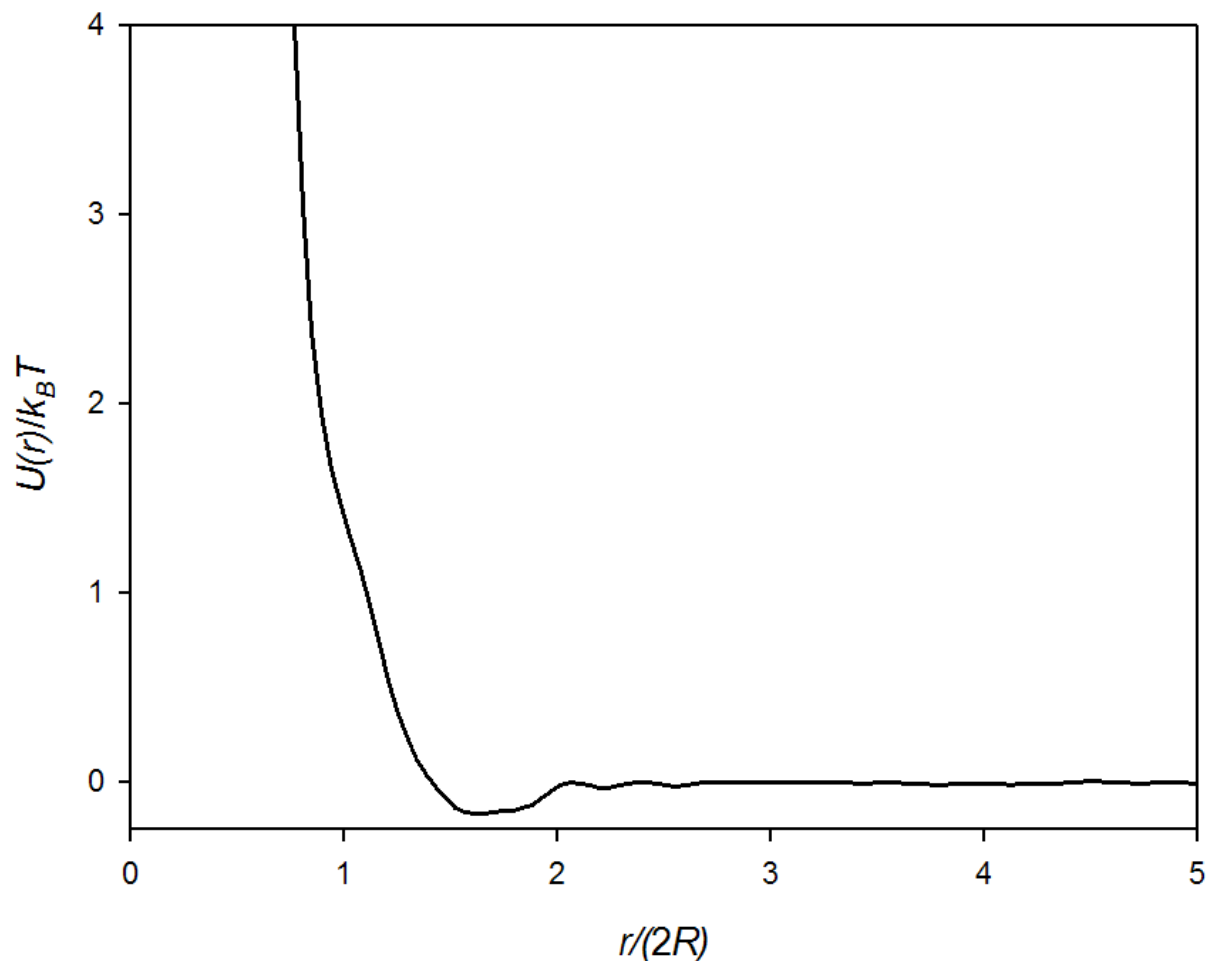## Supplementary Materials

Figure A.1. Interaction potential, $U(r)$, extracted from USAXS data using Percus-Yevick closure for silica nanoparticles of radius $R \approx 100$ nm grafted with 25 kg/mol PDMS in 13 kg/mol PDMS at a particle core volume of $\phi_c = 0.18$. The presence of an attractive well and divergence of the potential at $r/(2R) < 1$ suggest softening effects due to polydispersity. The effects of polydispersity on the extraction of interaction potentials and the treatment of polydispersity in prediction of mechanical properties are included in References 26 – 30.

