

Fabrication of sinusoidal substrates using 3D printing and imprint molding

Substrates with a sinusoidal cross-section were designed using computer-aided design (CAD) software and 3D printed with varying amplitudes and wavelengths using the ProJet 3500 multi-jet (MJP) 3D printer, developed by 3D Systems (nominal layer height resolution of 29 μm). The resin is jetted in layer-by-layer fashion to build up parts, and each layer is cured by exposure to UV light. Parts were printed using the Visijet M3-X resin. The designed parts do not require any support materials. However, a sacrificial layer of the S300 support material is deposited first to allow parts to be separated readily from the build plate. This layer can be removed by heating at 65°C. **Figure S1** shows a representation of the 3D CAD model.

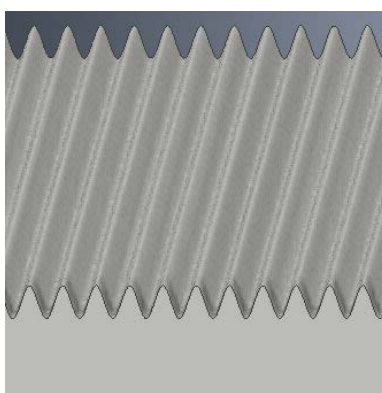


Figure S1. 3D CAD model showing sinusoidal surface topography on a substrate.

Table S1. Dimensions of sinusoidal surface topography.

sample	Wavelength (λ) (μm)	Amplitude (A) (μm)	Aspect ratio (A/ λ)
1	400	275	0.69
2	300	300	1.00
3	400	450	1.13

The dimensions of the substrates fabricated in this manner are listed in **Table S1**. Once substrates are printed, we used imprint molding to create replicas using poly(dimethylsiloxane) (PDMS). We achieved this by pouring a mixture of PDMS base and crosslinker in a 10:1 ratio over the 3D printed mold and curing at 50°C overnight. To de-mold the cured PDMS replica, the mold and cured PDMS were placed in a closed vessel and exposed to toluene vapor for 48 hours. Toluene vapor caused the

PDMS to swell at a slow rate resulting in its separation from the 3D printed mold. PDMS substrate was subsequently heated at 70°C for one hour to remove any residual toluene present in the bulk.

IR spectra of chemical modification of PDMS and LG substrates with PAA

We characterized the layer-by-layer (LBL) deposition of PEI (poly(ethylene imine)) and PAA (poly(acrylic acid)) on ultraviolet/ozone (UVO)-activated PDMS using Fourier Transform Infrared (FTIR) spectroscopy in the attenuated total reflection (ATR) mode and plot them in **Figure S2**. The LBL sample was analyzed after being washed with water to simulate the effect of the particle deposition in water. We used the FTIR spectra of pure PEI and PAA to demonstrate successfully chemical modification. Peaks at 1500-1650 cm^{-1} indicate the presence of amine groups, corresponding to PEI, whereas the peaks at 1750-1800 cm^{-1} show that there are carboxylic acid groups, corresponding to PAA, present on the surface.

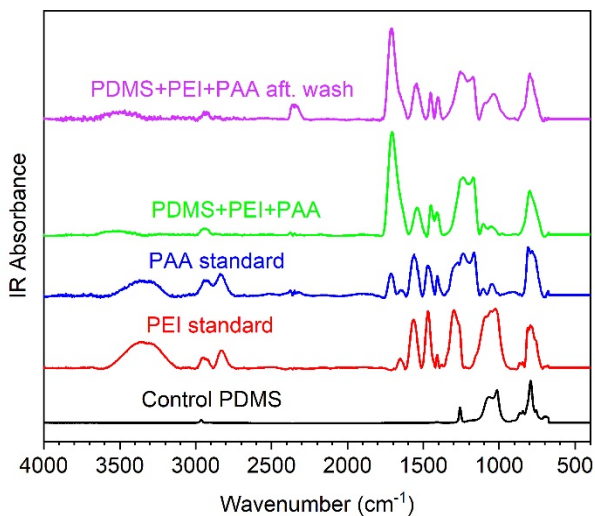


Figure S2. IR spectra showing surface functionalization of PDMS.

