# Supplementary Information for
# Thermodynamics Description of Startup Flow of Soft Particles Glasses

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## Elastic energy as a function of strain on linear scales

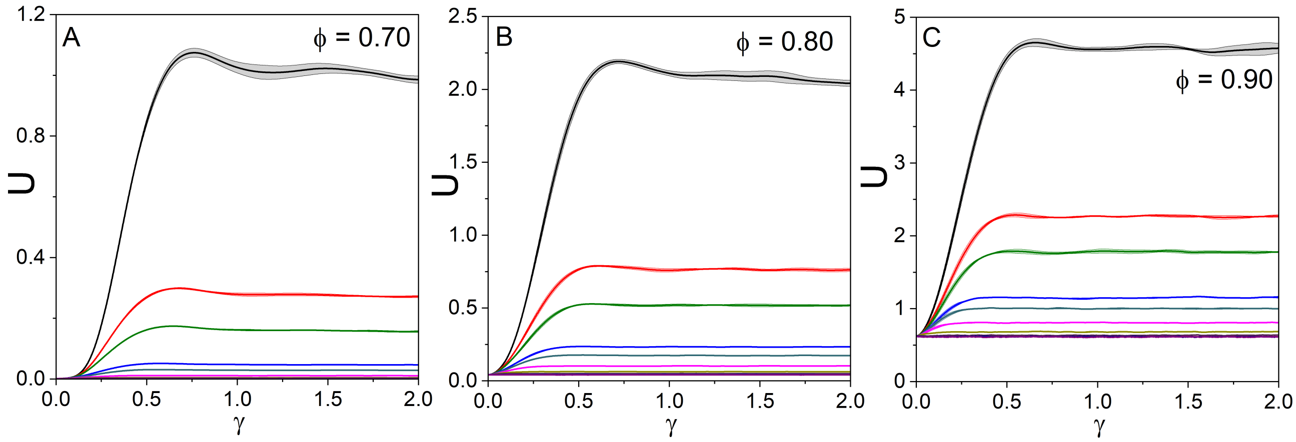


Fig. S1. Elastic energy, $U$, as a function of strain, $\gamma$, for different volume fractions in a linear scale plot.

