Electronic Supplementary Material

for

"Adhesion-Controlled Capillary Origami"

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1. **Zero Thickness Limit**

   Assuming that a film being lifted from an adhesive substrate by a fluid puddle has no thickness allows for the simple calculation of the balance between capillary lift and film-substrate adhesion. With this assumption, bending energies can be ignored as can any gravitational effects. Assuming the geometry shown in the figure, and that the film has a width b in the 'in page' direction, a simple 2D energy balance can be constructed. We write the simplest possible argument below in order to keep clear the basic underlying physics.
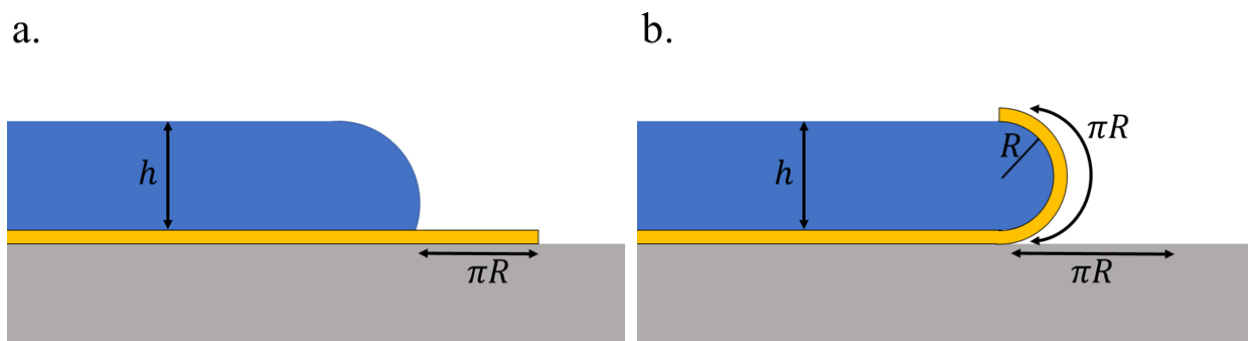
   a.   b.

   Figure 1a. schematic of a film in the flat state – it is adhered to a substrate with a water droplet on top of it. b. schematic of a film in the peeled state – here the film has bent around the edge of the fluid drop.

   a. **Energy of Flat State:**

   Before the film is (possibly) lifted by capillary forces, the total energy can be written:

   $$U_{flat} = \frac{U}{b} = \pi R(\gamma_{la} + \gamma_{fa} + k\gamma_{fs}) + \frac{1}{2}\rho g h^2 L + C.$$

   Here ρ is the fluid density, g the gravitational constant, L is the horizontal extent of the drop and C is a constant representing the energies of all the surfaces which do not change upon wrapping. R the radius of curvature at the edge of the water film. In this simple treatment, we consider the water edge to have the same shape before and after film wrapping. In our experiments, the substrate is patterned in order to control adhesion. This means that only a fraction (k) of the film's total surface area is ever in contact with the substrate. For example, $k = 1$ on an unpatterned substrate. Additionally, the subscripts refer to: a=air, l=liquid, f=film, s=substrate. See for example Ref. 4.

   b. **Energies of Peeled State:**

   Next, we consider the total energy after a substrate has been successfully peeled by the fluid drop:

$$U_{peeled} = \pi R\left(\gamma_{lf} + k\gamma_{fa} + k\gamma_{sa}\right) + \frac{1}{2}\rho g h^2 L + \frac{\pi B}{2R} + C.$$

Of course, here we assume zero thickness, so the bending term goes to zero, leaving:

$$U_{peeled} = \pi R\left(\gamma_{lf} + k\gamma_{fa} + k\gamma_{sa}\right) + \frac{1}{2}\rho g h^2 L + C.$$

c. **Energy Balance:**

To find the limiting value of adhesion (here through the fraction k) one can write the difference in energy between peeled and unpeeled states and then set it to zero.

$$\frac{\Delta U}{b} = (Peeled\ State) - (Flat\ State).$$

$$\frac{\Delta U}{b} = 0 = \pi R(\gamma_{lf} + k\gamma_{fa} + k\gamma_{sa} - \gamma_{la} - \gamma_{fa} - k\gamma_{fs}).$$

$$k(\gamma_{fs} - \gamma_{fa} - \gamma_{sa}) = (\gamma_{lf} - \gamma_{la} - \gamma_{fa}).$$

$$k = \frac{\gamma_{lf} - \gamma_{la} - \gamma_{fa}}{\gamma_{fs} - \gamma_{fa} - \gamma_{sa}}$$

This is the general solution for the boundary between peeled or flat favored state. In our case we can simplify further due to the film and substrate both being PDMS. Therefore $s = f$ and:

$$k = \frac{\gamma_{lf} - \gamma_{la} - \gamma_{fa}}{\gamma_{ff} - 2\gamma_{fa}} = \frac{\gamma_{la} + \gamma_{fa} - \gamma_{lf}}{G_c}.$$

d. **Calculation of k_{limit}**

Furthermore, we can use measured values for each of the relevant surface energies and directly calculate k. For example, literature sets the value for the PDMS surface as $\gamma_{fa} = 0.019$ N/m.[1] A water surface energy value was measured directly with the Wihelmy plate method using a KSV Layerbuilder apparatus, $\gamma_{la} = 0.049 \pm 0.0025$ N/m. This deviation from water's ideal surface tension is suspected to be due to the contamination of PDMS filler leaching into the liquid from the Sylgard 184 and acting as a surfactant. A similar effect is seen in the glycerine rather than water on PDMS.[2,3] Using γ_{fa} and a directly measured contact angle ($\theta_e = 92.6°$ - advancing contact angle) with the Young-Dupre Equation we find $\gamma_{lf} = 0.021$ N/m. In a direct measurement outlined below we find $G_c = 0.124$ N/m. Ultimately this yields the value $k_{limit} = 0.38$.