The same chemical modification was carried out on PDMS:UVO coated with liquid glass (LG). **Figure S3** shows the resulting FTIR spectra. We compare the FTIR spectra of samples modified with PEI and PAA to pure PEI and PAA to demonstrate the success of surface functionalization.

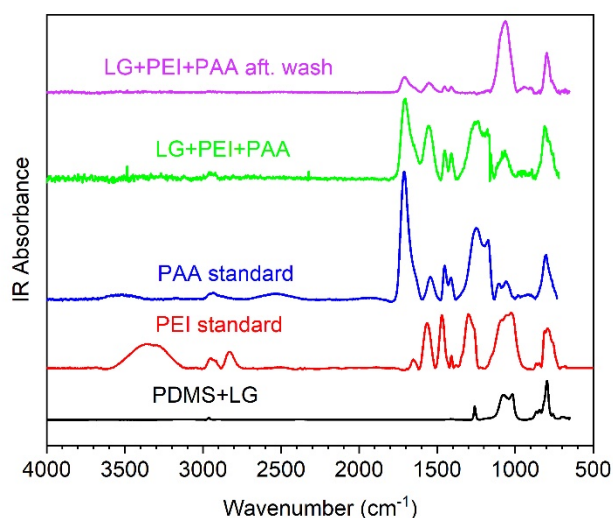


Figure S3. IR spectra showing LBL modification of the LG-coated PDMS surface.

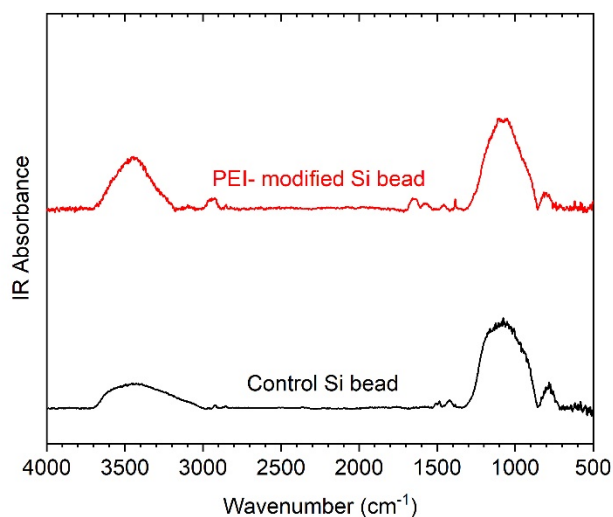


Figure S4. Surface modification of SiO_x particle with PEI.

Figure S4 plots the FTIR spectra of the parent SiO_x particles and those modified with PEI. Particles are pulverized using a mortar and pestle and combined with potassium bromide (KBr). A pellet is created and analyzed using transmission FTIR spectroscopy. In the case of LG surface modification, and SiO_x particle, the signal from the PAA and PEI peaks respectively is relatively weak. This

could indicate uneven or poor coverage by the PEI layer on LG or the particle, or weak bonding to the surface.

Calculation of particle position on sinusoidal surface profile

The surface profile $z=f(x)$ can be modeled as:

$$z = \frac{A}{2} \left[\cos\left(2\pi \frac{x}{\lambda}\right) - 1 \right] . \quad (S1)$$

In Equation (S1), A is the amplitude of the features, λ is the wavelength of the features. We consider particles having a radius R (or diameter D) that are attached to the surface at some point, making a single point contact (cf. **Figure S5**). In 2D representation, the particles are modeled as circles.

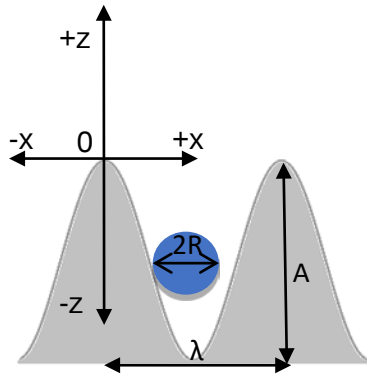


Figure S5. The coordinate system for particle attachment to sinusoidally-corrugated substrates.

The center point of each particle has coordinates $[x', z']$ and we seek to find an analytical way to express the $[x', z']$ coordinates as a function of x and z for a given set of A , λ and R . In **Figure S6** we present such a situation for $A=100$ a.u., $\lambda=100$ a.u. (thus the aspect ratio $AR=A/\lambda=1$) and $R=10$ a.u.

