Electronic supplementary information

Adsorption of "Soft" Spherical Particles onto Sinusoidally-Corrugated Substrates

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Computer simulation model

We provide details pertaining to the computer simulation setup, initialization, operation and optimization. We chose a 3D lattice Monte Carlo simulation scheme following the bond fluctuation model (BFM) for our system of polymer coated spheres adhesively interacting with sinusoidally-corrugated substrates. The BFM dictates a set of possible bond vectors and prevents overlap of polymer coarse grained elements in a 2D or 3D lattice. Initializing the system involves first the generation of a solid, inelastic substrate defined by y=A*sin(B*x) such that A determines the amplitude of the substrate while B determines the periodicity. A sphere is generated at a set, identical location (X,Y) with fixed height (Z) above the substrate. The sphere is modified with surface bound polymer chains which have optimized equidistant spacing based on the surface grafting density parameter (σ), length defined by chain element parameter (N). The polymer chains are generated orthogonal to the surface; it thus needs time to equilibrate and are given 1000*N Monte Carlo steps to equilibrate in the lattice. Once completed the polymer coated sphere is ready to move in the simulation box. Polymer, particle core and substrate are not allowed to overlap at any lattice site. There is a balance between movement of the entire particle (sphere and polymer together) and independent movement of the coarse-grained polymer chain elements. This balance is struck in optimization by achieving effective maximization of adhesion energy in long Monte Carlo step simulations wherein the particles are able to achieve adhesion energy maxima but maintain ability to release from local energy maxima. The move set probabilities are such that there is N/N+1 chance of attempted polymer chain move and a 1/N+1 chance of attempted entire sphere move. Moves are accepted based on periodic boundary

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conditions in x and y as well as rules of the BFM. This particle moves around in 3D space until it contacts the surface. Contact occurs when a substrate lattice site is adjacent to an adhesive polymer lattice site. Each attractive interaction is pair-wise and is a square-well potential with a value of ε kT and is calculated directly based on the distance between surface lattice sites (X, Y, Z) and polymer lattice sites (x, y, z), such that:

$$r = \sqrt{(X_{substrate} - x_{polymer})^2 + (Y_{substrate} - y_{polymer})^2 + (Z_{substrate} - z_{polymer})^2}$$

$$\begin{cases} r \le \sqrt{10} \rightarrow \varepsilon_{adhesion} = \varepsilon\, kT \\ r > \sqrt{10} \rightarrow \varepsilon_{adhesion} = 0\, kT \end{cases}$$

At this point moves become restricted to acceptance based on increasing adhesion energy of the system or on passing the Metropolis algorithm where the probability of acceptance of the move is proportional to $e^{\Delta E/kT}$, where ΔE is defined as $E_{system,i} - E_{system,i-1}$. This makes large changes to adhesion energy unlikely yet still allows particles to gradually probe the substrate for optimal adhesion locations. An additional move is introduced to the algorithm when the particle is in contact with the substrate. We call this a "stuck" move in which the hard core of the particle attempts to move but some of the surface-bound polymer is adhered to the surface. This move checks the possibility of the core of the particle to move while the adhered polymer remains in contact with the surface. Particles continue to probe the surface for a predefined number of Monte Carlo steps with their X,Y,Z positions, adhesion energy and highest adhesion energy location tracked. The simulation ends after the desired number of trials (typically 1000 trials) have occurred.

Particle settlement as a function of feature width and aspect ratio of the substrates

In Figure S1 we plot the relative penetration depth (RPD, left), topography match parameter (TMP, middle), and adhesion energy (E_{adh}, right) as a function of the aspect ratio (*i.e.*, ratio of feature height to feature width) for D=20, N=3, σ=0.04 and ε=1 kT. The RPD increases with decreasing the aspect ratio and increasing the feature width. The TMP dependence mirrors the behavior of RPD; namely, it increases with increasing the aspect ratio and decreasing the feature width. The adhesion energy exhibits a more complex behavior. Namely, it is relatively low for particles whose sizes match approximately the feature width of the substrates and increases for

all other situations. Settlement of particles on substrates with feature width that is larger than the particle size exhibits, in general, higher adhesion energies with decreasing aspect ratio.