**Strain dependence of first and second normal stresses differences and osmotic pressure**

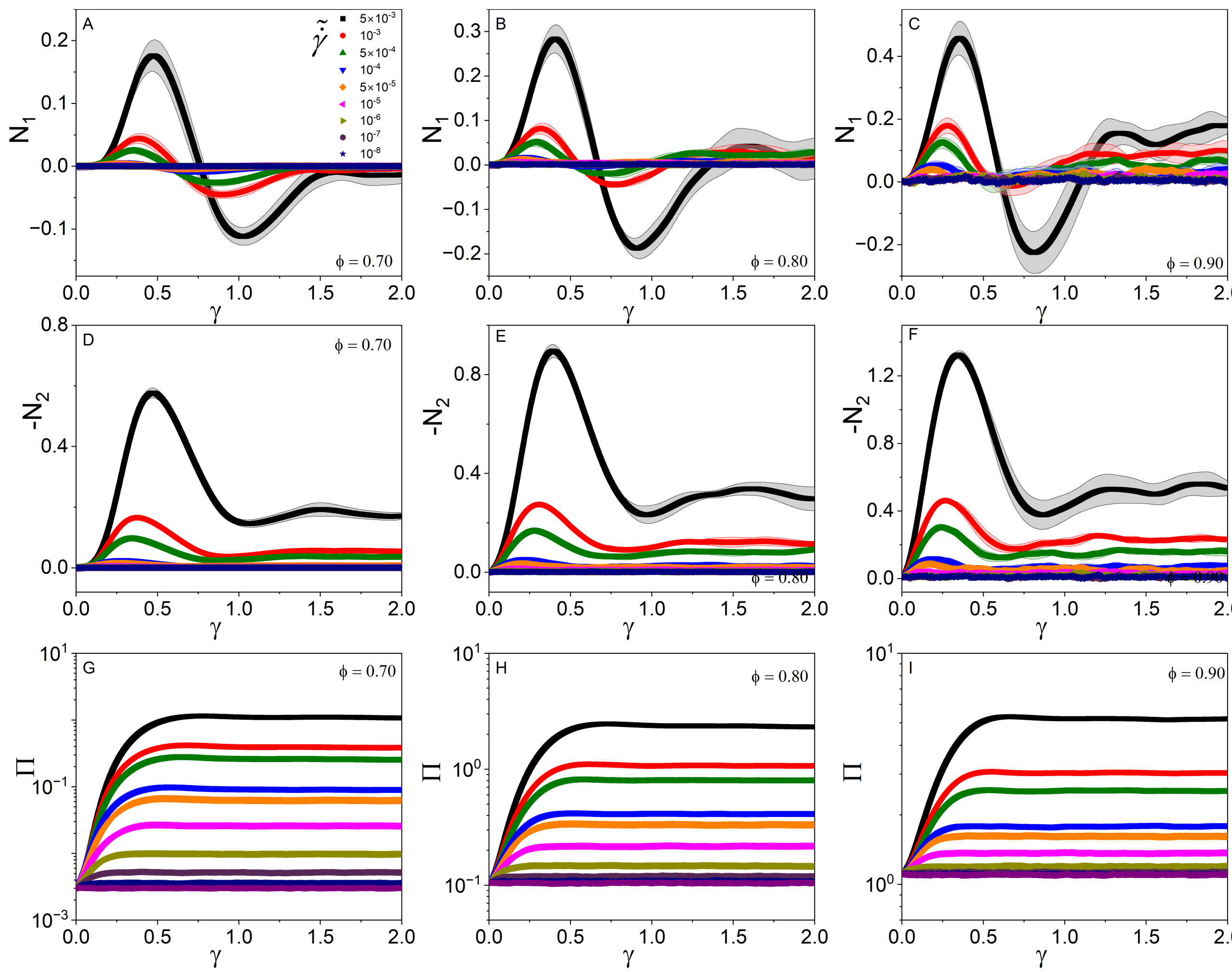


Fig. S2. First normal stress difference, $N_1$, (top row), second normal stress difference, $-N_2$, (middle row), and osmotic pressure, $\Pi$, (bottom row), as a function of strain, $\gamma$, for suspensions with volume fraction of $\phi = 0.70$, 0.80, and 0.90.

**Fit parameters for $S^E - \dot{\gamma}$ curves**

TABLE I. Parameters determined from fitting $S^E - \dot{\gamma}$ curves to $-S^E = a - b\ln(\dot{\gamma} + c)$ at different volume fractions.

| Volume fraction ($\phi$) | $a$ | $b$ | $c$ |
|---|---|---|---|
| 0.70 | $2.140 \pm 0.087$ | $0.495 \pm 0.009$ | $4.24 \times 10^{-8} \pm 1.16 \times 10^{-8}$ |
| 0.75 | $2.106 \pm 0.169$ | $0.560 \pm 0.020$ | $8.00 \times 10^{-7} \pm 2.45 \times 10^{-7}$ |
| 0.80 | $2.415 \pm 0.172$ | $0.568 \pm 0.022$ | $1.64 \times 10^{-6} \pm 4.57 \times 10^{-7}$ |
| 0.85 | $2.542 \pm 0.168$ | $0.589 \pm 0.022$ | $4.70 \times 10^{-6} \pm 1.10 \times 10^{-6}$ |
| 0.90 | $3.011 \pm 0.244$ | $0.558 \pm 0.031$ | $9.96 \times 10^{-6} \pm 2.66 \times 10^{-6}$ |

**Flow curve with Herschel Bulkley fit**

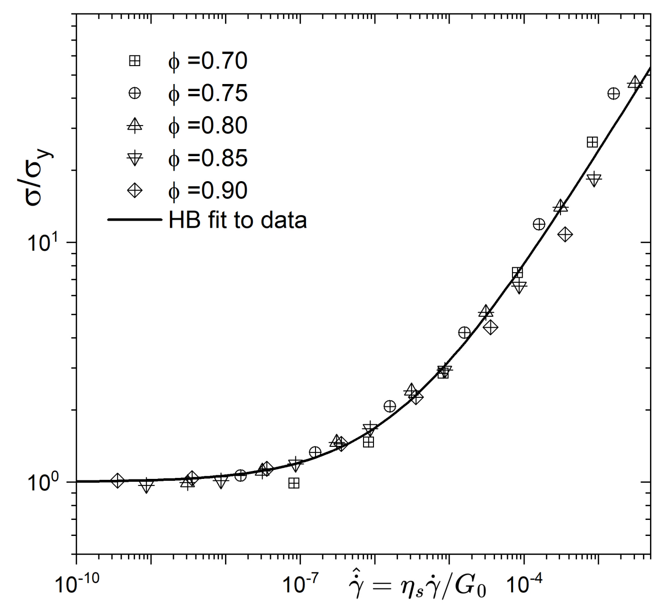


Fig. S3. Master curves of the dimensionless shear stress. The solid line is the Herschel-Bulkley (HB) equation according to $\sigma/\sigma_y = 1 + 789.28\hat{\dot{\gamma}}^{0.51\pm0.02}$.