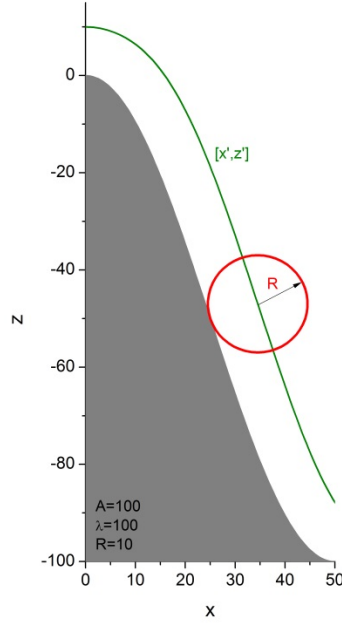


Figure S6. Graphical representation of the coordinates of a particle (*i.e.*, a sphere in the 2D representation) (COS) that is attached to a sinusoidally-corrugated substrate at a single point. The locus of the particle COS is marked with a dark green color.

We first determine the tangent to the surface for each x .

$$\frac{dz}{dx} = -\frac{A\pi}{\lambda} \left[\sin\left(2\pi\frac{x}{\lambda}\right) - 1 \right] \quad (\text{S2})$$

The line perpendicular to the tangent has a slope of:

$$\text{perp} = \left[\frac{A\pi}{\lambda} \left[\sin\left(2\pi\frac{x}{\lambda}\right) - 1 \right] \right]^{-1} \quad (\text{S3})$$

Note that based on geometry the equation for “perp” has to satisfy the condition:

$$\frac{dz}{dx} * \text{perp} = -1 \quad (\text{S4})$$

The angle of the line whose slope=perp (α) is:

$$\alpha = \tan^{-1}(\text{perp}) \quad (\text{S5})$$

The x' and z' coordinates for the circle, having a radius of R , are:

$$x' = x + R * \cos(\alpha) \quad (S6)$$

$$z' = z + R * \sin(\alpha) \quad (S7)$$

Image analysis to obtain particle position in sinusoidal channels

Particle position is recorded by measuring the distance between the particle and the crest of the sinusoid. To do this, we first image the substrates with particles deposited upon them with a laser confocal scanning microscope (LCSM). The instrument used is the Keyence VKx1000. **Figure S7** shows an LCSM image of a particle settling in a sinusoidal channel.

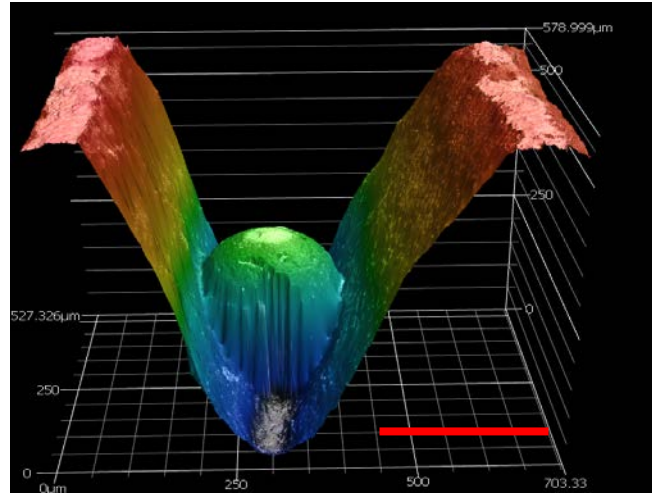


Figure S7. 3D surface map of sinusoidal surface profile and particle. Colors indicate height. Scale bar: 500 μm.

We analyzed images using the Keyence Multifile Analyzer software by taking cross-sections of the system at the particle (perpendicular to the length of the channel), as shown in **Figure S8**.

