

SUPPLEMENTARY INFORMATION

Surface Stresses and a Force Balance at a Contact Line

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SI1. Simulation Details

Model and Interactions: Coarse-grained molecular dynamics simulations were performed to study interactions of polymeric droplets, solid spherical and cylindrical nanoparticles with elastic (gel-like) substrates (see **Figure 3**).^{1,2} Polymeric droplets consisted of bead-spring chains each having $N = 32$ beads (monomers) with diameter σ connected by the FENE bonds. Nanoparticles are modeled by assembly of Lennard-Jones beads of diameter σ arranged in a hexagonal closed-packed (HCP) lattice and connected to their nearest neighbors by the FENE bonds. Elastic substrates were made by randomly cross-linking bead-spring chains with the degree of polymerization $N = 32$. We have varied the number of cross-links per chain to cover a wide range of gel shear modulus.^{1,2}

The interactions between all beads in a system were modeled by the truncated-shifted Lennard-Jones (LJ) potential³

$$U_{\text{LJ}}(r_{ij}) = \begin{cases} 4\epsilon_{\text{LJ}} \left[\left(\frac{\sigma}{r_{ij}} \right)^{12} - \left(\frac{\sigma}{r_{ij}} \right)^6 - \left(\frac{\sigma}{r_{\text{cut}}} \right)^{12} + \left(\frac{\sigma}{r_{\text{cut}}} \right)^6 \right] & r_{ij} \leq r_{\text{cut}} \\ 0 & r_{ij} > r_{\text{cut}} \end{cases} \quad (\text{SI.1})$$

where r_{ij} is the distance between the i th and j th beads and σ is the bead diameter. In the case of a droplet on elastic substrate the Lennard-Jones interaction parameter ϵ_{LJ} was equal to $1.5k_{\text{B}}T$ for interactions between beads of the same type, and was equal to 0.40, 0.75 or $1.20 k_{\text{B}}T$ for interactions between beads of different types (where k_{B} is the Boltzmann constant and T is the absolute temperature). The cutoff distance r_{cut} is set to 2.5σ . Interaction parameters used in simulations of nanoparticle systems is set to $1.5 k_{\text{B}}T$ for all pairs with $r_{\text{cut}} = 2.5\sigma$

The connectivity of the beads into polymer chains, the cross-link bonds and bonds belonging to beads forming nanoparticles were modeled by the finite extension nonlinear elastic (FENE) potential⁴

$$U_{\text{FENE}}(r) = -\frac{1}{2} k_{\text{spring}} R_{\text{max}}^2 \ln \left(1 - \frac{r^2}{R_{\text{max}}^2} \right) \quad (\text{SI.2})$$

with the spring constant $k_{\text{spring}} = 30k_B T/\sigma^2$ and the maximum bond length $R_{\text{max}} = 1.5\sigma$. The repulsive part of the bond potential is modeled by the LJ-potential with $r_{\text{cut}} = 2^{1/6}\sigma$ and $\epsilon_L = 1.5k_B T$.

Simulations were carried out in a constant number of particles and temperature ensemble. The constant temperature was maintained by coupling the system to a Langevin thermostat implemented in LAMMPS.⁵ The motion of the i th bead is described by the following equation

$$m \frac{d\vec{v}_i(t)}{dt} = \vec{F}_i(t) - \zeta \vec{v}_i(t) + \vec{F}_i^R(t) \quad (\text{SI.3})$$

where m is the bead mass set to unity for all particles in a system, $\vec{v}_i(t)$ is the bead velocity, and $\vec{F}_i(t)$ denoted the net deterministic force acting on the i th bead. The stochastic force $\vec{F}_i^R(t)$ had a zero average value and δ -functional correlations $\langle \vec{F}_i^R(t) \vec{F}_i^R(t') \rangle = 6k_B T \zeta \delta(t - t')$. The friction coefficient ζ was set to $\zeta = m/\tau_{\text{LJ}}$, where τ_{LJ} is the standard LJ-time $\tau_{\text{LJ}} = \sigma(m/\epsilon)^{1/2}$ with value of $\epsilon = 1.0 k_B T$. The velocity-Verlet algorithm with a time step $\Delta t = 0.01\tau_{\text{LJ}}$ was used for integration of the equation of motion. The system was periodic in x and y directions with system dimensions listed in **Tables SI1a** and **SI1b**. All simulations were performed using LAMMPS.