### Two-dimensional pair distribution function

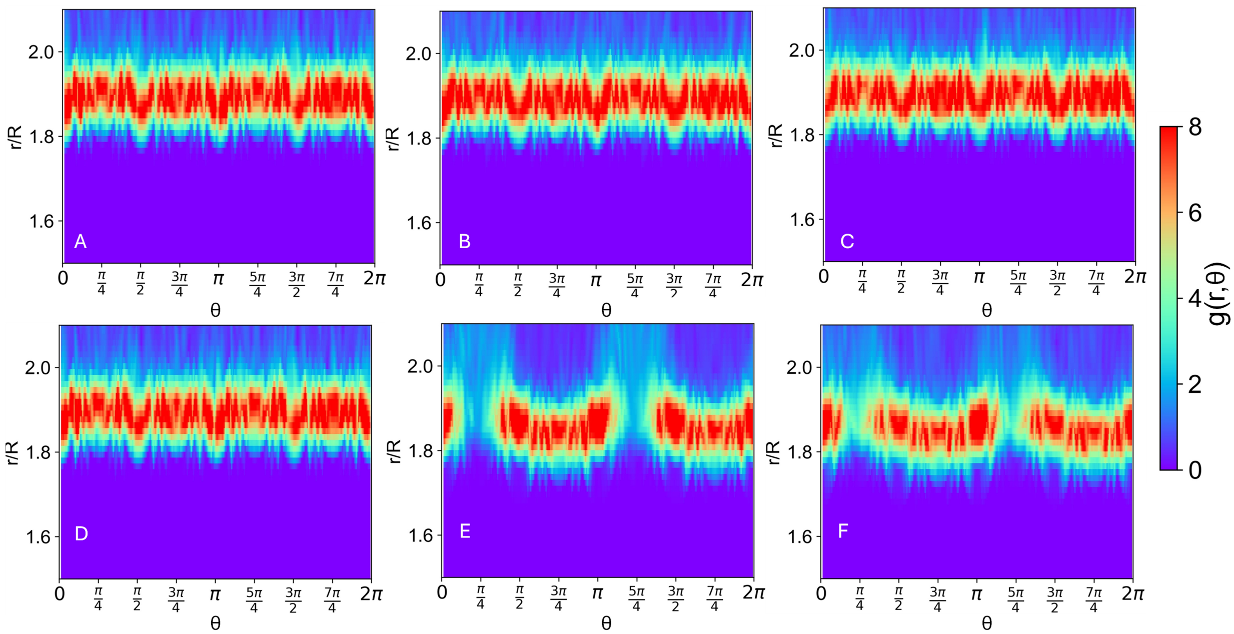


Fig. S4. Two-dimensional pair distribution function, $g(r, \theta)$, at shear rate of (A-C) $\tilde{\dot{\gamma}} = 10^{-9}$ and (D-F) $\tilde{\dot{\gamma}} = 10^{-4}$ for $\phi = 0.80$ at different strains:(A, D) $\gamma = 0$, (B, E) $\gamma = \gamma_p$, and (C, F) $\gamma = \gamma_{st}$.

**Expansion of pair distribution function based on spherical harmonics**

The pair distribution function $g(\mathbf{r})$ is expanded using spherical harmonics series, $Y_{lm}(\mathbf{n})$ [1, 2], as:

$$g(r) = g_s(r) + \sum_{l=1}^{\infty} \sum_{m=-l}^{l} g_{lm}(r) Y_{lm}(\theta, \phi), \tag{1}$$

