Supporting information for

Viscoelastic cluster densification in sheared colloidal gels

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S1 Stress jump measurements

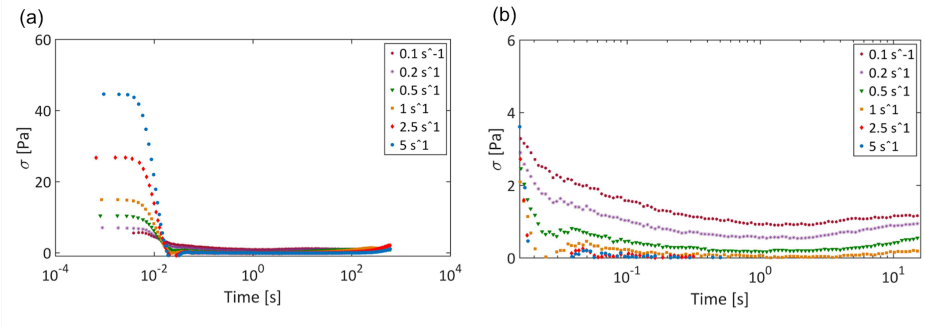

Figure S.1: Stress jump experiments for a 10 vol% suspension of PMMA particles in a linear PDMS-decalin matrix. (a) Step down in shear rate to a final value of 0.0001 s^{-1} after preshear steps at several initial values of shear rate $\dot{\gamma}_i$, which are shown in the legend. (b) Initial part of the stress relaxation curves corresponding to the raw data used to calculate the elastic contributions. The measurements have been performed with a force rebalanced transducer rheometer (ARES G2), in order to catch fast transients. As expected, the higher the $\dot{\gamma}_i$ the greater the stress jump, because of the higher hydrodynamic stresses. The relaxation times increase when the $\dot{\gamma}_i$ is decreased as a direct consequence of the increased aggregate size.

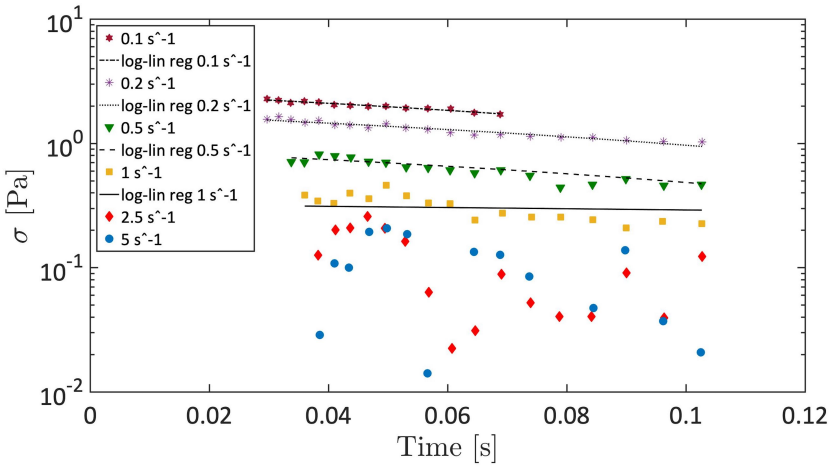

Figure S.2: Log-linear regression of the relaxation data between 30 ms and 70-100 ms for a 10 vol% suspension of PMMA particles in a linear PDMS-decalin matrix. The intercept is the stress at time zero, i.e. the time at which the flow has been arrested, and corresponds to the elastic contribution. After 100 ms, phenomena such as floc build-up at rest start interfering with the viscoelastic response of the material, and thus a deviation from single exponential behaviour is observed. The maximum time used for the data fitting depends on the initial shear rate.

