Supplemental Information for:  Structure-property relationships of sheared carbon black suspensions determined by simultaneous rheological and neutron scattering measurements

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“True” Yield Stress Measurement:

The “true” yield stress of the suspension was measured by performing creep experiments with applied stresses ranging from 70 Pa to 100 Pa. Prior to each applied stress, the suspension was presheared at a shear rate of 2500 s⁻¹ for 600 s. As seen in Fig. S.1., the yield stress of the presheared suspension is between 70 Pa and 90 Pa and depends on the time allowed for the yielding event to occur.
FIG. S.1. Yield stress measurement for a suspension of Vulcan XC-72 carbon black in propylene carbonate with $\phi_{\text{eff}} = 0.274$ after preshearing at 2500 $s^{-1}$ for 600 s. Displacement angle plotted against time for a range of applied stresses. The yield stress range measured is between 70 Pa and 90 Pa.

**Determining the Wall Thickness of the Rheo-SANS Couette:**

Spatiotemporally resolved Rheo-SANS experiments were performed using a hollow titanium Couette geometry. To determine the thickness of titanium along the height of the Couette, a finely-spaced height scan was performed with no sample in the geometry. The results are shown in Fig. S.2 as the transmission measured at each height in the Couette, $T_{\text{Couette}}$, normalized to the transmission of the open beam with no Couette geometry, $T_{\text{open beam}}$, plotted against the normalized height of the Couette, $h/h_0$. For our spatiotemporally resolved experiments, a region of constant thickness was chosen. The boundaries of this region are highlighted by the dashed lines at $h/h_0 = 0.25$ and $h/h_0 = 0.75$. Outside of this region, the thickness of titanium increases, as indicated by a decrease in the normalized transmission.
FIG. S.2. Transmission of the Couette used in spatiotemporally resolved Rheo-SANS experiments measured at different heights along the Couette as $h/h_0$. Dashed vertical lines show maximum and minimum heights measured.

**Rheo-Electric Measurements:**

Typical data from rheo-electric measurements taken under quasi-steady conditions are shown in Fig. S.3 as conductivity, $\kappa$, in part (a) and permittivity, $\varepsilon'$, in part (b). The conductivity was evaluated at $f = 20$ kHz, as indicated by the vertical dashed lines in both plots. Both the conductivity and permittivity measured at 2500 s$^{-1}$ are roughly an order of magnitude higher than that measured at 250 s$^{-1}$. 
FIG S.3. Results from rheo-electric measurements performed at 250 s$^{-1}$ (red triangles) and 2500 s$^{-1}$ (black circles) on a suspension of Vulcan XC-72 carbon black in propylene carbonate with $\phi_{eff} = 0.274$. (a) Conductivity, $\kappa$, plotted against frequency, (b) permittivity, $\varepsilon'$, plotted against frequency. The vertical dashed line marks a frequency of $f = 20$ kHz.

Rheo-uSANS Fitting Details:

Data from Rheo-uSANS measurements performed at $Bi^{-1} > 1$ were fit to a single level Beaucage model [1], [2] to obtain a radius of gyration, $R_g$. To fit the slit smeared data, the Beaucage model was slit smeared using a vertical $q$-resolution of 0.117 Å$^{-1}$ [3]. Fits to the data are shown in Fig. S.4 and are scaled for clarity. Fit information is shown in Table S.I. In the Beaucage model, Guinier’s law is unified with a structurally limited power law such that both a radius of gyration and power law slope can be determined. Two prefactors are used in this model where $G$ is the Guinier scaling factor and $B$ is a prefactor that is specific to the type of power-law scattering measured. Due to the complexity of this suspension, the relationship between these prefactors and specific properties of the suspension is difficult to interpret.
FIG. S.4. Results from a Rheo-uSANS measurement performed at $Bi^{-1} > 1$ for a suspension of Vulcan XC-72 carbon black in propylene carbonate with $\phi_{eff} = 0.274$. Data are slit-smereared and scaled on the y-axis as noted. Fits to a Beaucage model are slit smeared and are shown as solid black lines with fit parameters shown in Table S.I.

TABLE S.I. Parameters from fitting Rheo-uSANS results shown in Fig. S.3 to a Beaucage model. Error reported is one standard deviation.

<table>
<thead>
<tr>
<th>Shear Rate ($s^{-1}$)</th>
<th>$R_g$ (nm)</th>
<th>Power</th>
<th>$B$ (1/cm)</th>
<th>$G \times 10^7$ (1/cm)</th>
<th>$\chi^2$</th>
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<tbody>
<tr>
<td>5000</td>
<td>1310 ± 36.4</td>
<td>1.93 ± 0.0225</td>
<td>0.101 ± 0.0155</td>
<td>1.03 ± 0.0421</td>
<td>0.673</td>
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<tr>
<td>4000</td>
<td>1310 ± 40.2</td>
<td>1.95 ± 0.0211</td>
<td>0.0919 ± 0.0133</td>
<td>1.03 ± 0.0443</td>
<td>1.04</td>
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<tr>
<td>2500</td>
<td>1330 ± 39.8</td>
<td>1.97 ± 0.0204</td>
<td>0.0794 ± 0.0112</td>
<td>1.22 ± 0.0509</td>
<td>1.09</td>
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<tr>
<td>1500</td>
<td>1680 ± 39.8</td>
<td>1.95 ± 0.0178</td>
<td>0.0889 ± 0.0110</td>
<td>2.04 ± 0.0805</td>
<td>1.48</td>
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<tr>
<td>1000</td>
<td>1910 ± 42.3</td>
<td>1.99 ± 0.0158</td>
<td>0.0665 ± 0.00745</td>
<td>3.01 ± 0.0118</td>
<td>1.20</td>
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<tr>
<td>500</td>
<td>2270 ± 37.3</td>
<td>2.02 ± 0.0147</td>
<td>0.0525 ± 0.00555</td>
<td>5.89 ± 0.0203</td>
<td>2.39</td>
</tr>
</tbody>
</table>

Accumulated Strain
**Fig. S.5.** Results from a Rheo-uSANS measurement performed for a suspension of Vulcan XC-72 carbon black in propylene carbonate with $\phi_{eff} = 0.274$. Data are plotted as measured viscosity against the accumulated strain for (a) $Bi^{-1} > 1$ and (b) $Bi^{-1} < 1$.

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**References**


Notes

Disclosure: Certain commercial equipment, instruments, or materials are identified in this paper to specify the experimental procedure adequately. Such identification is not intended to imply recommendation or endorsement by the National Institute of Standards and Technology, nor is it intended to imply that the materials or equipment identified are necessarily the best available for the purpose.