Elastic Substrates: The elastic (gel-like) substrates were made by confining polymer chains into a slab with thickness H_0 then the chains were randomly cross-linked by the FENE bonds (see for details^{1,2}). After cross-linking, the confining slab was removed and substrates were equilibrated in the external potential

$$U_w(z) = \epsilon_w \left[\frac{2}{15} \left(\frac{\sigma}{z} \right)^9 - \left(\frac{\sigma}{z} \right)^3 \right] \quad (\text{SI.4})$$

where ϵ_w was set to $1.0 k_B T$. Substrates were equilibrated by performing MD simulation runs lasting $10^4 \tau_{\text{LJ}}$. The equilibrium substrate thickness was varied between 20σ and 64σ depending on the droplet and particle size. The shear modulus of the substrate was obtained from 3-D simulations of a polymeric network with the same degree of cross-linking and values of interaction parameters as described in ref⁶.

Table SI1a: System Sizes Used in Droplet Simulations

Droplet Radius $R_d [\sigma]$	10.92	15.81	19.78	25.74	30.64	34.62
Substrate Dimensions $L_x = L_y [\sigma]$	45.2	65.6	82.1	106.9	127.3	143.9

Table SI1b: System Sizes Used in Nanoparticle Simulations

Spherical Nanoparticles			Cylindrical Nanoparticles		
$R_p [\sigma]$	$L_x = L_y [\sigma]$	$L_z [\sigma]$	$R_{\text{cyl}} [\sigma]$	$L_x = L_y [\sigma]$	$L_z [\sigma]$
9.8	45.2	65.2	9.6	45.2	65.2

14.3	65.6	85.6	14.8	65.6	85.6
17.9	82.1	102.1	17.8	82.1	102.1
23.3	106.9	126.9	22.9	106.9	126.9
27.7	127.3	147.3	27.3	127.3	147.3
31.4	143.9	163.9	31.4	143.9	163.9

Polymeric Droplets: The polymer droplets were made by confining polymer chains into a spherical cavity (see for details⁶). After removing the confining cavity, droplets were equilibrated by performing MD simulation runs lasting $10^4\tau_{LJ}$. The size of equilibrium droplets R_d was obtained by calculating their radius of gyration.

Nanoparticles: The solid nanoparticles with cylindrical and spherical shapes were generated by arranging beads into HCP lattice and connecting them by elastic bonds with twelve closest neighbors. Nanoparticles were relaxed by performing MD simulation runs lasting $100\tau_{LJ}$. After equilibration the bond length between beads forming nanoparticles is equal to 0.97σ .

Droplet Spreading or Particle Adhesion: Simulations of droplets spreading or nanoparticle adhesion on elastic substrates began by placing a droplet/nanoparticle at a distance of 2.0σ from a substrate. A harmonic potential with the spring constant $K_{sp} = 100k_B T/\sigma^2$ was applied to the droplet center of mass for $100\tau_{LJ}$ to bring it in contact with a soft substrate then the potential was removed. The system was equilibrated for $4 \times 10^4\tau_{LJ}$ followed by the production run lasting $10^4\tau_{LJ}$.

Work of Adhesion: The work of adhesion between droplet and substrate or between particle and substrate was calculated using the potential of the mean force. In these simulations, two films having structures of elastic (gel-like) substrate and polymeric droplet or solid particle with initial dimensions $10\sigma \times 10\sigma \times 10\sigma$ were pushed towards each other. During simulation runs the z -component of the center of mass of the polymeric film or solid substrate was fixed at $z = 0.0\sigma$. The center of mass of elastic film, z_{cm} , was tethered at z^* by a harmonic potential

$$U(z_{cm}, z^*) = \frac{1}{2} K_{sp} (z_{cm} - z^*)^2 \quad (SI.5)$$

with the value of the spring constant K_{sp} varying between 200 and $500 k_B T/\sigma^2$. The location of the film's tethering point was moved with an increment $\Delta z^* = 0.1\sigma$. For each location of the tethering point, a simulation run lasting $5 \times 10^3\tau_{LJ}$ was performed during which distribution of the center of mass of the film was calculated. The weighted histogram analysis method (WHAM)⁷ was applied to calculate potential of the mean force between two films from distribution functions of the film center of mass. The potential of the mean force was used to calculate work of adhesion, $W = \Delta F/A$, as a function of the interaction parameter and the cross-linking density of the elastic substrates.

