Electronic Supplementary Material (ESI) for Soft Matter.

Supplementary Information:

Pressure Tunable Adhesion of Rough Surfaces

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1. Interfacial Contact Videos During Indentation Testing

The evolution of the contact area between the pressure tunable adhesive (PTA) and the flat cylindrical probe increases as the probe is compressed into the substrate. When the direction of the probe is reversed after the maximum compressive load, P_m, is reached, the contact area remains constant until the normal load becomes positive (adhesive). The formation and separation of the interface differs with different droplet heights. SI Videos 1- 3 show the probe (radius, a = 0.5 mm) – PTA interfacial contact formation and separation during compression and retraction of the probe from the substrate, respectively. The vertical displacement of the probe is 1 μm/s and $P_m =$ 100 mN for all videos. SI Video 1 displays an adhesion test performed on the smallest ($h = 0.5\,\mu$m) droplet size. SI Video 2 and SI Video 3 are representative adhesion videos of intermediate ($h = 1.6\,\mu$m) and large ($h = 2.9\,\mu$m) droplet sized PTA surfaces, respectively.

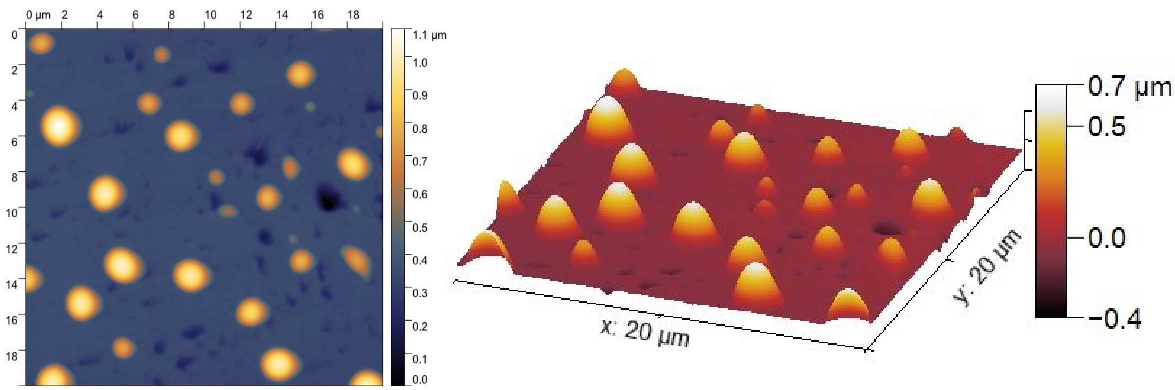
SI Video 1: Contact, compression, and separation of flat cylindrical punch on PTA surface (droplet height $h = 0.5\,\mu$m). The outline of the probe can be clearly seen throughout the test. The probe first advances towards the surface, contact (darkest regions) first occurs around the probe circumference and then the contact moves towards the center as the compressive load increases. Different regions of the bare PDMS between droplets "jump" into contact. At $P_m =$ 100 mN, only the tops of the droplets in the center of the projected contact area are touching the probe. The PDMS in the center does not make contact with the probe (light region) and does not play a significant role in the adhesive response of the surface during this test. As the probe is retracted, this "top contact" region increases by growing radially outward until separation occurs. Time has been accelerated slightly for ease of observation. The timestamp in the lower left corner reflects the lapsed time of the test.

SI Video 2: Contact, compression, and separation of flat cylindrical punch on PTA surface (droplet height $h = 1.6\ \mu$m). See caption for SI Video 1 for all other details.

SI Video 3: Contact, compression, and separation of flat cylindrical punch on PTA surface (droplet height $h = 2.9\ \mu$m). See caption for SI Video 1 for all other details.

2. Atomic Force Microscopy

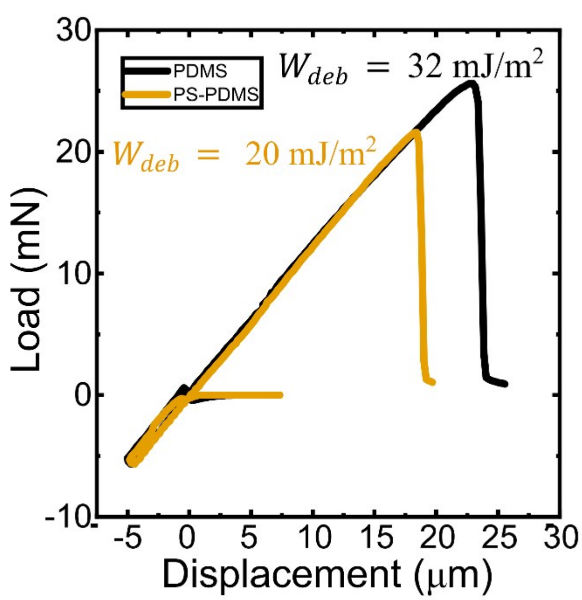
Due to the lateral resolution limits of the optical profilometer (OP) used to characterize droplet diameters, atomic force microscopy (AFM) scans were obtained on the smallest droplet PTA surfaces to confirm the average droplet dimensions. While the OP height measurements were in good agreement with the AFM-determined droplet heights, the droplet diameters were dramatically different than those measured with OP. Height scans were obtained with tapping mode over a scan size of 20 μm by 20 μm. Representative 2D and 3D scan results for the smallest droplet PTA surfaces ($h = 0.5\ \mu$m) are shown in SI Figure 1(a) and (b), respectively.