2. **Zero Film-Adhesion Limit**

The limit of the maximum thickness a film could have such that it could be bent to the peeled state can similarly be calculated. In our notation, the limit amounts to letting $k = 0$ as well as adding a term proportional to the bending energy of the sheet.

a. **Energy of the Flat State:**

$$U_{flat} = \frac{U}{b} = \pi R(\gamma_{la} + \gamma_{fa}) + \frac{1}{2}\rho g h^2 L + C$$

b. **Energies of Peeled State:**

$$U_{peeled} = \frac{U}{b} = \frac{1}{2}\left(B\left(\frac{1}{R}\right)^2 \pi R\right) + \frac{1}{2}\rho g h^2 L + \pi R(\gamma_{lf}) + C$$

Where B is the bending modulus, given by $B = \frac{Et^3}{12(1-\nu^2)}$.

c. **Energy Balance:**

$$\frac{\Delta U}{b} = Peeled\ State - Flat\ State$$

$$0 = \pi R \left(\frac{1}{2} B \left(\frac{1}{R} \right)^2 + \gamma_{lf} - \gamma_{la} - \gamma_{fa} \right)$$

Assuming the bending modulus as above, we find:

$$\gamma_{la} + \gamma_{fa} - \gamma_{lf} = \frac{1}{2} \left(\frac{Et^3}{12(1-v^2)} \left(\frac{1}{R^2} \right) \right)$$

Next, we isolate thickness:

$$t = \left(\frac{(\gamma_{la} + \gamma_{fa} - \gamma_{lf})(24R^2(1-v^2))}{E} \right)^{1/3}$$

We note that the fluid drop thickness is twice the radius of curvature and assume that the puddle height remains the same before and after wrapping. The puddle height is easily calculated,[4]

$$h = 2\sqrt{\frac{\gamma_{la}}{\rho g}} \sin \left(\frac{\theta_e}{2} \right).$$

With the puddle height we finally arrive at a thickness of

$$t = \left(\frac{(\gamma_{la} + \gamma_{fa} - \gamma_{lf})(24(\gamma_{la} \sin^2(\frac{\theta_e}{2}))(1-v^2))}{E\rho g} \right)^{1/3}.$$

Using the values listed above, as well as a Poisson ratio of $v = 0.5$ (commonly used for PDMS elastomers, $E = 1.69\ MPa = 1.69 \times 10^6$ Pa (directly measured in [5]), $pg = 9800$ kg/m²s and $\theta_e = 92.6°$ (Directly measured advancing contact angle), we arrive at the thickness limit of: $t_{limit} = 0.000110$ m or $110\ \mu m$.

3. **Energy Balance Function**

Following the same approach used in deriving limits based upon the free energy of the system, a more complete limit can be constructed for arbitrary values of adhesion and film thickness.

   a. **Energies of Flat State:**

$$U_{flat} = \frac{U}{b} = \pi R [\gamma_{fa}(2-k) + \gamma_{fs}k + \gamma_{wa} + \gamma_{sa}(1-k)] + \frac{1}{2}\rho g h^2 L + C$$

   b. **Energies of Peeled State:**

$$U_{peeled} = \frac{U}{b} = \pi R \left[\gamma_{lf} + \gamma_{fa} + \gamma_{sa} + \frac{1}{2} B \left(\frac{1}{R} \right)^2 \right] + \frac{1}{2}\rho g h^2 L + C$$

Once again, we assume the puddle thickness is fixed by capillarity,

$$R^2 = \frac{\gamma_{la}}{\rho g} \sin^2 \left(\frac{\theta_e}{2} \right),$$

and so,

$$U_{peeled} = \pi R \left[\gamma_{lf} + \gamma_{fa} + \gamma_{sa} + \frac{Et^3 \rho g}{24\gamma_{la}(1-v^2)\sin^2(\frac{\theta_e}{2})} \right] + \frac{1}{2}\rho g h^2 L + C.$$

Peeling is favored if $U_{peeled} \leq U_{flat}$, and we take 0 as the limiting case. Hence, we have:

$$U_{peeled} - U_{flat} = 0.$$

Or, after some simplification:

$$\gamma_{lf} - \gamma_{fa} - \gamma_{la} + k(\gamma_{fa} + \gamma_{sa} - \gamma_{fs}) + t^3 \frac{E\rho g}{24\gamma_{la}(1-v^2)\sin^2(\frac{\theta_e}{2})} = 0.$$

And once again isolating the thickness gives:

$$k[t] < \frac{\left(\gamma_{la}+\gamma_{fa}-\gamma_{lf}-t^3\frac{E\rho g}{24\gamma_{la}(1-v^2)\sin^2\left(\frac{\theta_e}{2}\right)}\right)}{(\gamma_{fa}+\gamma_{sa}-\gamma_{fs})}.$$

This is the general solution for the fraction of adherence to the substrate where peeling will occur. Using experimental values will give a value for k below which, peeling is expected to occur. In the case of these experiments, $s = f$ leading to the equation of the boundary as:

$$k[t] = \frac{\left(\gamma_{la}+\gamma_{fa}-\gamma_{lf}-t^3\frac{E\rho g}{24\gamma_{la}(1-v^2)\sin^2\left(\frac{\theta_e}{2}\right)}\right)}{(G_c)}.$$

Substituting the parameter values noted above we find,

$$k[t] < 0.38 - (2.9 \times 10^{11})t^3,$$

in the case of the materials presented here.

