Supplementary information for: Geometry-controlled instabilities for soft-soft adhesive interfaces

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Section S1: Fracture mechanics to determine P'_s, δ'_s, and G

To calculate the adhesion energy (G) of confined soft-soft interfaces, values of the force and displacement in the absence of adhesion must be determined. These parameters are linked through the compliance of the soft system. These variables have been determined for the case of hard-on-soft adhesion using a correction factor f that accounts for effects of geometric confinement[1]:

$$\delta' = \delta_H f_\delta = \frac{a^2}{R}\left(0.4 + 0.6 exp\left(-1.8\frac{a}{h}\right)\right) \quad \text{Eq. S1}$$

$$C = C_0 f_C = \frac{(1-\nu^2)}{2Ea}\left(1 + \frac{4}{3}\left(\frac{a}{h}\right) + \frac{4}{3}\left(\frac{a}{h}\right)^3\right)^{-1} \quad \text{Eq. S2}$$

where a is the contact radius, R is the probe's radius of curvature, h is the thickness of the elastic layer, v is Poisson's ratio, and E is the elastic modulus. The compliance is defined as the slope of displacement versus force:

$$C \equiv \frac{d\delta'}{dP'} \quad \text{Eq. S3}$$

In the case of the soft-soft system, δ'_s must include components of both soft layers:

$$\delta'_s = \frac{1}{2}(\delta'_1 + \delta'_2) = \frac{a^2}{2R}\left(0.8 + 0.6\, exp\left(-1.8\frac{a}{h_1}\right) + 0.6\, exp\left(-1.8\frac{a}{h_2}\right)\right) \quad \text{Eq. S4}$$

As a result, P'_s can be found by substituting Eq. S3 and Eq. 2 into Eq. S3 above to yield:

$$P'_s = \int_0^a \frac{1}{\frac{1-\nu^2}{2Ea'}\left(\left(1+\frac{4}{3}\left(\frac{a'}{h_1}\right)+\frac{4}{3}\left(\frac{a'}{h_1}\right)^3\right)^{-1} + \left(1+\frac{4}{3}\left(\frac{a'}{h_2}\right)+\frac{4}{3}\left(\frac{a'}{h_2}\right)^3\right)^{-1}\right)} \frac{a'\left(1.6 + e^{-\frac{1.8a}{h_1}}\left(1.2 - \frac{1.08a'}{h_1}\right) + e^{-\frac{1.8a'}{h_2}}\left(1.2 - \frac{1.08a'}{h_2}\right)\right)}{2R}\, da'$$

Eq. S5

From numerical calculation of P'_s and the expression for δ'_s, the strain energy release rate, G, is calculated for each point during the measurement using Equation 5 in the main text.