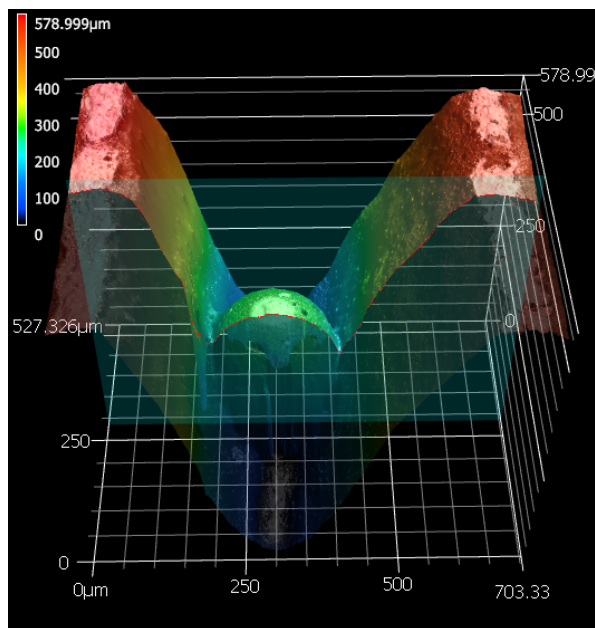


Figure S8. Cross-section of sinusoidal surface profile and particle system. Colors indicate height.

Figure S9 depicts the profile obtained by cross-sectional analysis. This representation enables determining pertinent dimensions, such as substrate periodicity, and the center of the particle relative to the sinusoidal substrate profile.

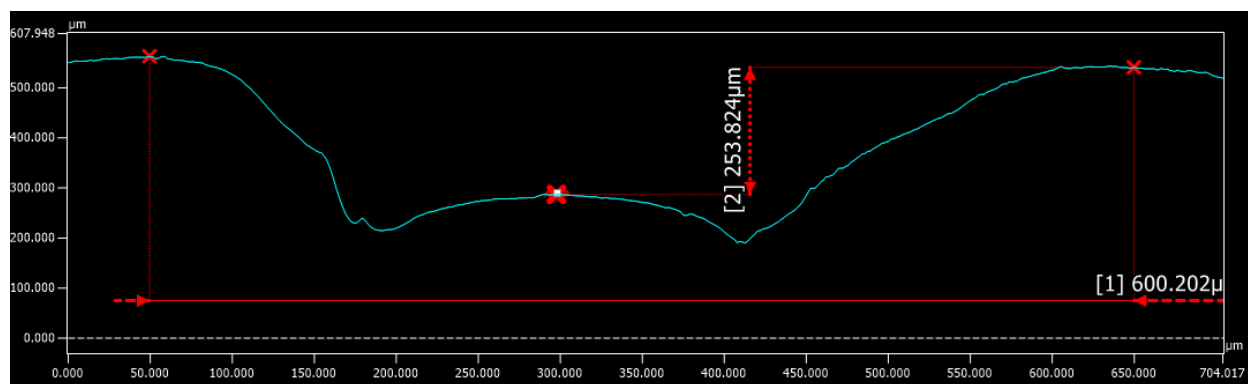


Figure S9. Particle dimensions and position with respect to sinusoidal features are measured from the cross-section.

Surface modulus of PDMS and LG substrates and respective chemical modifications

Surface moduli for each PDMS and LG substrate collected by the CFM are shown in **Figures S10** and **S11**. The images show that surface modification using LG results in a non-uniform coverage of the surface.