4. **Comments on Calculations**

Obviously, our treatment here is very simple. A more complete treatment would focus on minimizing h in the free energy functionals discussed above. While this approach is more thorough, we don't arrive at any conclusions which are more meaningful for our experiment in doing so, and thus cannot justify the added complexity.

Further, representing the sheet with Euler's elastica would give a more realistic idea of the actual curvature present at the film edge. These calculations would be complex due to the moving boundaries (where the film contacts the substrate and where the film contacts the fluid) as well as the deformability of the fluid surface. We leave this idea for the more theoretically inclined to pursue.
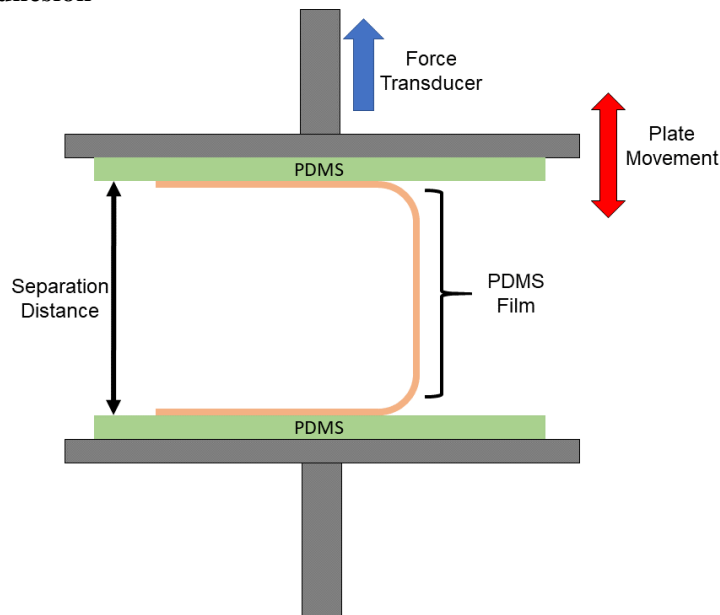
5. PDMS-PDMS Adhesion

Figure 2. Experimental "tape loop" adhesion setup. Here a thin PDMS film (orange) is placed between two plates coated with a thick PDMS layer. Forces are then measured as the film is compressed, and then pulled open in tension. The force-displacement curve gives the film modulus, work of adhesion as well as the energy release rate.

This section outlines an experiment to determine the adhesion values of the PDMS-PDMS interface using the methods of Elder *et al.*. [6] The difference here was that the top and bottom plates were glass coated with 10:1 PDMS rather than being glass alone. A PDMS "tape loop" was placed between the two plates and compressed past the system's elastocapillary length. Then the plates were separated, while the force was recorded. Plate velocities are listed below. A typical force vs. motor position curve is shown below, with positive values representing a compressive force on the bottom plate whereas a negative force represents a tension on the bottom plate and the adhesive effect of the tape loop.
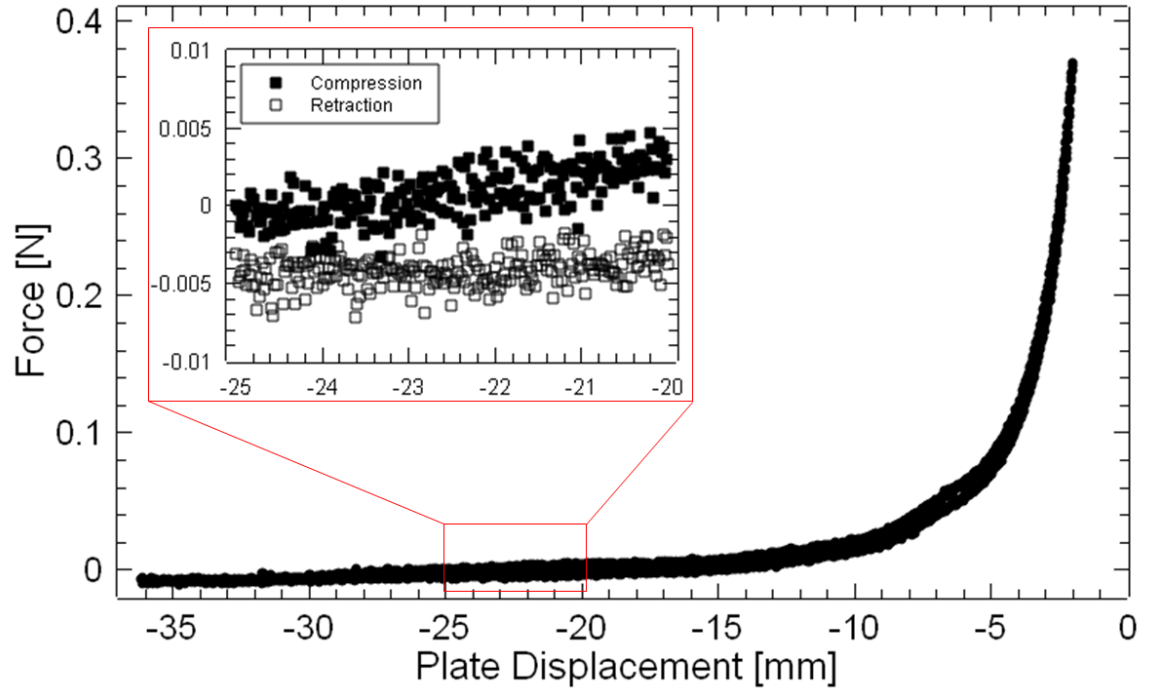
Figure 3. A typical force-displacement curve for a PDMS loop in contact with two PDMS surfaces. Inset shows the difference between the closing and opening part of the data in the 'plateau' part of the cycle.