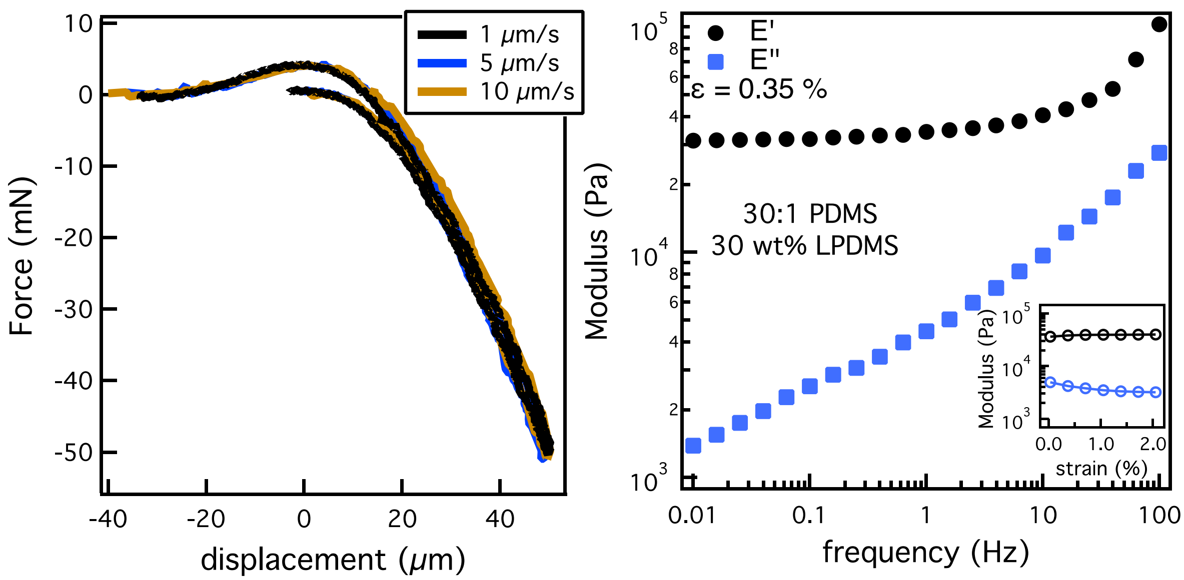

Figure S1: Rate-dependent force-displacement curves (left) and DMA (right) of PDMS samples used for experiments. The largely rate-independent force capacity and frequency-independent storage modulus ($0.01 < \omega < 10$ Hz) indicate a primarily elastic character of the network.

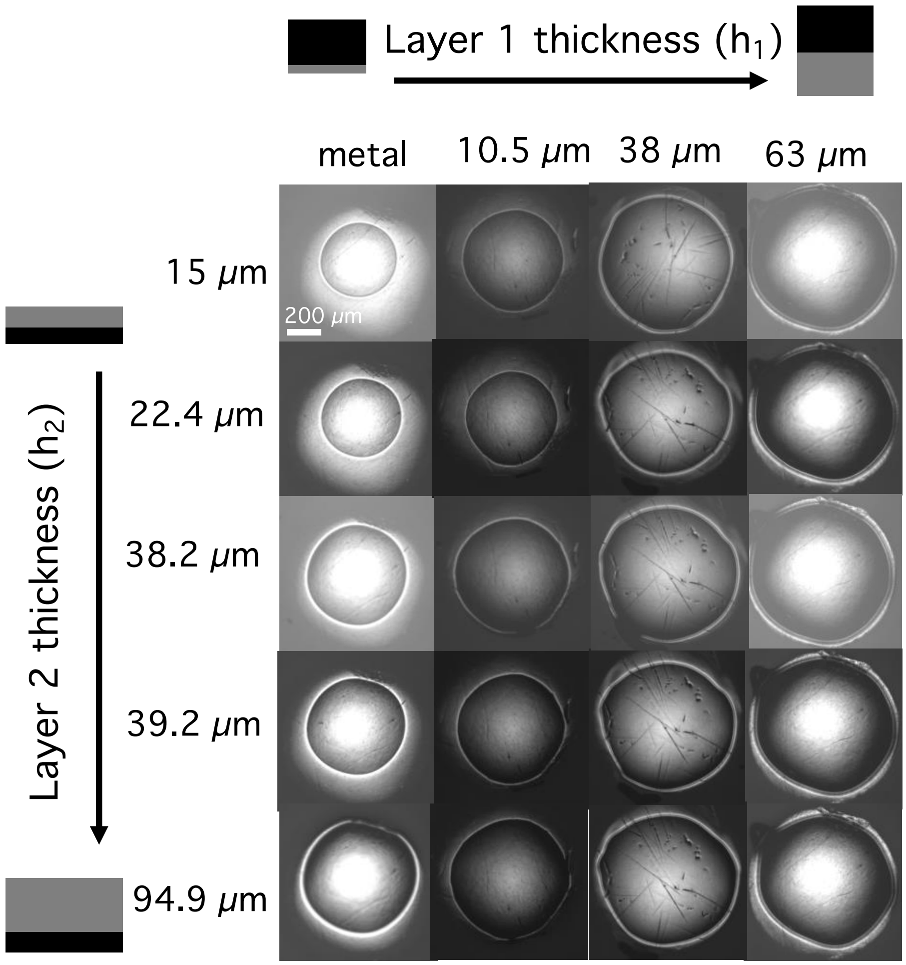

Figure S2: Maximum contact radius images. Images were contrast corrected by 20% for visualization.

