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Effect of Dynamically Heterogeneous Interphases on Particle Dynamics of Polymer Nanocomposites

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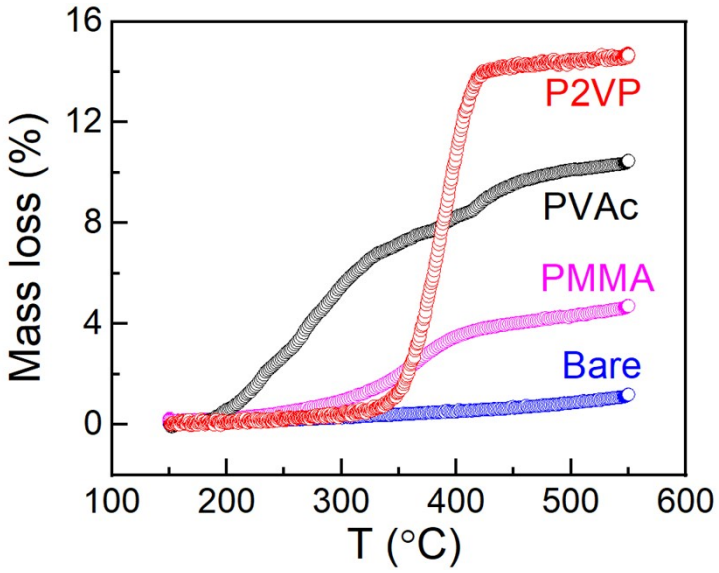

Figure S1. TGA data of polymer adsorbed SiO_2 nanoparticles.

The chain densities were calculated by $\sigma = a\frac{N_A \rho r}{3M}$, where σ is the chain density (chains/nm^2), a is the mass ratio between polymer and particle from the TGA result, N_A is the Avogadro constant, ρ is the density of the particle (SiO_2, 2.65 g/cm^3), r is the mean radius of the particle, ~6.25 nm, M is the molecular weight of the polymer.

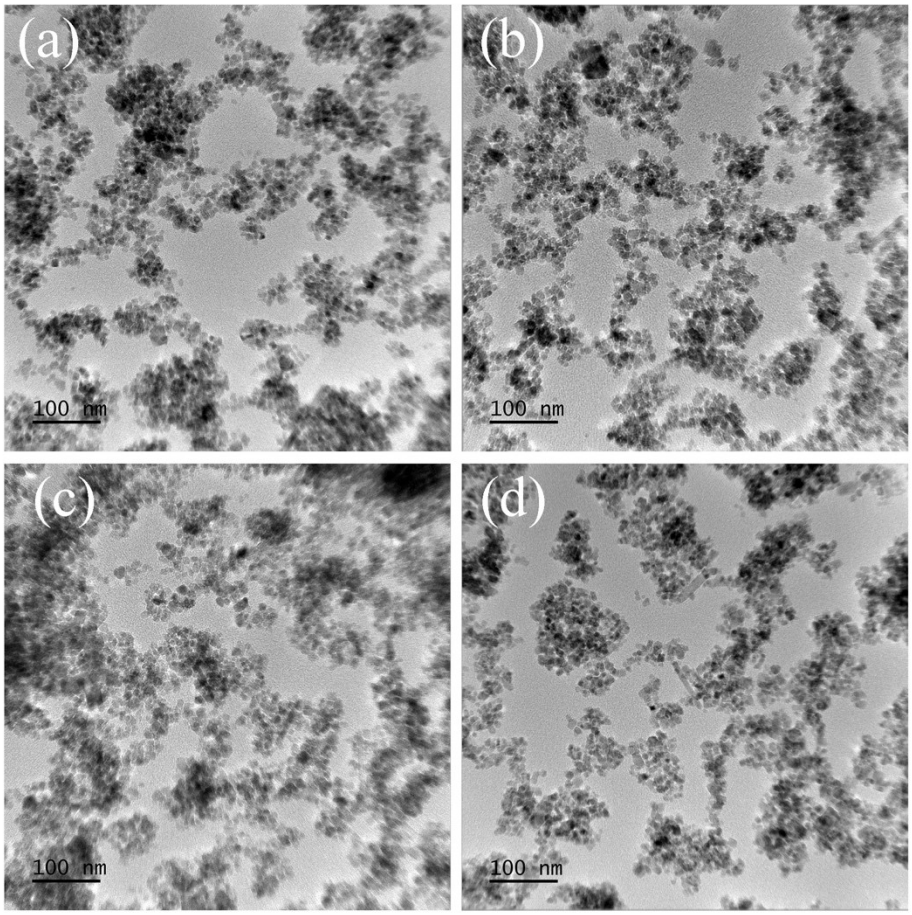

Figure S2. TEM images of cryomicrotomed PMA nanocomposites. Data show aggregation states of Fe_3O_4 nanoparticles adsorbed with (a) 35 kDa PMMA, (b) 50 kDa PVAc, and (c) 40 kDa P2VP. (d) PMA nanocomposite with bare Fe_3O_4 nanoparticles. All samples are at 10 vol% loading[1]. Adapted from *J. Appl. Phys.* 2021, *130* (6), 064701 with the permission of AIP Publishing.