The difference in measured force between the compression and separation curves at the same plate position were calculated while the loop was in the peel 'plateau' (see figure 2 and 3). During this part of the force-displacement cycle the force is equal to the loop width times the measured (negative) force. The experiment was repeated multiple times and at several different speeds (data shown in Figure 4 Below). For our analysis we use the G_c value of 0.124 N/m which was measured at the slowest velocity 9×10^{-5} m/s.
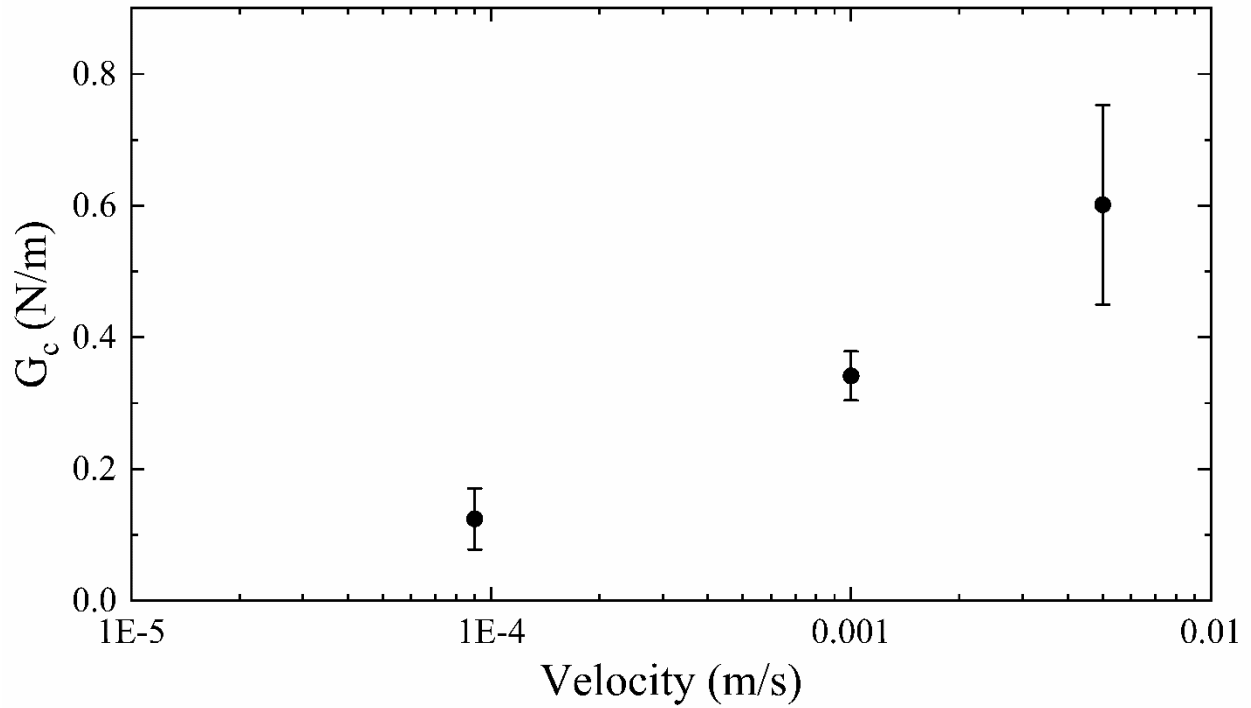
Figure 4. Gc values for PDMS-PDMS contact at various velocities. In this work the value at the lowest velocity is used as it is near the 'zero velocity' limit of the true work of adhesion. The trend suggests that very slow peels would have a smaller adhesion value, but experiments at these speeds proved difficult. Likewise, the capillary peel experiments did not show peel over very long time-frames, with capillary peel observed within a few minutes on all of the peeled films.