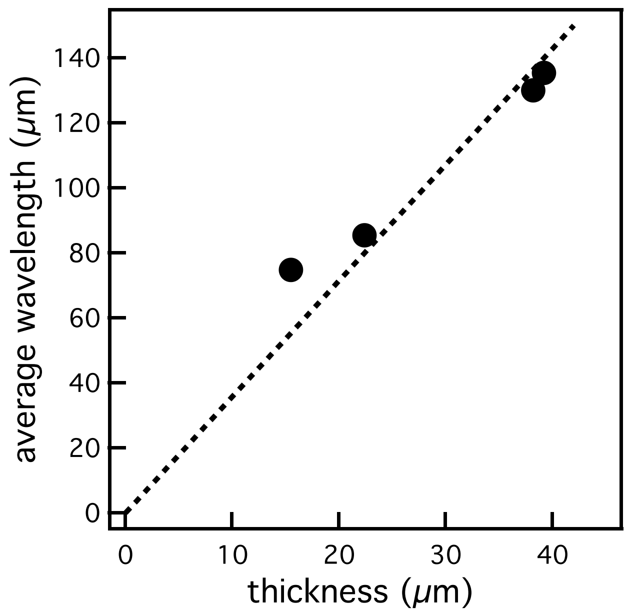

Figure S3: The average instability wavelength with thickness of PDMS. The best fit slope from least-squares fitting analysis was 3.57, comparable to a linear relationship of approximately 3 from previous work.[2]

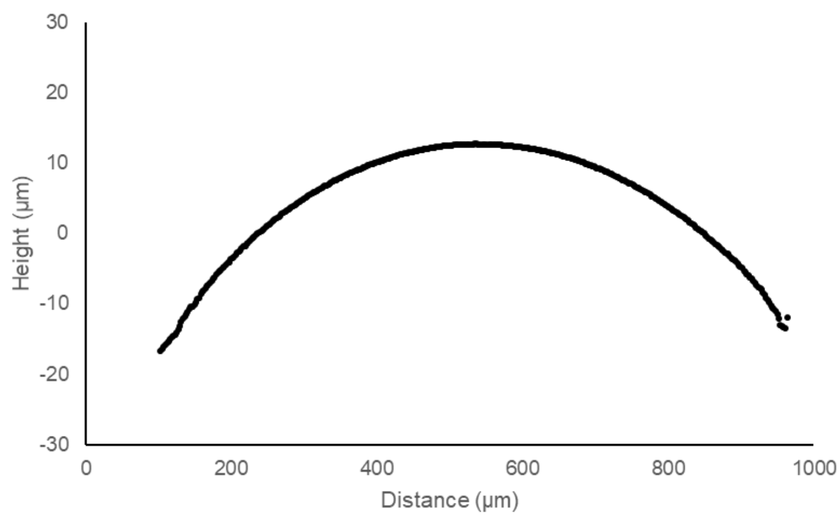

Figure S4: Optical profilometry of a polished probe used for contact adhesion tests. The radius of curvature is 3.5 mm and the root mean square (rms) surface roughness was less than 1 micron.

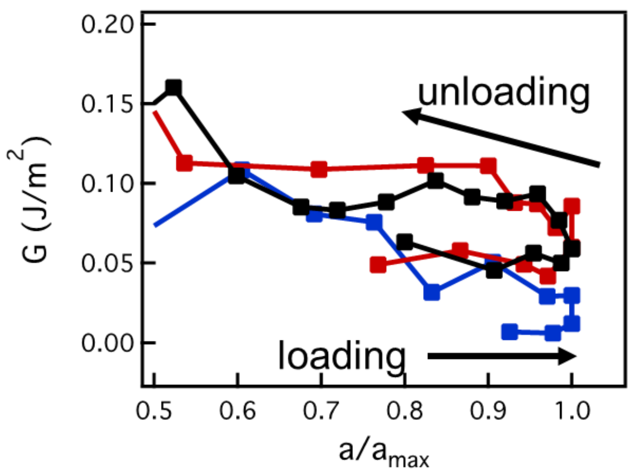

Figure S5: Plots of the energy release rate as a function of normalized contact radius for h_2 = 95 µm. During the loading phase when contact area is increasing, *G* remains nearly constant. After the maximum contact force is reached, the probe retracts from the substrate and the energy release rate increases. Once *G* overcomes the work of adhesion from surface interactions at the PDMS interface, the contact area decreases until debonding occurs. The value of *G* where the contact radius begins to decrease is defined as G_0. Similar values of G_0 are observed for each soft contact pair with h_2 = 95 µm with an average G_0 = 0.05 J/m^2, consistent with self-adhesion of PDMS based on its surface energy.[3]

References

1. K. R. Shull, D. Ahn, W.-L. Chen, C. M. Flanigan and A. J. Crosby, *Macromolecular Chemistry and Physics*, 1998, **199**, 489–511.
2. J. Y. Chung, K. H. Kim, M. K. Chaudhury, J. Sarkar and A. Sharma, *Eur. Phys. J. E*, 2006, **20**, 47–53.
3. A. C. M. Kuo, *Polymer Data Handbook,* Oxford University Press, New York, 2009.