in which the expansion coefficients are $g_{lm}(r)$ and are determined as $g_{lm}(r) = \frac{\int g(r) Y_{lm}(\mathbf{n}) d\mathbf{n}}{\int Y_{lm} Y_{lm}(\mathbf{n}) d\mathbf{n}}$, where $\mathbf{n}$ is the outward unit normal to a spherical surface and thus $d\mathbf{n} = \sin\phi d\phi d\theta$ is the solid angle element on the sphere (see Fig. S5 for the definition of $\theta$ and $\phi$). The spherical harmonics are given by:

$$Y_{lm}(\theta, \phi) = \sqrt{\frac{(2l+1)}{4\pi} \frac{(l-m)!}{(l+m)!}} P_{lm}(\cos\theta) e^{im\phi}, \tag{2}$$

where $P_{lm}(\cos\theta)$ is the associated Legendre function. It is more convenient to work with the real representation of the spherical harmonic function defined as:

$$S_{l,|m|}(\theta, \phi) = \sqrt{2}\,\mathrm{Re}[Y_{l|m|}(\theta, \phi)], \tag{3}$$

$$S_{l,-|m|}(\theta, \phi) = \sqrt{2}\,\mathrm{Im}[Y_{l|m|}(\theta, \phi)]. \tag{4}$$

These functions constitute an orthonormal basis set. Then the equation for $g(r)$ can be rewritten as:

$$g(r) = g_s(r) + \sum_{l=1}^{\infty} \sum_{m=-l}^{l} g_{lm}(r) S_{lm}(\theta, \phi). \tag{5}$$

The first few harmonics used are:

$$
\begin{aligned}
S_{00} &= \frac{1}{2\sqrt{\pi}}, \\
S_{20} &= \sqrt{\frac{5}{16\pi}}(3\cos^2\phi - 1), \\
S_{22} &= \sqrt{\frac{15}{16\pi}}\cos 2\theta \sin^2\phi, \\
S_{2,-2} &= \sqrt{\frac{15}{16\pi}}\sin 2\theta \sin^2\phi, \\
S_{40} &= \frac{3}{16\sqrt{\pi}}(35\cos^4\phi - 30\cos^2\phi + 3), \\
S_{42} &= \frac{3}{8}\sqrt{\frac{5}{\pi}}\cos 2\theta(7\cos^2\phi - 1), \\
S_{4,-2} &= \frac{3}{8}\sqrt{\frac{5}{\pi}}\sin 2\theta(7\cos^2\phi - 1), \\
S_{44} &= \frac{3}{16}\sqrt{\frac{35}{\pi}}\cos 4\theta \sin^2\phi, \\
S_{4,-4} &= \frac{3}{16}\sqrt{\frac{35}{\pi}}\sin 4\theta \sin^2\phi.
\end{aligned}
$$

These coefficients are shown at different stages of the startup flow at high (Fig. S6) and low (Fig. S7) shear rates for suspensions with a volume fraction of $\phi = 0.80$.

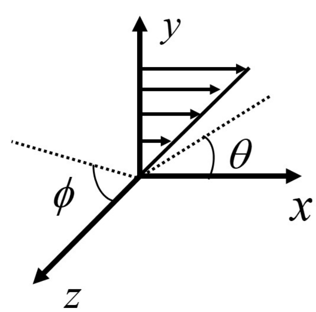


Fig. S5. Definitions of $\theta$ and $\phi$ angles in shear flow. $\theta$ is measured clockwise from the positive x-axis, while $\phi$ is measured from the z-axis. The velocity profile of the imposed shear flow is given as $\mathbf{u} = (\dot{\gamma}y, 0, 0)$, where $\dot{\gamma}$ is the shear rate.