6. Double Fold Substrate Map
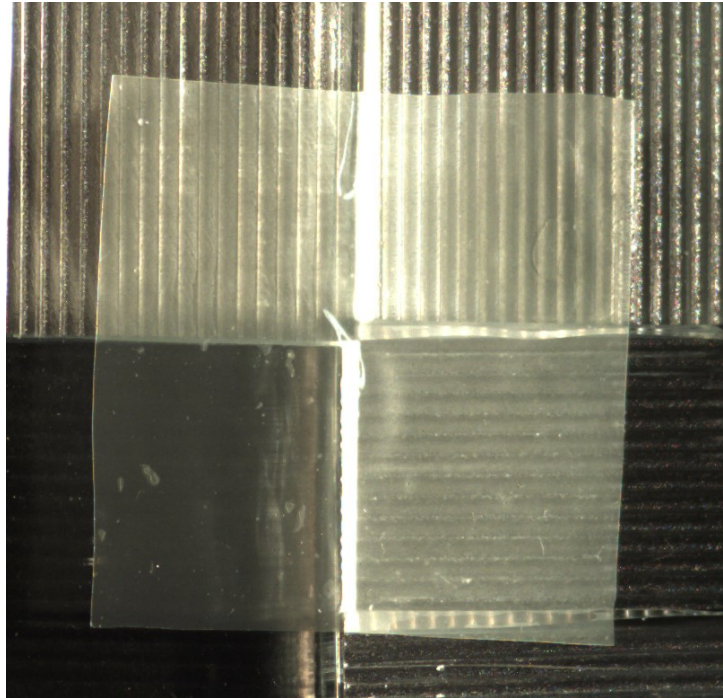
Figure 5. A film resting on an adhesion patterned substrate.

A thin elastomeric film (polyvilylsiloxane for visibility) is placed upon a substrate consisting of regions with high film-substrate adhesion (nearly 100%) and regions of very low film-substrate adhesion (around 1%). The basic idea is that by adding water to either the center of the film, or some other general and large region, the desired folds will be facilitated. The rectangular film is shown in figure 5 and figure 6.
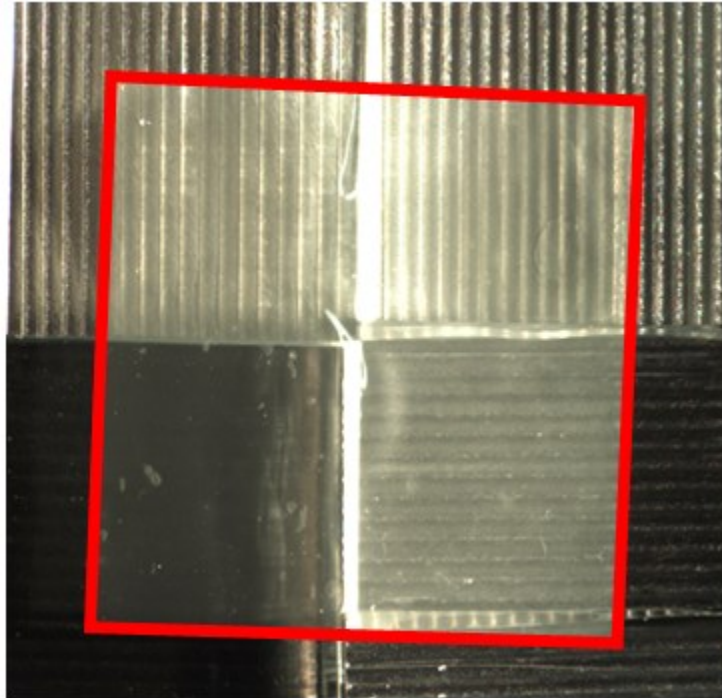

Figure 6. A film resting on an adhesion patterned substrate. Film is outlined in red for clarity.
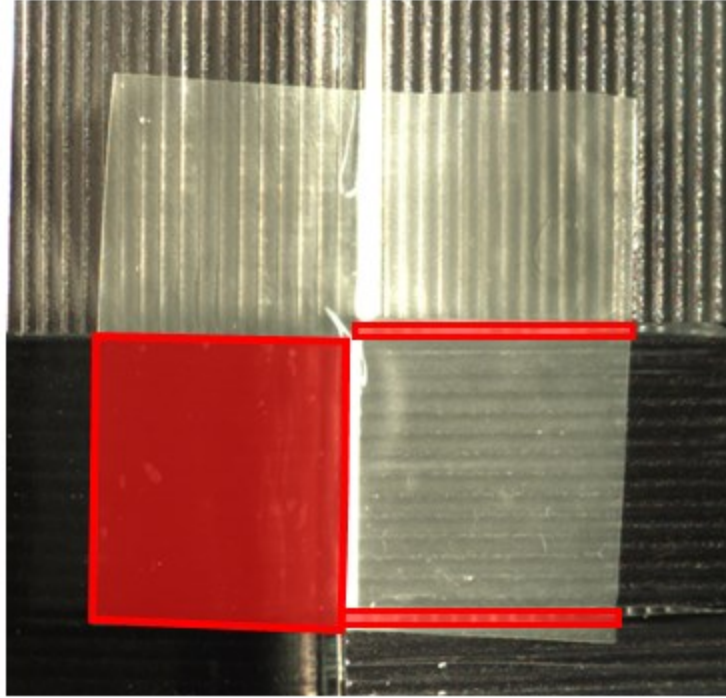
Figure 7. Film resting on an adhesion patterned substrate. Red regions highlight high adhesion areas of the pattern.