S2 Stress contributions in the viscoelastic matrix

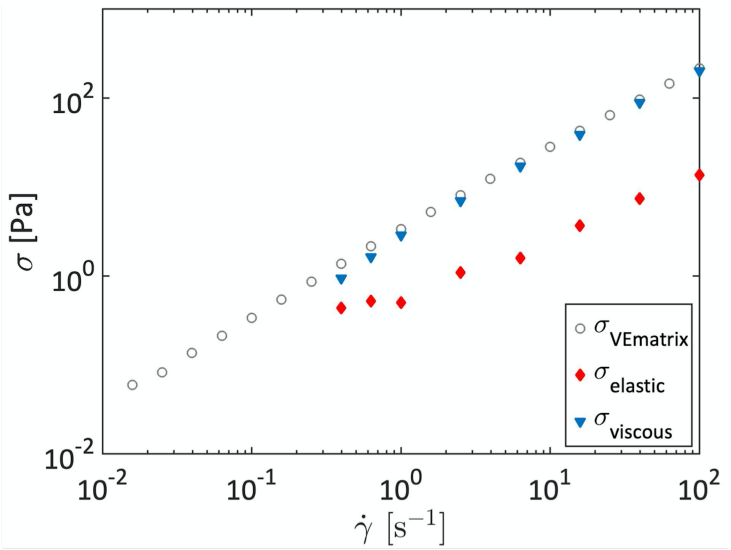

Figure S.3: Steady state rheology for the viscoelastic matrix. Total stress at steady state (open symbols) reported along with elastic (red filled symbols) and viscous (blue filled symbols) contribution for a branched PDMS-decalin matrix. The two contributions have been measured performing stress jump experiments.

S3 Calculation of the fractal dimension

The fractal dimension was determined based on the experimental data using the model of Wu and Morbidelli [1]. The model relates the fractal dimension of the flocs with two scaling exponents, $f(D_f) \equiv m_\gamma$ and $f(D_f) \equiv m_G$, obtained from oscillatory shear rheometry. More precisely, $f(D_f) = m_\gamma$ is the scaling exponent of the critical strain amplitude γ_{crit}, which denotes the end of the linear viscoelastic deformation regime at a given frequency, whereas $f(D_f) = m_G$ is the scaling exponent of the frequency-independent plateau value of the elastic modulus G' in the linear viscoelastic regime. The two scaling exponents derived by Wu and Morbidelli [1] are defined as:

$$m_\gamma = \frac{2 - \beta_{Wu}}{3 - D_f} \tag{S.1}$$

$$m_G = \frac{\beta_{Wu}}{3 - D_f} \tag{S.2}$$

where β_{Wu} is a parameter dependent on the relative contributions of intra-floc and backbone elasticities. By combination of Eq. S.1 and S.2, an expression of D_f as function of the two scaling exponents is obtained as:

$$D_f = \frac{3 \cdot (m_\gamma + m_G) - 2}{m_\gamma + m_G}. \tag{S.3}$$

Note that m_γ and m_G correspond to the slopes of the curves in Fig. S.4 (a) and (b) respectively.

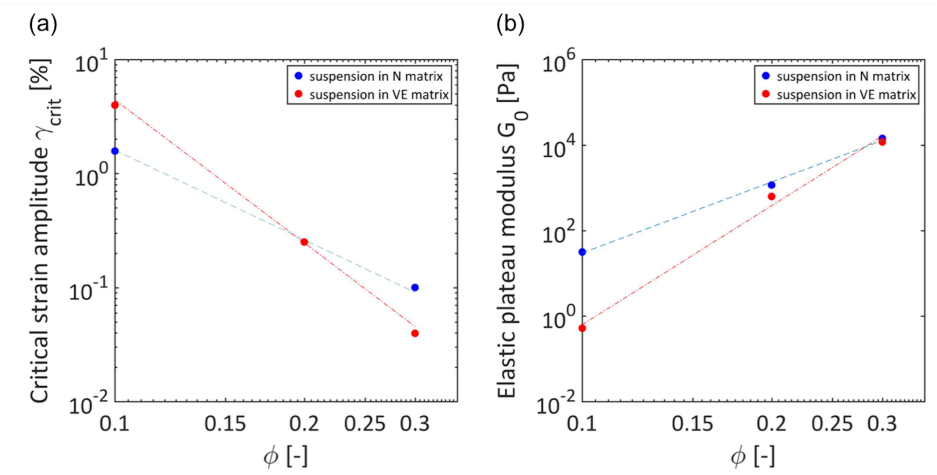

Figure S.4: Fractal model analysis for the determination of the fractal dimension D_f. (a) Critical strain amplitude γ_{crit}, which was defined using the 95 % criterion, and (b) elastic plateau modulus G_0, as function of the solid volume fraction ϕ for both suspensions in Newtonian (N) and viscoelastic matrix (VE). Every data point was obtained performing strain amplitude measurements at a frequency $\omega = 0.25$ rad/s while increasing the strain amplitude γ from 0.001 to 20%. Increasing the volume fraction the limit of linearity γ_{crit} is decreasing whereas G_0 is increasing.

References

[1] H. Wu and M. Morbidelli. A model relating structure of colloidal gels to their elastic properties. *Langmuir*, 17(4):1030–1036, 2001.