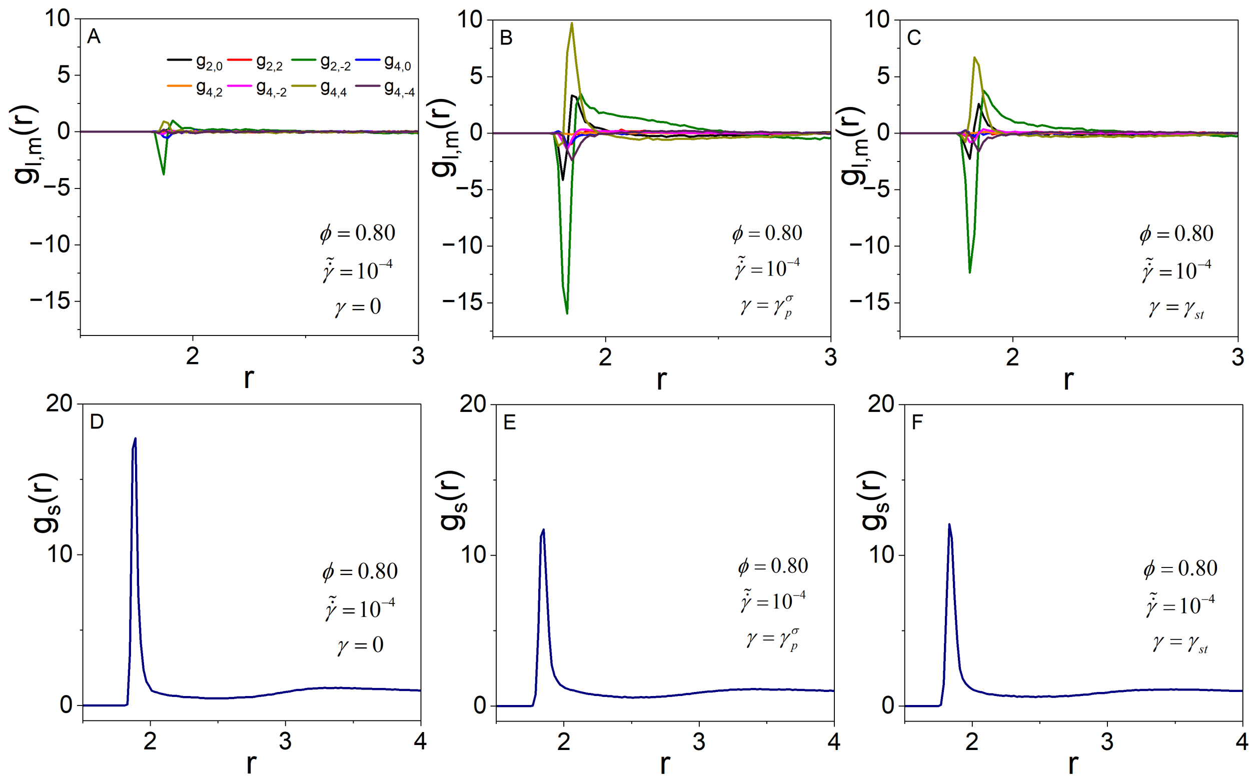


Fig. S6. Expansion of $g(\mathbf{r})$ based on spherical harmonics coefficients at (A and D) equilibrium, (B and E) peak stress, and (C and F) steady state (D-F shows the spherically symmetric contribution, $g_s(r)$) for suspensions with volume fraction of $\phi = 0.80$ at shear rate of $\tilde{\dot{\gamma}} = 10^{-4}$.