The adhesive regions are placed to guide the two folds necessary for the double-fold design to be created (Figure 7). A peel will not begin in the adhesive regions and will usually peel along the path of least resistance. This means that a water drop placed in the center of the film will lift the top half of the film first. This will create a peel front on the low-adhesion substrate that will progress towards the center line until it encounters a region of high-adhesion. The film will continue to peel from the low-adhesion substrate until the peel front has reached the high-adhesion region at which point peeling is arrested. The peel front encounters the transition to the high-adhesion zone nearly parallel, effectively transitioning from peeling from an area with 1% adhesion directly to nearly 100% adhesion, which cannot be overcome with capillary peeling. This first step is often very quick. Before and after images can be seen in figure 8.
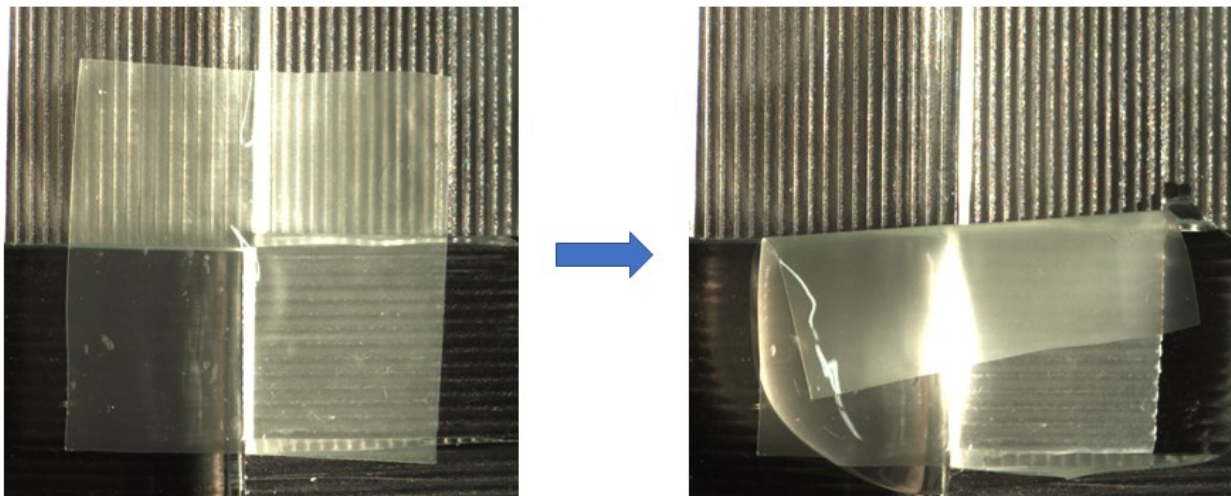
Figure 8. Before fluid loading and after fluid loading. Here the film peels readily from the low adhesion region (top two quadrants) and folds around the water. The film does not peel further than the horizontal high adhesion 'line' along the pattern center.

After the first fold, the water dries leaving the film in an elongated racquet formation. This can also create a small amount of peeling along the rounded edge of the racquet after the flat regions of the film self-contact and become stuck. Water is then added to the top of this film creating a new peel front on the right side of the now double-thick film. The horizontal high adhesion line means that peeling will not occur top down and the high adhesion left lower quadrant means peeling will not begin from the left side. Peeling can only occur from the right side of the pattern, and only if the 'double thick' film is still thin enough to be below the peel criteria outlined in the main paper. Figure 9 shows the process. We note the final folded film is often offset by a small angle due to the rounded fold from the first step of the process.
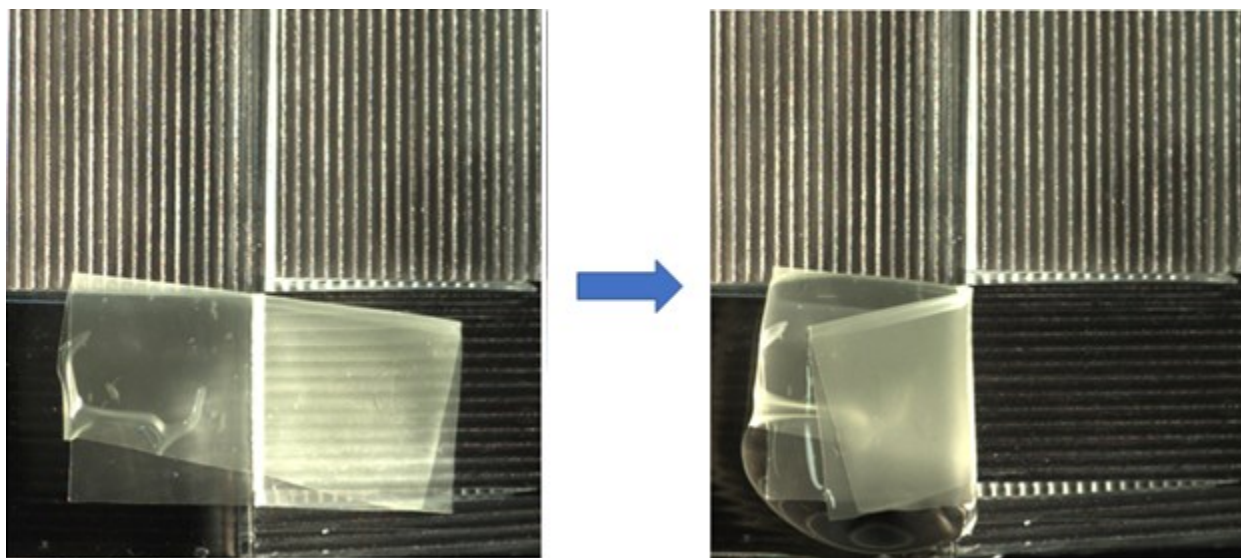

Figure 9. Second step of the folding process. A water drop added to the top of the (dried) structure causes peeling from the right, resulting in a double fold.

