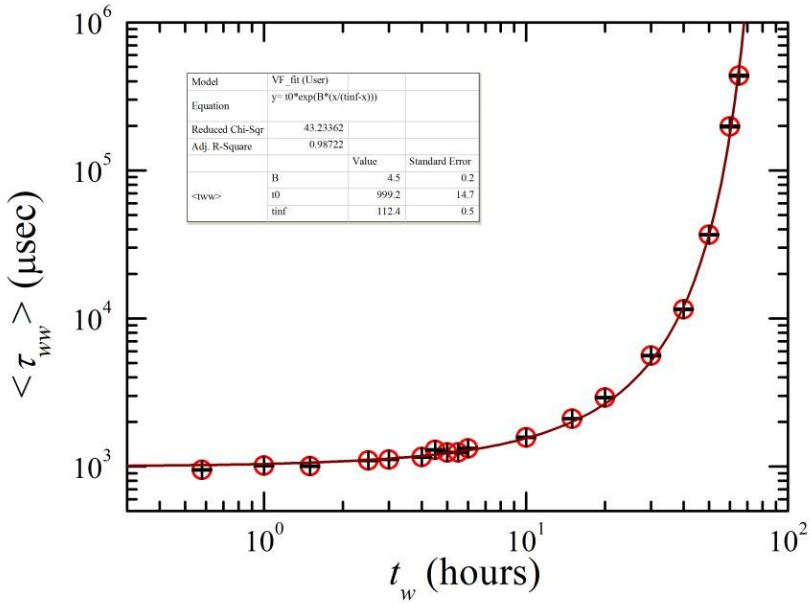

Figure S9: The mean slow relaxation timescales of a 2.5% w/v Laponite sample, extracted from fits of the data plotted in Figure S5 to equation 1, versus time since preparation of the sample. The solid line is a fit to the modified VFT form (equation 3).

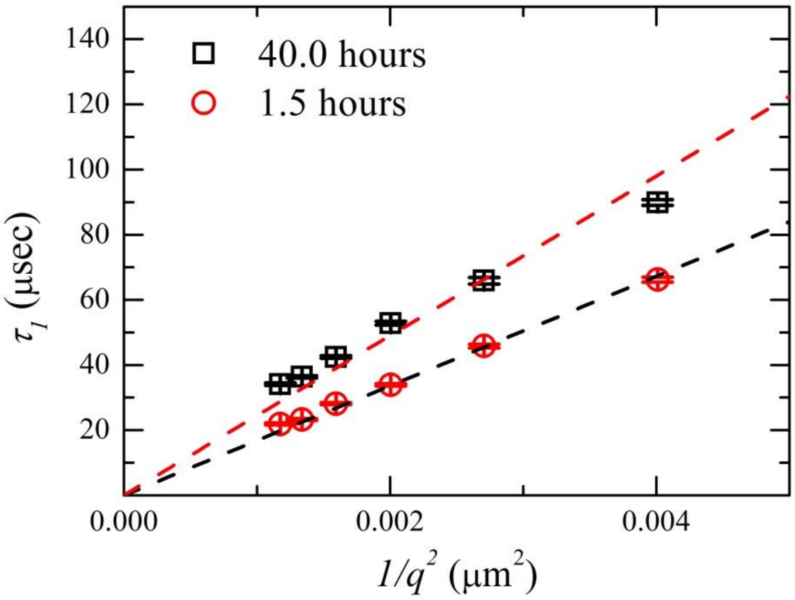

Figure S10: The diffusive dynamics of the fast relaxation time (τ_1) is shown above for a 2.5% w/v Laponite sample for two different waiting times t_w. The dashed lines are linear fits passing through the origin.

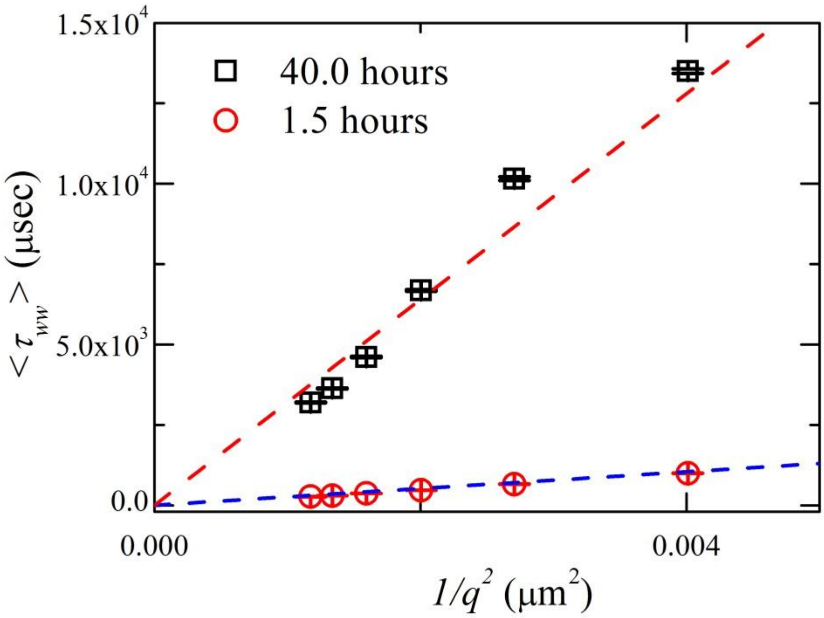

Figure S11: The diffusive dynamics of the mean slow relaxation time is shown above for a 2.5% w/v Laponite sample for two different waiting times t_w. The dashed lines are linear fits passing through the origin.

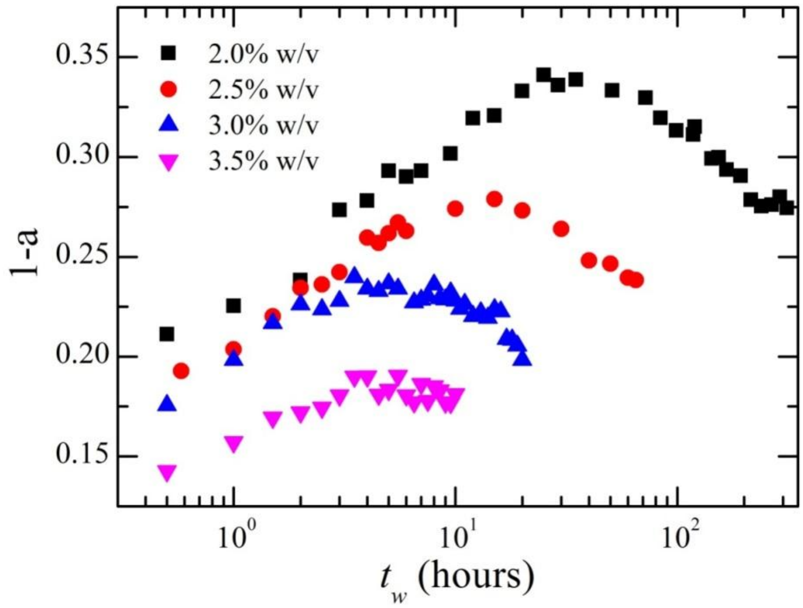

Figure S12: The fitting parameter (1-a) in equation (1) is plotted for different concentrations of Laponite.