The maximum magnitude of attraction due to van der Waals (vdW) interactions for 109 nm silica spheres in a PDMS melt is 0.01 k_BT, which occurs at larger separation distances due to the stretching of graft polymers. Thus, we do not plot the VdW interaction in Fig. A.1 as it is

much smaller than the well depth in Fig. A.1. This agrees with the expectation that for repulsive

spheres, vdW forces are negligible. To calculate the potential energy of attraction due to vdW

forces, the standard equation for two spheres of equal radius was used [1]:

$$U_A = -\frac{A}{6}\left[\frac{2R^2}{f_1(R,r)} + \frac{2R^2}{f_2(R,r)} + \ln\left(\frac{f_1(R,r)}{f_2(R,r)}\right)\right]$$ (A.1)

where A is the Hamaker constant, r is interparticle separation distance, $f_1(R,r) = r^2 + 4Rr$ and

$f_2(R,r) = r^2 + 4Rr + 4R^2$.
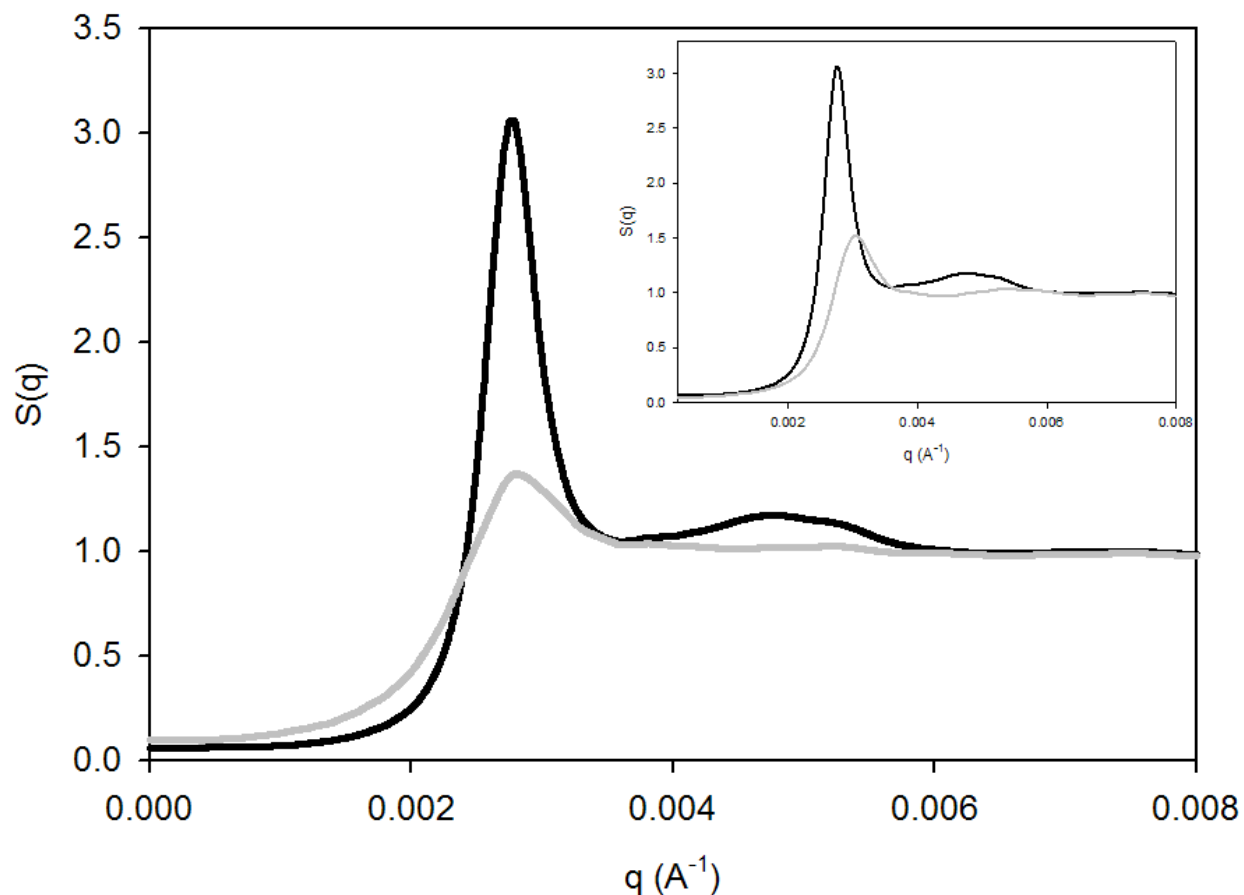
Figure A.2. Structure factor $S(q)$ for silica nanoparticles of radius $R \approx 100$ nm grafted with 25 kg/mol PDMS in 2kg/mol (black) and 13kg/mol (gray) PDMS at a volume fraction of $\phi_c = 0.27$. Inset shows $S(q)$ of the particles at ϕ_c^*, the concentration at which the grafted polymer brushes impinge, i.e. the overlap concentration,[2] for $\phi_{c(2k)}^* = 0.27$ and $\phi_{c(13k)}^* = 0.37$ which corresponds to an effective volume fraction, $\phi_{eff} \approx 0.60$ Despite a lower concentration of scatterers, the magnitude of the structure factor in the lower molecular weight melt greatly exceeds that in the higher molecular weight melt. This effect corresponds to the mechanical reinforcement observed in the lower molecular weight melt due to the stretching of the polymer brush.

Reference:

1.   Hiemenz, P. C.; Rajagopalan, R., *Principles of Colloid and Surface Chemistry*. Third Edition ed.; Taylor and Francis Group, LLC: Boca Raton, 1997.

2.   McEwan, M.; Green, D., Rheological Impacts of Particle Softness on Wetted Polymer-Grafted Silica Nanoparticles in Polymer Melts. *Soft Matter* **2009,** 5, (8), 1705-1716.