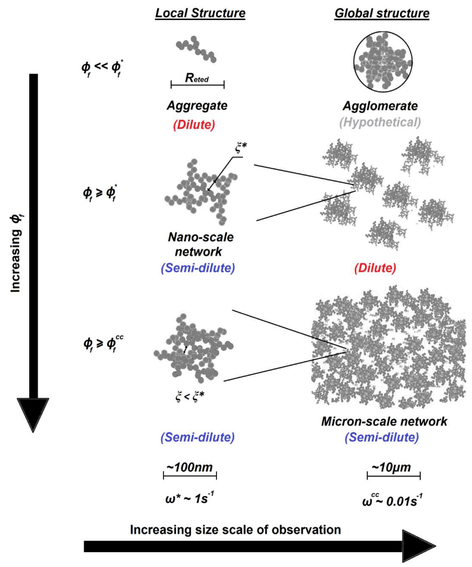
Impact of an Emergent Hierarchical Filler Network on Nanocomposite Dynamics[1]

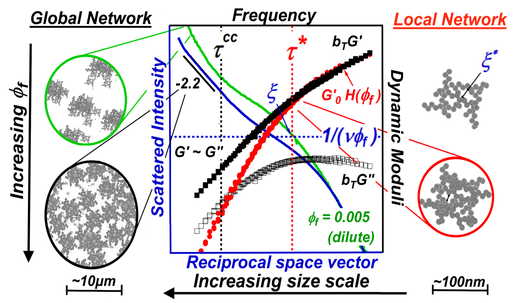
Kabir Rishi[a], Gregory Beaucage[a], Vikram Kuppa[b], Andrew Mulderig[a], Vishak Narayanan[a], Alex McGlasson[a], Jan Ilavsky[c], Mindaugas Rackaitis[d]

[a] Department of Materials Science, University of Cincinnati, Cincinnati, OH 45221, USA
[b] Nonstructural Materials Division, University of Dayton Research Institute, Dayton, OH 45469 USA
[c] Advanced Photon Source, Argonne National Laboratory, Argonne, IL, 60439 USA
[d] Bridgestone Americas Center for Research and Technology, Akron OH 44301, United States

Model[1]



Overview



Methods

- Commercial PBD (M_w ~220 kg/mol) milled with 6PPD (antioxidant) and varying amount of Vulcan 8 carbon black (reinforcing filler) for 6 mins at 130 °C and 60 rpm.
- Scattering from ~1.2 mm (thk.) flat samples measured at Advanced Photon Source, Argonne National Laboratory using the ultra-small-angle X-ray scattering (USAXS) facility located at the 9 ID beam line, station C.
- Dynamic response from 20 mm (dia.) x ~3 mm (thk.) disks measured on a Discovery HR-2 rheometer with a parallel plate geometry. Fixed oscillatory strain of 0.1%.
- Micrographs obtained through TEM in STEM mode from ~80 nm thin sections cooled below T_g of the nanocomposites

Details

Small angle X-ray scattering and TEM

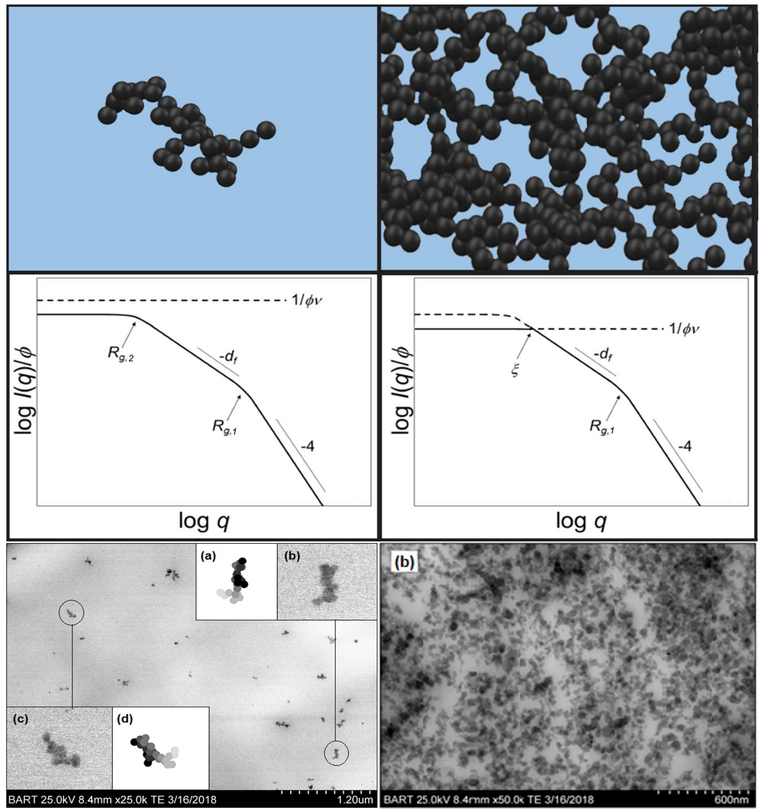
- Structural parameters in the dilute regime computed from the Unified fit.[2,3]