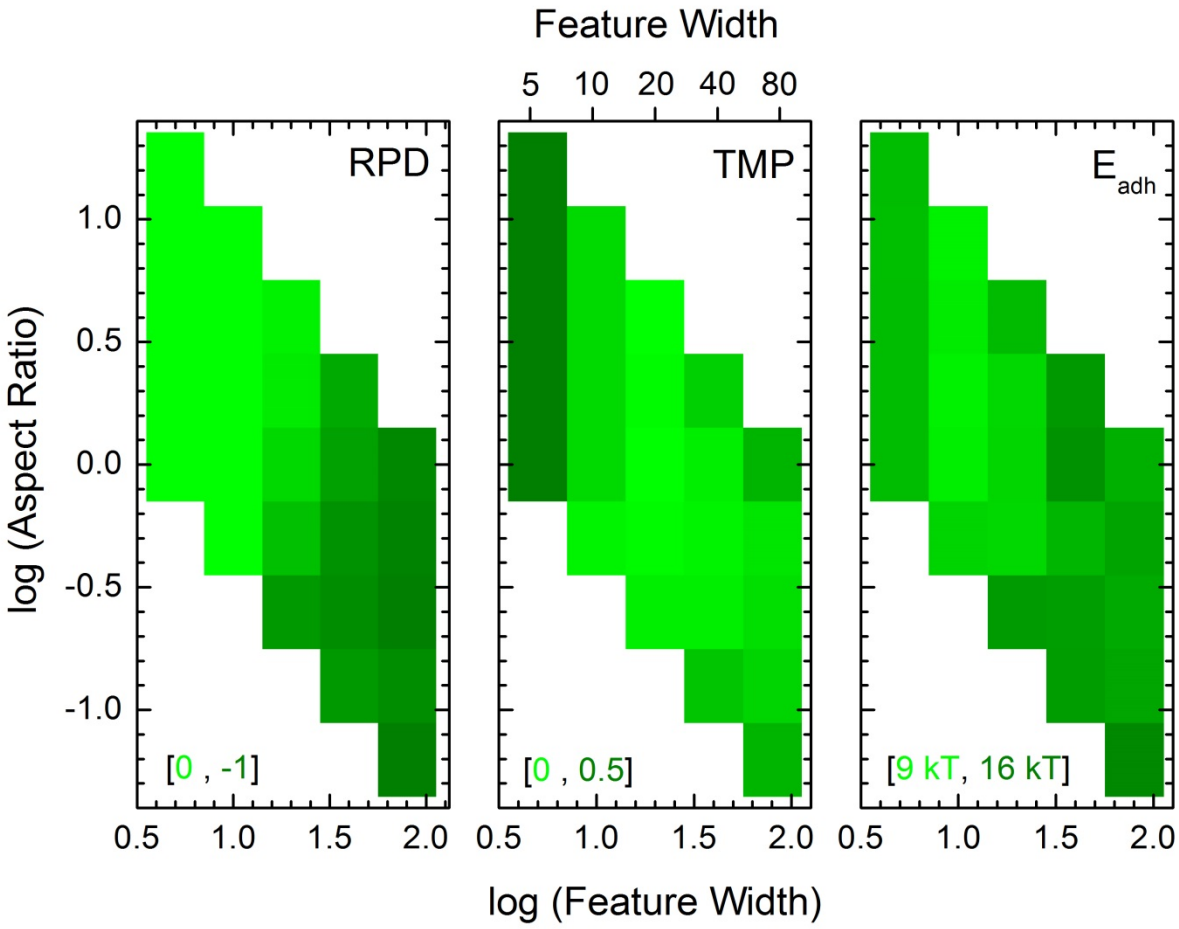

Figure S1. Log(aspect ratio) versus Log(feature width) for adhesion energy of D=20, N=3, σ=0.04 and ε=1 kT among RPD, TMP and adhesion energy measurements. The aspect ratio is defined as a ratio of feature height to feature width. Colored shades of green where lightest represent minimal values and darkest indicates maximal values of the respective measurement. In the regions where both RPD and TMP are light green (minimized) we detect lowest adhesion energies.