Table SI2a: Work of Adhesion $W [k_B T/\sigma^2]$ between Droplets and Elastic Substrates

$G [k_B T/\sigma^3]$	$\varepsilon_{LJ} [k_B T]$		
	1.20	0.75	0.40
0.000	2.31	0.95	0.24
0.024	2.32	0.96	0.24
0.072	2.33	0.97	0.25
0.162	2.34	0.98	0.25
0.252	2.36	0.99	0.25
0.498	2.39	1.00	0.26
0.833	2.46	1.03	0.27

Table SI2b: Work of Adhesion $W [k_B T/\sigma^2]$ between Nanoparticles and Elastic Substrates

$G [k_B T/\sigma^3]$	0.024	0.072	0.162	0.252	0.498	0.833
$W [k_B T/\sigma^2]$	4.01	4.14	4.36	4.44	4.49	4.61

Surface Tension: The surface tension of the polymeric droplet and elastic substrates was evaluated by integrating the difference of the normal $P_N(z)$ and tangential $P_T(z)$ to the interface components of the pressure tensor in the system of films having structures of elastic substrate and polymeric melt and with initial dimensions $10\sigma \times 10\sigma \times 10\sigma$. Note that in our simulations, the z direction was normal to the interface.

$$\gamma = \int_{-\xi}^{\xi} (P_N(z) - P_T(z)) dz \quad (\text{SI.6})$$

where 2ξ is the thickness of the interface that was determined from the monomer density profile as an interval within which the monomer density changes from zero to its bulk value. The results of surface tension calculations are presented in **Table SI3**. The surface tension of the substrate/polymer interface was calculated from the work of adhesion, $\gamma_{SL} = \gamma_S + \gamma_L - W$.

Table SI3: Surface Tension

$\gamma_L [k_B T/\sigma^2]$	1.812						
$G [k_B T/\sigma^3]$	0.000	0.024	0.072	0.162	0.252	0.498	0.833
$\gamma_S [k_B T/\sigma^2]$	1.81	1.84	1.88	1.97	2.03	2.21	2.40
$\varepsilon_{LJ} [k_B T]$	$\gamma_{SL} [k_B T/\sigma^2]$						
1.20	1.31	1.34	1.36	1.44	1.48	1.63	1.75
0.75	2.67	2.70	2.72	2.80	2.85	3.03	3.18
0.40	3.39	3.41	3.45	3.53	3.59	3.76	3.94

Table SI4: Droplets on Elastic Substrates

ϵ_{LJ} [$k_{\text{B}}T$]	G [$k_{\text{B}}T/\sigma^3$]	γ_{S} [$k_{\text{B}}T/\sigma^2$]	Υ_{S} [$k_{\text{B}}T/\sigma^2$]	f_{el} [$k_{\text{B}}T/\sigma^2$]	$f_{\text{el,z}}$ [$k_{\text{B}}T/\sigma^2$]	h [σ]	θ_1 [$^\circ$]	θ_2 [$^\circ$]	θ_{f} [$^\circ$]	θ_{s} [$^\circ$]
1.20	0.000	1.81	1.81			0.00	43.0	68.6		
	0.024	1.84	1.91	0.09	0.09	0.87	49.9	58.9	70.1	4.8
	0.072	1.88	2.24	0.34	0.34	2.92	62.9	39.6	71.5	12.6
	0.162	1.97	2.57	0.47	0.47	2.13	67.7	30.0	72.2	14.3
	0.252	2.03	2.83	0.54	0.54	1.94	67.3	26.2	70.3	13.6
	0.498	2.21	3.60	0.73	0.73	1.23	68.9	18.6	76.0	11.5
	0.833	2.40	4.67	0.91	0.91	1.09	70.2	13.2	83.6	9.4
0.75	0.000	1.81	1.81			0.00	91.9	43.6		
	0.024	1.84	2.03	0.13	0.07	0.38	95.9	37.6	29.1	2.6
	0.072	1.88	2.60	0.39	0.34	2.01	101.9	26.6	53.7	6.5
	0.162	1.97	3.16	0.56	0.52	1.89	104.0	21.0	59.7	7.0
	0.252	2.03	3.48	0.64	0.59	1.56	103.6	18.8	59.1	7.3
	0.498	2.21	4.33	0.76	0.73	1.22	106.1	14.3	65.4	6.8
	0.833	2.40	6.05	0.98	0.95	1.05	106.7	10.2	71.7	5.3
0.40	0.000	1.81	1.81			0.00	131.3	24.7		
	0.024	1.84	2.23	0.16	0.11	0.75	133.2	20.2	40.4	1.1
	0.072	1.88	2.77	0.28	0.25	0.91	135.3	16.5	59.3	1.6
	0.162	1.97	3.55	0.43	0.39	1.14	135.6	13.3	63.9	2.0
	0.252	2.03	3.95	0.46	0.43	0.91	137.1	11.6	67.7	2.2
	0.498	2.21	5.29	0.61	0.58	0.89	136.4	9.0	69.4	2.3
	0.833	2.40	6.55	0.71	0.67	0.73	135.1	7.6	69.5	2.0

