Supplementary material for

"Colloidal Gels Tuned by Oscillatory Shear"
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Asakura–Oosawa potential:

We model a tunable attraction strength and range via Asakura–Oosawa potential:

$$U_{AO}(r) = -\Pi V_{overlap} \qquad \text{Eq. S1}$$

Where Π is the osmotic pressure of depletant and $V_{overlap}$ is the overlap volume of the two excluded volumes surrounding the pair of particles which has been modified for particles of different sizes. We modify the potential in a small range around contact, to have a flat potential. This change is made to reduce the amount of force the particles exert on each other when in direct contact, leading to more stable simulations. To this end, we have used an exponential weighting function as follows:

$$U_{AO-\mathrm{modified}}(r) = U_{AO} + \alpha(U_{AO} - U_0) \qquad \text{Eq. S2}$$

where,

$$\alpha = \exp[-(\frac{r-(r_1+r_2)}{2L})^{\nu}] \qquad \text{Eq. S3}$$

where L represents the region where the modified AO potential deviates from the standard one. The exponent ν identifies the deviation from AO potential for distances r<2(R+L). Fig. S1 shows the comparison between true AO potential and the modified one with L=0.01 and ν=3.5 used in the present study. This results in the constant attraction strength where there are no attractive forces for r<2R+0.01. Increasing the exponent ν results in a sharper deviation from the AO potential.

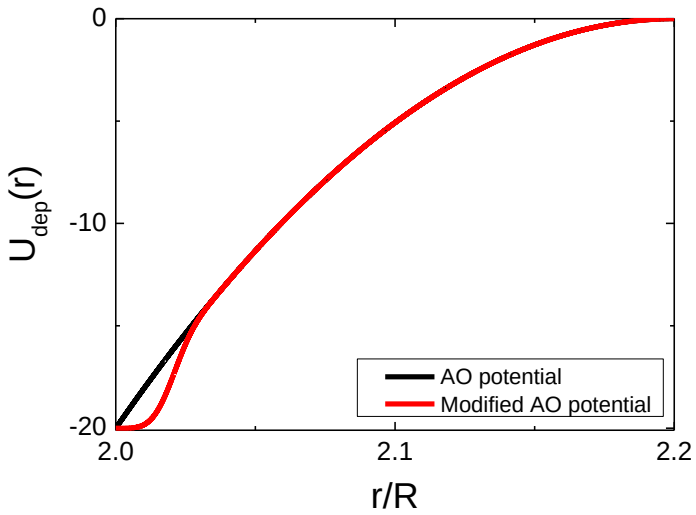

Fig. S1: Plot of the Asakura–Oosawa potential (solid black curve) and the modified Asakura–Oosawa potential utilized in the current study (solid red curve) with the polymer-to-colloid size ratio $\xi = R_g/R = 0.1$ and attraction strength at contact of $U_{dep}(2R) = -20\ k_BT$.

Calculation of the void volume:

We define the void volume as the volume of a sphere with its center in the empty space and the radius equal to the nearest particle surface as shown schematically in Fig. S2. The simulation box is divided to the small pieces in which dV << R^3. Probing all the empty space gives the distribution of voids and provides information on the structural heterogeneity on all length scales. With this method a specific portion of the space might counts several times in its calculations which makes it robust in mapping the heterogeneous distribution of void sizes. The average void volume <VV>, used in the present study is calculated from the probability density function as:

$$\langle V V \rangle = \int_0^\infty P(V) dV \qquad \text{Eq. S4}$$

where P(V) is the void volume distribution

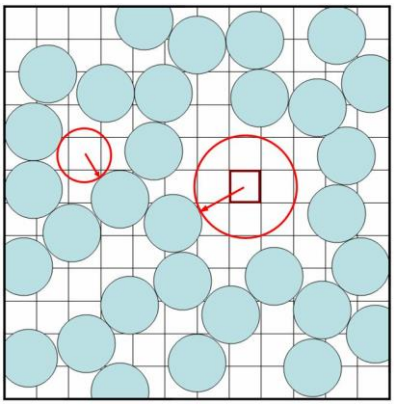

Fig S2: A two dimensional representative of the void volume calculation as discussed in the text.

Distribution of bonds under oscillatory pre-shear:

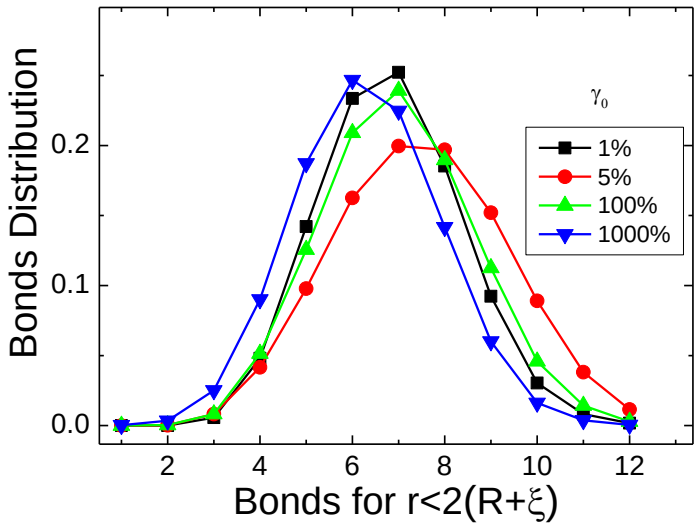

Fig. S3: The bonds distribution under shear as a function of strain amplitude (γ_0) as indicated taken from BD simulations at φ=0.44, $U_{dep}(2R) = -20k_BT$, $\xi = 0.1$.