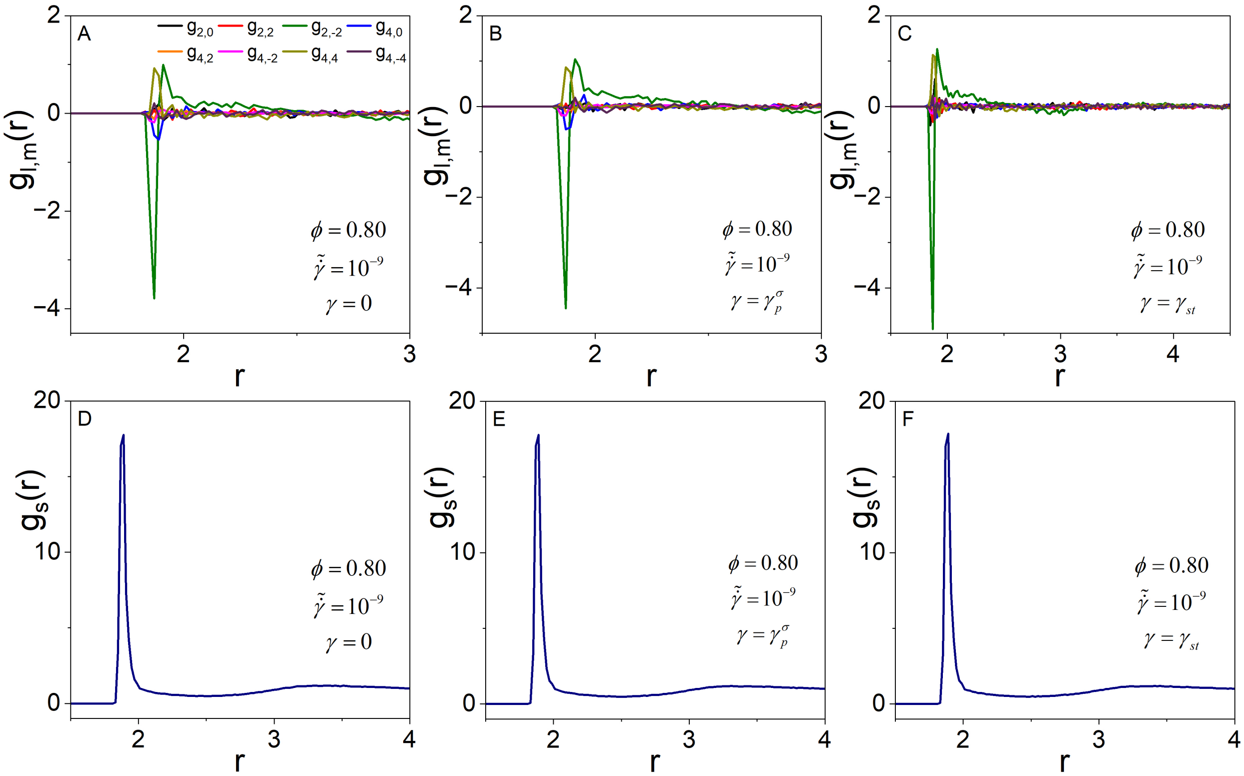


Fig. S7. Expansion of $g(\mathbf{r})$ based on spherical harmonics coefficients at (A and D) equilibrium, (B and E) peak stress, and (C and F) steady state (D-F shows the spherically symmetric contribution, $g_s(r)$) for suspensions with volume fraction of $\phi = 0.80$ at shear rate of $\tilde{\dot{\gamma}} = 10^{-9}$.

**Derivative of excess entropy as a function of strain**

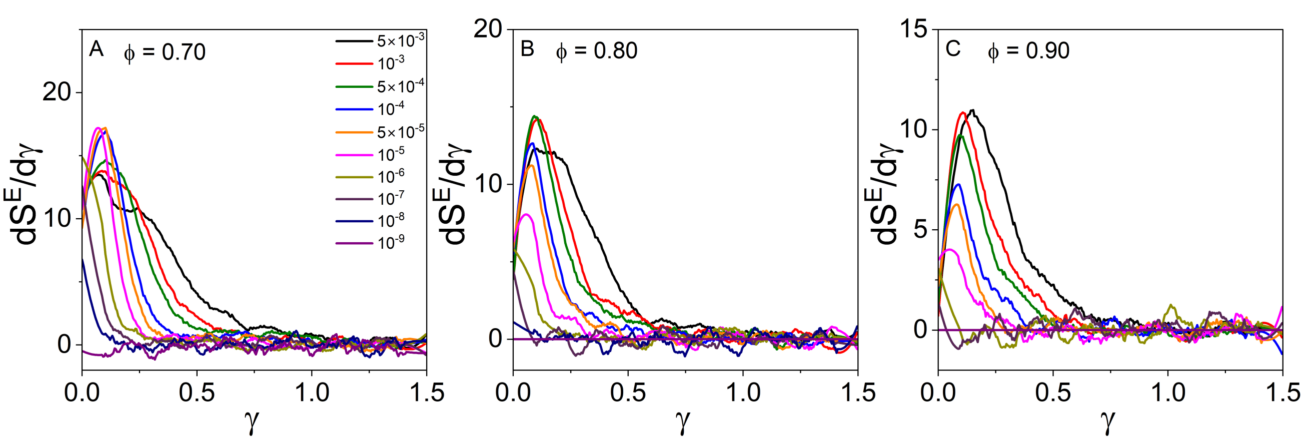


Fig. S8. Derivative of excess entropy with respect to strain, $\left(\frac{\partial S^E}{\partial \gamma}\right)_V$, as a function of strain, $\gamma$, at different shear rates for suspensions with volume fraction of (A) $\phi = 0.70$, (B) $\phi = 0.80$, and (C) $\phi = 0.90$. The color-coding in all sub-figures is the same as (A).