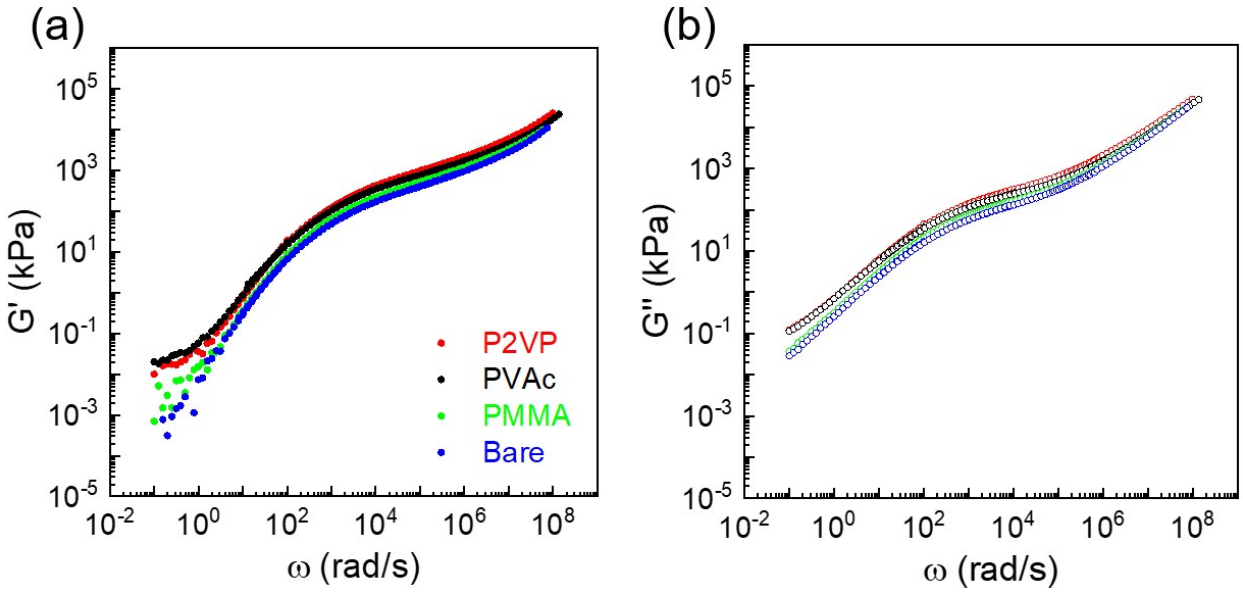

Figure S3. TTS data of polymer adsorbed SiO_2 composites. **(a)** Storage moduli and **(b)** loss moduli of composites. The reference temperature is 140 °C.

SAXS data analysis of PMA-SiO_2 Composites:

The SAXS data is analyzed by fitting the Unified model[2]:

$$I(q) = \sum_{i=1}^{n}\left[G_i exp\left(-\frac{q^2 R_{gi}^2}{3}\right) + B_i exp\left(-\frac{q^2 R_{g(i+1)}^2}{3}\right)\left\{\left[erf\left(\frac{qR_{g,i}}{\sqrt{6}}\right)\right]^3 / q\right\}^{P_i}\right]$$

G_i is the Guinier prefactor, B_i is the prefactor specific to the type of power-law scattering, R_{gi} is the structural size, and p_i is the power-law exponent that determines the shape of fractals, n represents the maximum number of structural levels for fitting.

Table S1. The number of levels for unified function and corresponding R_g (nm) are listed. The

ratio between G_i represents the population of different R_{gi}'s.

Sample	n	R_{g1}	R_{g2}	R_{g3}	R_{g4}	R_{g5}	G_1:G_2:G_3:G_4:G_5
bare	1	7.2±0.2	-	-	-	-	-
PMMA	2	10.8±1	123±10	-	-	-	1:30
PVAc	3	12.4±2	39.5±5	87.5±5	-	-	1:13:41
P2VP	5	9.5±0.5	26.8±2	77.5±4	165±7	176±5	1:12:213:894:342

References

1. Wu, D.; Feng, Y.; Li, R.; Ozisik, R.; Akcora, P., Entanglement density and particle dynamics in rigid interfacial layers of polymer nanocomposites. *J. Appl. Phys.* **2021,** *130* (6), 064701.
2. Beaucage, G., Approximations Leading to a Unified Exponential/Power-Law Approach to Small-Angle Scattering. *J. Appl. Crystallogr.* **1995,** *28* (6), 717-728.