A video of this process is available as a supplementary file.
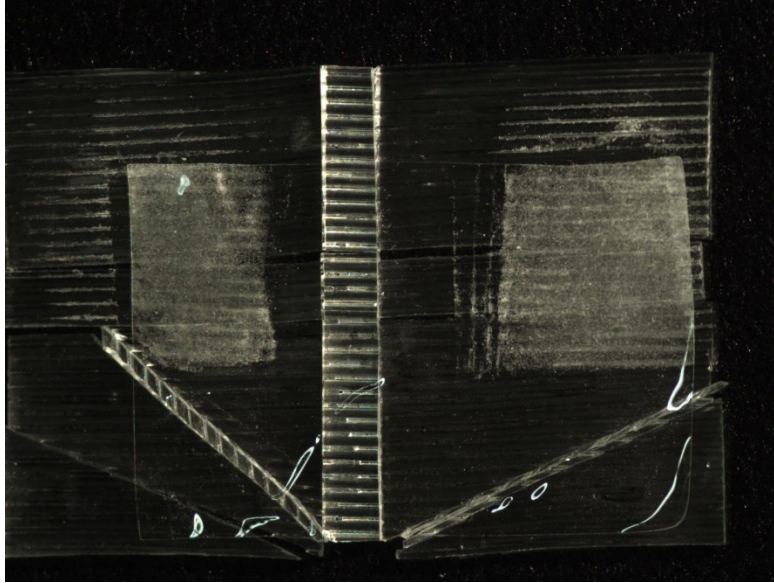
7. Airplane Substrate Map

Figure 10. a rectangular sheet resting on an adhesion guiding substrate.

A thin, rectangular PDMS film is placed on a prepared adhesion guiding substrate (Figure 10). This setup has regions of high or low adhesion similar to the double-fold, but with the added complexity of parts of the top of the film being coated with nonstick particles, effectively lowering the adhesion of those areas to anything they contact to nearly 0%. The adhesive areas of the substrate are also coated with a release layer that dissolves in water allowing the structure to detach from the substrate easily. The film is outlined in Figure 11.
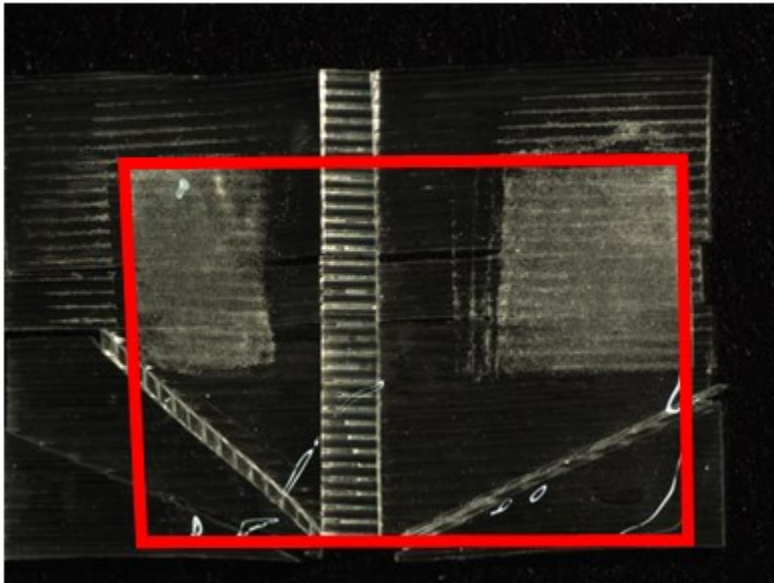

Figure 11. A rectangular film resting on a guided adhesion substrate. Film highlighted in red.

In figure 12, the areas highlighted in red are nearly 100% adhered to the substrate, and areas highlighted in blue have a nonstick coating applied to the top of the film.
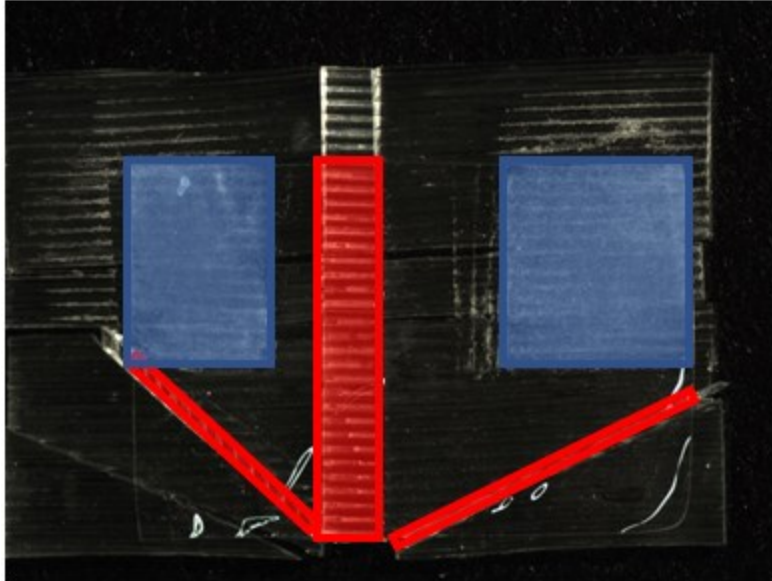
Figure 12. Regions of high adhesion (red) and zero adhesion (blue) are highlighted.