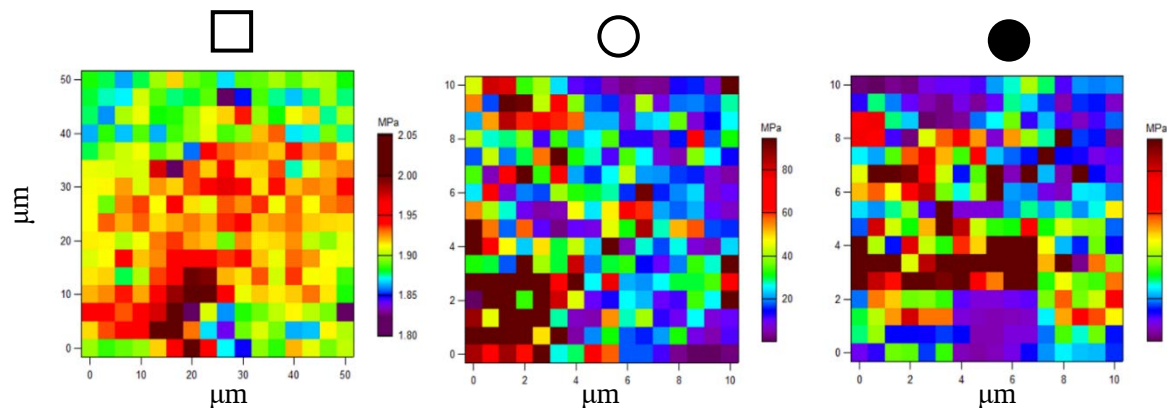


Figure S10. AFM scans showing the measured surface modulus of PDMS as probed by SiO_x particle as a function of chemical modification. Experimental conditions represented by the symbols are as follows: PDMS- SiO_x (□), PDMS:UVO/PAA- SiO_x (○), PDMS:UVO/PAA- SiO_x /PEI (●).

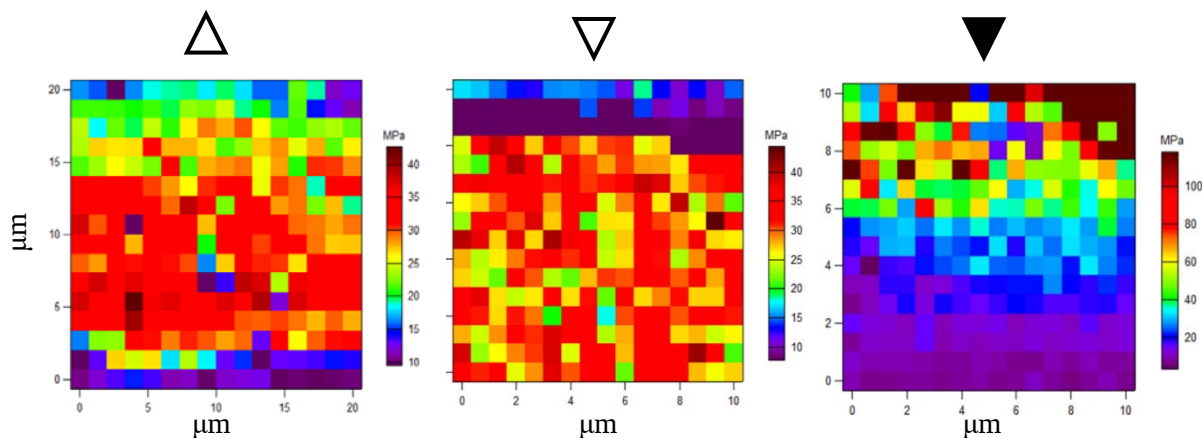


Figure S11. AFM scans showing the measured surface modulus of LG-coated PDMS as probed by SiO_x particle as a function of chemical modification. Experimental conditions represented by the symbols are as follows: PDMS:UVO/PEI/LG- SiO_x (△), PDMS:UVO/PEI/LG/PAA- SiO_x (▽), PDMS:UVO/PEI/LG/PAA- SiO_x /PEI (▼).

The average moduli and work done to separate the two surfaces for each case shown above are calculated and recorded in **Table S2**.

Table S2. Average moduli and work done to separate two surfaces calculated in each case from AFM force mapping.

Substrate	Particle	Modulus (MPa)	Work (pJ)
PDMS	SiO _x	1.6 ± 0.05	2.24 ± 0.038
PDMS:UVO/PAA	SiO _x	28.9 ± 29	0.554 ± 0.595
PDMS:UVO/PAA	SiO _x /PEI	24 ± 24	0.147 ± 0.205
PDMS:UVO/PEI/LG	SiO _x	43 ± 47	0.073 ± 0.030
PDMS:UVO/PEI/LG/PAA	SiO _x	38 ± 22	0.019 ± 0.021
PDMS:UVO/PEI/LG/PAA	SiO _x /PEI	14 ± 39	0.331 ± 0.384

We use the CFM to calculate interactions between the probing particle and substrates. **Figures S12 and S13** show the interactions between surfaces described in **Table 1**. The maps display inhomogeneity in the surface interactions, which could be attributed to the non-uniform chemical coating on both the substrate and the particle.