Table SI5: Solid Particles on Elastic Substrates

G [$k_B T/\sigma^3$]	Cylindrical Nanoparticles			Spherical Nanoparticles		
	R_p [σ]	f_{el} [$k_B T/\sigma^2$]	θ_{el} [$^\circ$]	R_p [σ]	f_{el} [$k_B T/\sigma^2$]	θ_{el} [$^\circ$]
0.072	9.6	1.06	50.9	9.8	0.45	103.4
	14.8	1.12	42.1	14.3	0.56	78.0
	17.8	0.98	32.7	17.9	0.61	60.0
	22.9	1.22	21.1	23.3	0.66	47.5
	27.3	1.20	18.9	27.7	0.66	41.3
	31.4	1.43	13.8	31.4	0.86	28.8
0.162	9.6	1.66	33.2	9.8	1.09	49.7
	14.8	1.71	21.8	14.3	1.29	35.4
	17.8	2.14	15.1	17.9	1.68	26.6
	22.9	2.03	10.8	23.3	1.62	20.0
	27.3	2.31	10.0	27.7	1.71	15.0
	31.4	2.32	6.9	31.4	1.94	11.9
0.252	9.6	2.08	21.3	9.8	1.50	31.7
	14.8	2.53	16.8	14.3	1.83	24.5
	17.8	2.57	10.4	17.9	2.05	18.6
	22.9	2.62	7.5	23.3	2.22	15.2
	27.3	2.71	4.0	27.7	2.19	8.5
	31.4	2.85	3.4	31.4	2.51	7.8
0.498	9.6	2.90	13.3	9.8	2.79	22.0
	14.8	2.87	5.7	14.3	2.66	11.7
	17.8	3.32	5.3	17.9	2.88	11.3
	22.9	3.40	3.5	23.3	2.97	3.9
	27.3	3.52	2.9	27.7	2.87	1.1
	31.4	3.64	2.7	31.4	3.51	9.0
0.833	9.6	3.27	8.3	9.8	3.06	14.8
	14.8	3.54	5.9	14.3	3.39	8.9
	17.8	3.83	5.2	17.9	3.69	13.9
	22.9	3.91	3.1	23.3	3.58	5.8
	27.3	3.99	3.4	27.7	3.61	4.6
	31.4	3.96	2.4	31.4	3.87	8.9

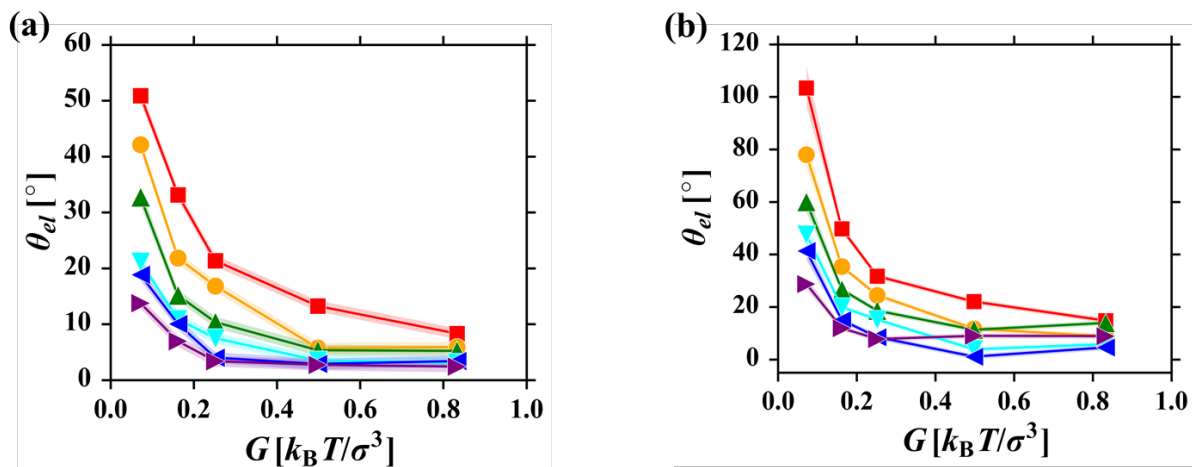


Figure S1: Dependence of the orientation of the elastic force on the substrate shear modulus. The orientation of the elastic force is obtained from analysis of the force balance at the triple phase contact line for (a) cylindrical and (b) spherical nanoparticles. In panel (a) the following symbols are used for particles with size $R_p = 9.8 \sigma$ (red squares), 14.3σ (orange circles), 17.9σ (green triangles), 23.3σ (cyan inverted triangles), 27.7σ (blue left triangles), 31.4σ (purple right triangles). In panel (b) the symbols are $R_p = 9.6 \sigma$ (red squares), 14.8σ (orange circles), 17.8σ (green triangles), 22.9σ (cyan inverted triangles), 27.3σ (blue left triangles), 31.4σ (purple right triangles).

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