Effects of oscillatory preshear and preshear history in nonlinear response: Large amplitude oscillatory shear tests in experiments (fig. S4 and S5) and start-up shear test in BD simulations (fig S6).

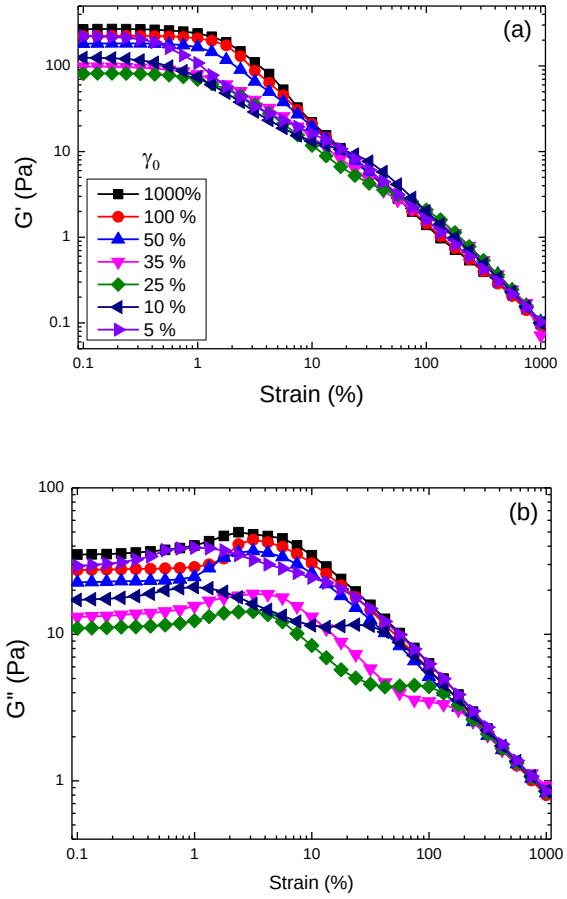

Fig. S4. Experiments: (a) Storage modulus and (b) loss modulus versus strain at frequency ω=0.1 rad/s for different pre-shearing strain amplitudes as indicated.

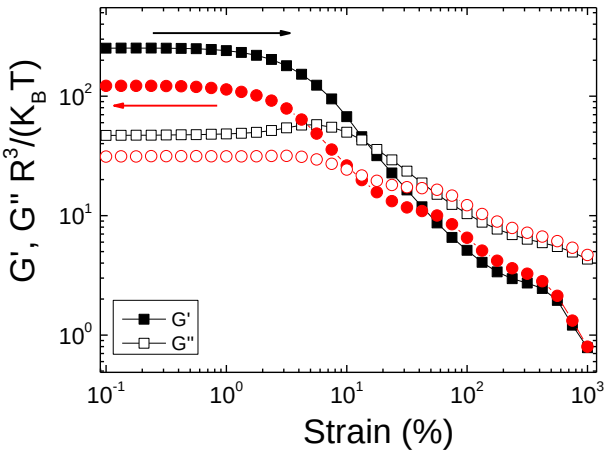

Fig. S5: Storage modulus G' (solid symbols) and loss modulus G'' as a function of strain amplitude. Black curves correspond to the gel prepared by rejuvenation at strain amplitude of 800% and frequency 10 rad/s. Red curves correspond to strain sweeps from high to low strain amplitudes. Arrows indicate the direction of strain sweep.

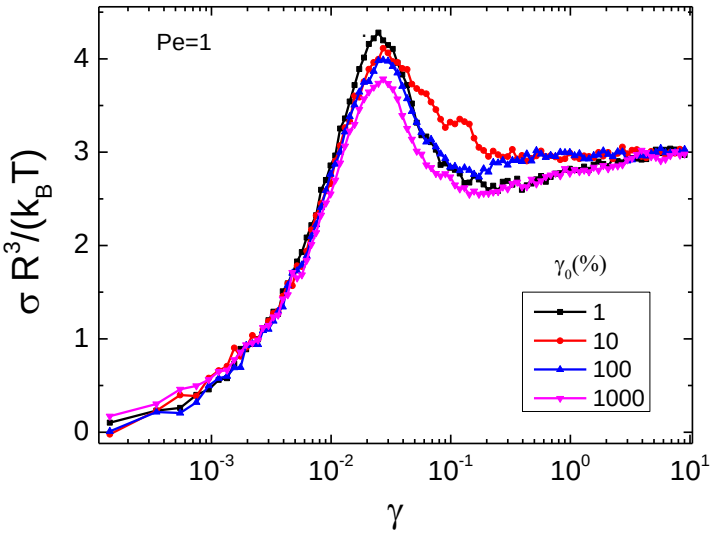

Fig. S6: BD simulations of step rate tests conducted at Pe=1 on a colloidal gel at 100 t_B after flow cessation of different oscillatory strain amplitudes as indicated. The test performed on a gel with φ=0.44, $U_{dep}(2R) = -20k_BT$ and $\xi = 0.1$.