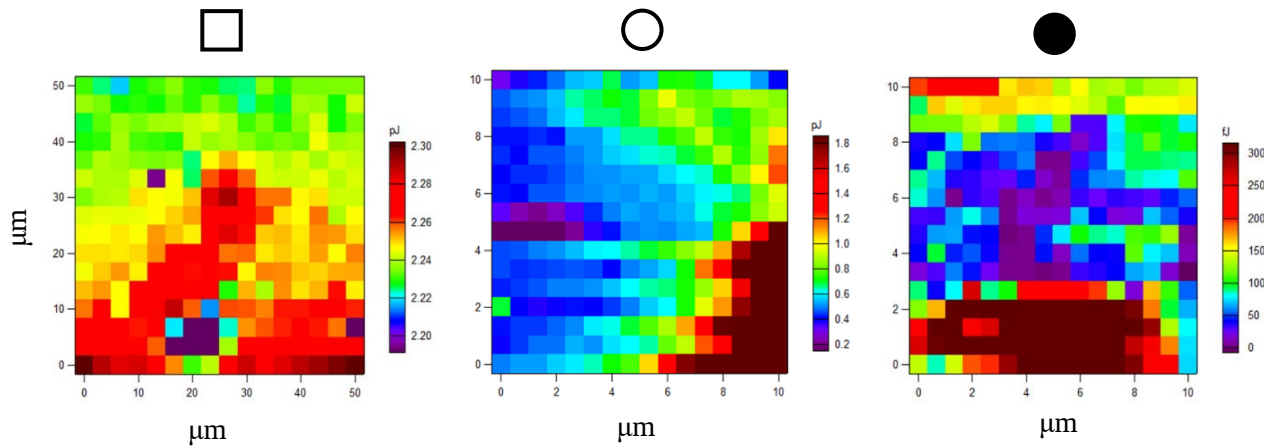


Figure S12. AFM scans showing the calculated work required to separate PDMS substrate from SiO_x particle as a function of chemical modification. Experimental conditions represented by the symbols are as follows: PDMS-SiO_x (□), PDMS:UVO/PAA-SiO_x (○), PDMS:UVO/PAA-SiO_x/PEI (●).

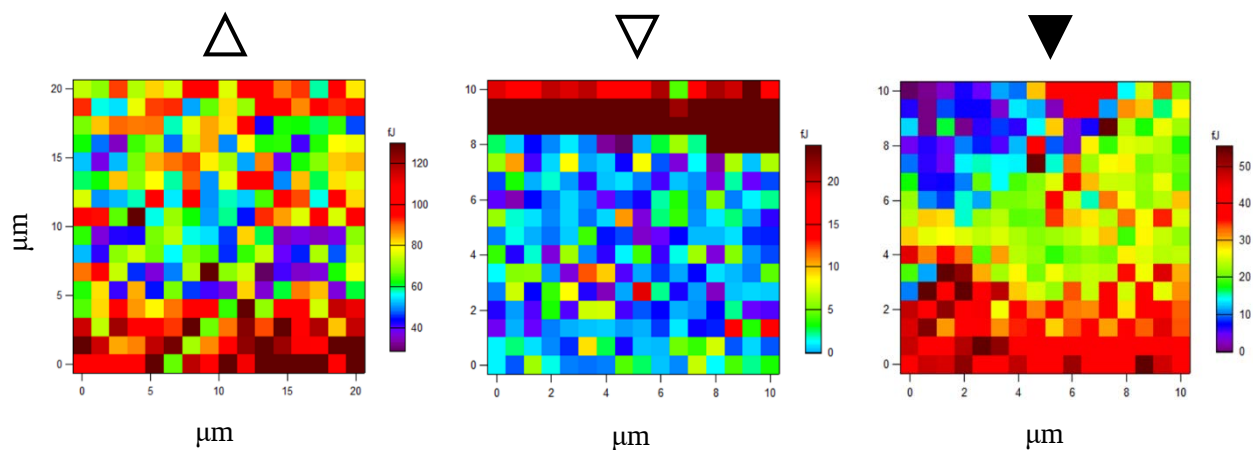


Figure S13. AFM scans showing the calculated work required to separate LG-coated PDMS surface from SiO_x particle as a function of chemical modification. Experimental conditions represented by the symbols are as follows: PDMS:UVO/PEI/LG- SiO_x (Δ), PDMS:UVO/PEI/LG/PAA- SiO_x (∇), PDMS:UVO/PEI/LG/PAA- SiO_x /PEI ($\blacktriangledown$).