**Correlation of excess entropy with first and second normal stresses as well as osmotic pressure**

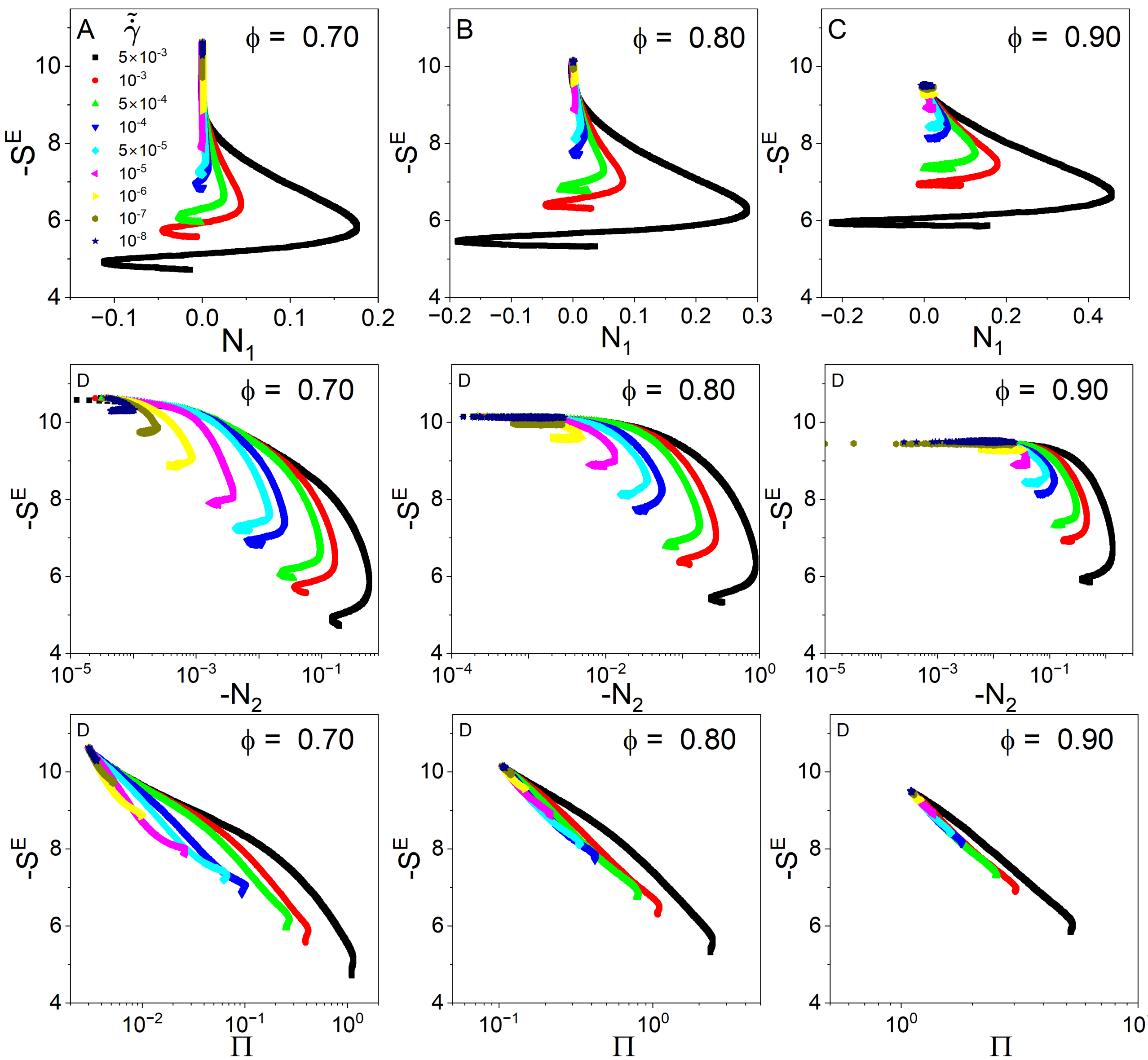


Fig. S9. Excess entropy, $-S_E$, as a function of first normal stress, $N_1$, (top row), second normal stress, $-N_2$, (middle row), and osmotic pressure, $\Pi$, (bottom row) for different volume fractions.


[1] J. R. Seth, L. Mohan, C. Locatelli-Champagne, M. Cloitre and R. T. Bonnecaze, *Nat. Mater.*, 2011, **10**, 838–843.

[2] J. F. Morris and B. Katyal, *Phys. Fluids*, 2002, **14**, 1920–1937.