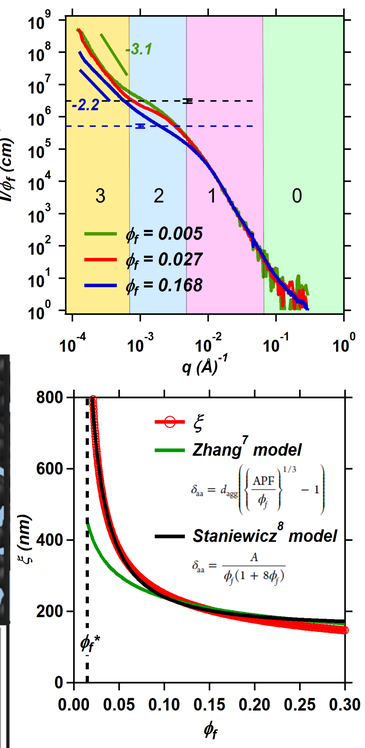
$$I_0(q) = \sum_0^3 \left\{ G_i \exp\left(\frac{-q^2 R_{g,i}^2}{3}\right) + \exp\left(\frac{-q^2 R_{g,i+1}^2}{3}\right) B_i (q_i^{*})^{-P_i} \right\} \quad \text{where,} \quad q_i^{*} = \frac{q}{\left(\operatorname{erf}\left\{\frac{kqR_{g,i}}{\sqrt{6}}\right\}\right)^3}$$

- In semi-dilute systems, structural features are screened and approximated by RPA.[4]

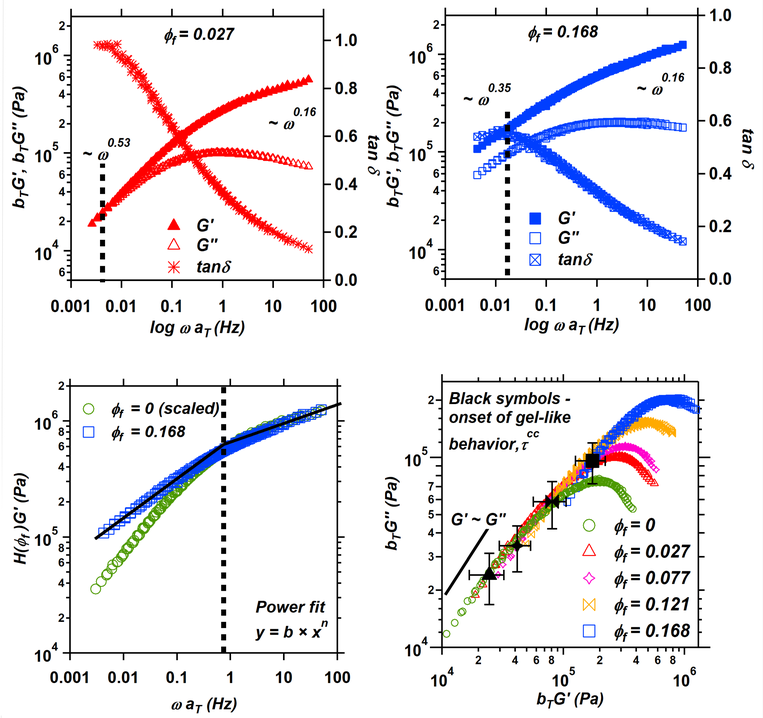
$$\left\{\frac{I(q,\phi_f)}{\phi_f}\right\}^{-1} = \left\{\frac{I(q,\phi_{f,0})}{\phi_{f,0}}\right\}^{-1} + \nu\phi_f$$

- The size of the emergent aggregated filler network is the mesh size, $\xi = 2\pi/q$.[5,6]





Dynamic viscoelastic response



Conclusion

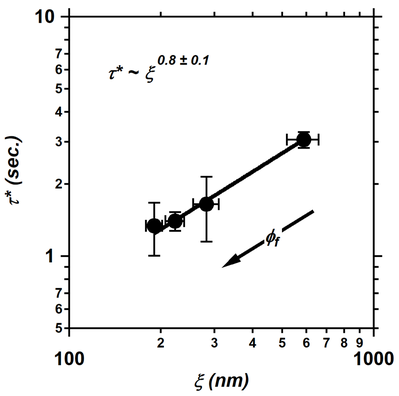
Local nano-scale network

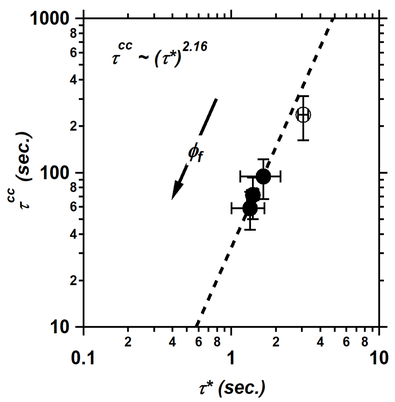
$$\xi \sim (\tau^*)^{1/\alpha} \overset{9}{\sim} (\tau^*)^{d_s}$$

$$c = d_f/d_{\min} \overset{10}{=} d_s$$

Global micron-scale network

$$\tau^{cc} \sim (\tau^*)^{d_f = 2.2}$$





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For further information, please contact:
Gregory Beaucage beaucag@ucmail.uc.edu
Kabir Rishi rishikr@mail.uc.edu