SI Figure 1: AFM scans of smallest droplet surfaces ($h = 0.5\ \mu$m). (a) 2D height image of a single scan. Color scale on left indicates height. (b) To get a better understanding of the aspect ratio of the droplets, a 3D scan of the same area confirms that the droplets are smooth and round.

3. Adhesion Testing of Smooth Interfaces

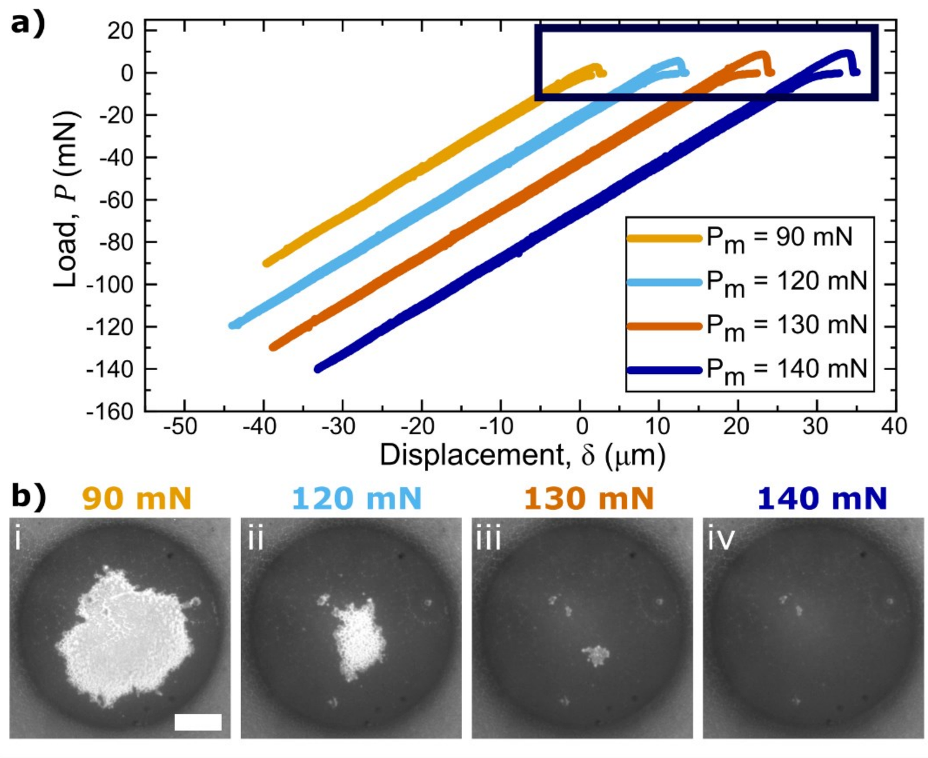
To determine the adhesive properties of smooth interfaces with an identical chemical makeup to the PTA surfaces, control experiments of flat punch indentation testing was performed with a cylindrical alumina silica probe contacting a polydimethyl siloxane (PDMS) surface (SI Figure 2, black line) and a polystyrene (PS)-coated PDMS (thin film ($t =$ 97nm)) bilayer(SI Figure 2, orange line). The load versus displacement curves show that there is a significant adhesive response in both contact scenarios. The work of debonding (W_{deb}) is determined by integrating the tensile portions of these curves and then normalizing by the projected area of the probe. The test parameters were held fixed at a probe displacement rate of 1 μm/s, probe radius of 0.5 mm and $P_m =$ 5 mN. For the PDMS surface, $W_{deb} = 32mJ/m^2$ and for the PS-coated PDMS surface, $W_{deb} = 20mJ/m^2$.



SI Figure 2: Adhesion testing results for PDMS and PS-coated PDMS contacted with a clean alumina silica probe. Even at very low compressive loads, full contact was reached and a large work of debonding was obtained. As expected, the adhesive strength of the elastomeric substrate coated with a thin glassy thermoplastic polymer film was slightly lower than the uncoated elastomeric surface.

4. Droplet Density Effect on Work of Debonding

Multiple flat punch adhesion tests varying in maximum compressive load, P_m, were performed on a region of interest having an areal droplet density, ρ_A, of 0.41. As P_m increases, the amount of PDMS-probe interfacial contact area increases as seen in SI Figure 4(b). The increase in interfacial contact results in a greater adhesive response that is apparent in SI Figure 4(a) as the critical pull-off load, P_c, increases with increasing applied load. The adhesive strength of the 0.41 density sample is significantly lower than the 0.33 density sample when comparing P_c values for the two probed regions of interest, even when apparent full contact is made signifying that droplet density plays an important role in altering the adhesive response.



SI Figure 3: Load-displacement curves for indentation tests performed on h = 0.5 µm droplet size samples at various P_m (a) and the respective images of PDMS-probe interfacial contact at P_m (b) for surfaces with $\rho_A \approx$ 0.41. Apparent full contact is achieved at 140 mN. The scale bar is 250 µm and applies to all images.