The fold progression is: (a.) bottom corners upwards onto wings, (b.) right wing onto left wing, (c.) top wing folds off bottom wing, (d.) the release layer is dissolved, allowing the airplane to float freely. This progression is visualized in the following sequence. A video of the process is available in supplementary material.
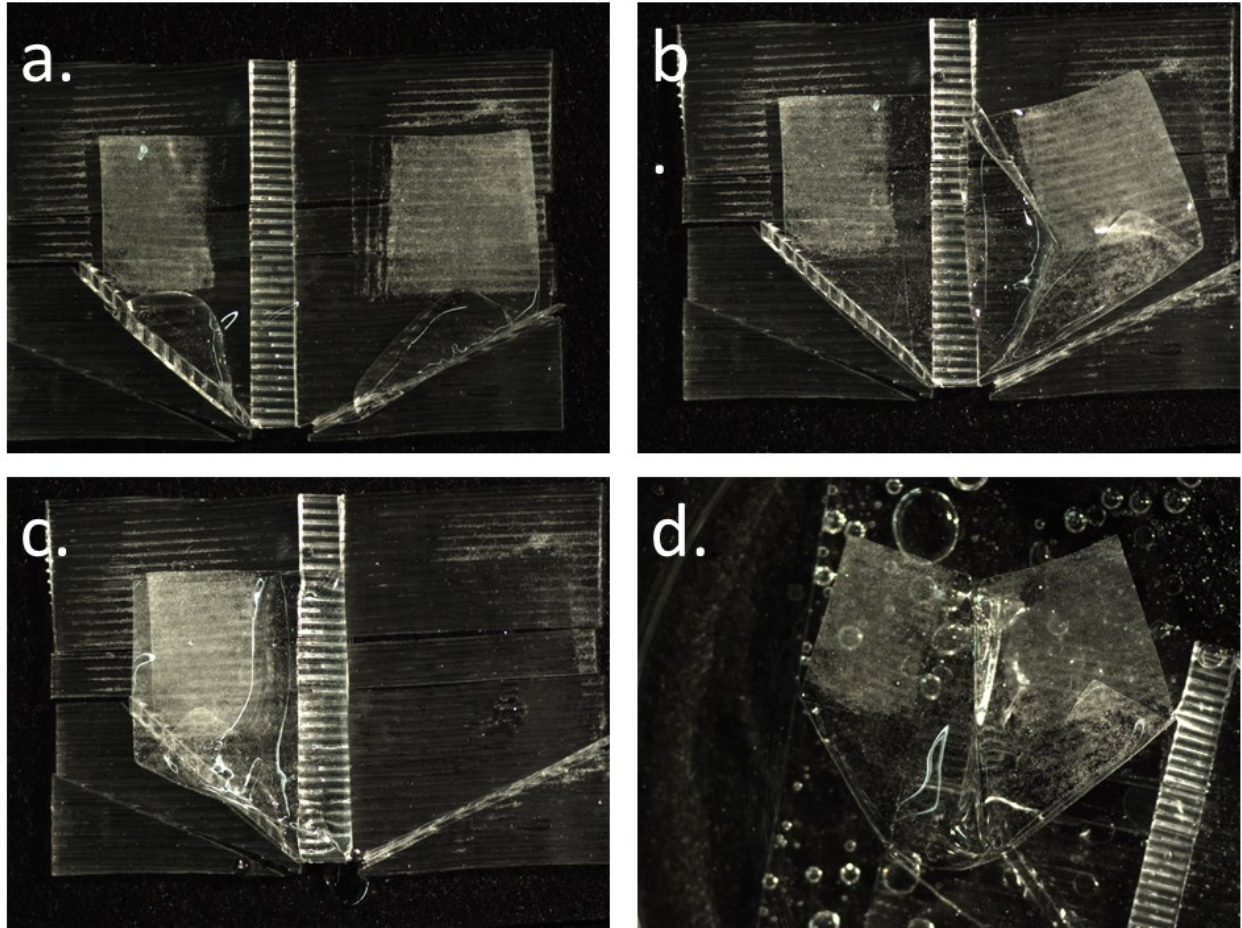
Figure 13. Several stages in airplane assembly. a.) bottom corners folded up with fluid drops. Fluid is then dried. B.) A second drop is added in the center of the film causing the right hand side to fold to the left. c.) shows the end of this stage of the process. A fluid drop is then added to the top of the film which folds the 'wing' open. d.) Shows the released airplane.

8. References

1. Efimenko, Kirill, William E. Wallace, and Jan Genzer. "Surface modification of Sylgard-184 poly (dimethyl siloxane) networks by ultraviolet and ultraviolet/ozone treatment." *Journal of colloid and interface science* 254 (2002): 306-315.
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3. Style, Robert W., et al. "Universal deformation of soft substrates near a contact line and the direct measurement of solid surface stresses." *Physical review letters* 110 (2013): 066103.
4. P.-G. De Gennes, F. Brochard-Wyart and D. Quéré, "Capillarity and wetting phenomena: drops, bubbles, pearls, waves", Springer Science & Business Media, 2013.
5. Elder, Theresa, Damith Rozairo, and Andrew B. Croll. "Origami inspired mechanics: measuring modulus and force recovery with bent polymer films." *Macromolecules* 52 (2019): 690-699.
6. Elder, Theresa, et al. "Adhesion of a tape loop." *Soft Matter* 16 (2020): 10611-10619.