Supplementary Materials for
Kuttsukigami: Sticky Sheet Design

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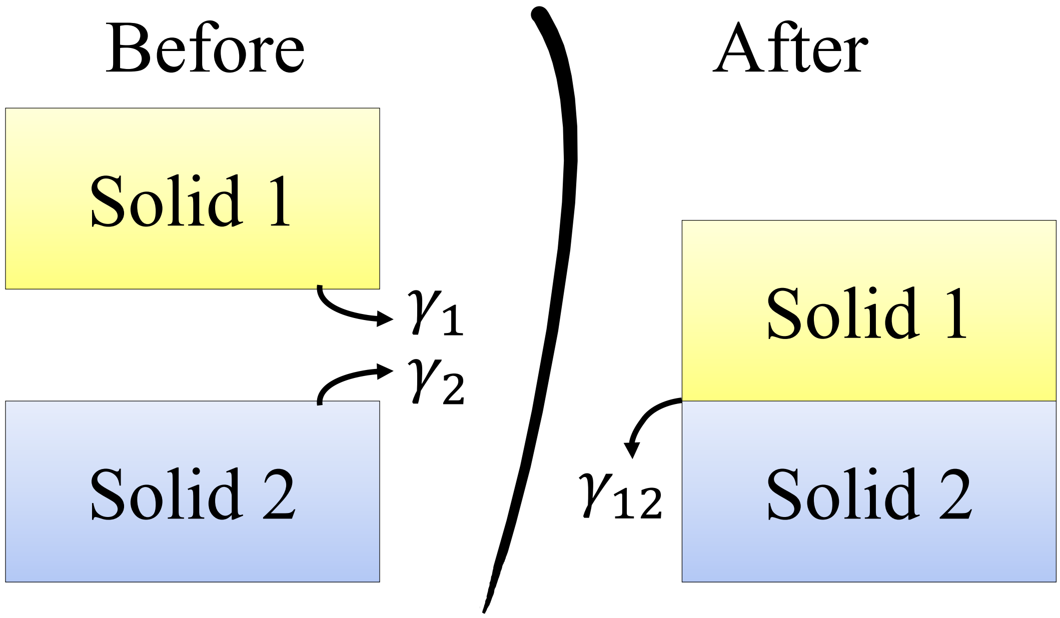

Fig. S1. Schematic showing two solids first out of contact, and later in contact. When contact is formed, surface area of solid 1 and solid 2 is replaced by a solid 1/solid 2 interface.

I. KUTTSUKIGAMI

We honour the Asian cultures from which origami was originally perfected as an art-form, by using the Japanese language to name the technique of "Sticky Origami" discussed in this work. Origami itself is a word formed from combination of "ori" or folding with "kami" or paper. Similarly, Kirigami, which uses cuts to create structure and function, is a word combining "kiri" or cut with "kami". Following the pattern, one naturally arrives at Kuttsukigami by combining "Kuttsuku" or stick together, with "kami".

II. ADHESION BASICS

For the convenience of those not intimately familiar with the science of adhesion, we present a short discussion of the important general concepts. Figure S1 shows a schematic of an adhesive process. Before separation, two materials 1 and 2 form an interface 12 along which a crack might propagate and eventually separate the two materials. If separated, the

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materials 1 and 2 must create new interface of the same total area (A) of the former 12 interface. An energy is associated with each interface, U_i, which is proportional to the total contact area, e.g. $U_i = \gamma_i A$, where γ_i is a proportionality constant associated with interface i. Quite commonly γ_i is referred to as a surface energy.

The question of adhesion is then most simply defined by comparison of the energies in the two different states. For example,

$$\Delta U_{interface} = \gamma_2 A + \gamma_1 A - \gamma_{12} A = wA, \tag{S1}$$

defines the difference in energy between the separated and contacting states. $w = \gamma_1 + \gamma_2 - \gamma_{12}$ defines the Young-Dupré work of adhesion. If ΔU is less than zero ($w < 0$) free energy is lower when the solids are in contact. This is a simple way to define adhesion: w relates to how strongly the two solids are 'bonded' to one another.

Unfortunately most materials, especially soft materials like the elastomers and gels discussed in the main manuscript, have additional losses associated with the motion of the crack which must propagate along the 12 interface in order to separate the two materials. From this point of view, adhesion is best determined by examining the process of the propagation of the interfacial crack. In this case, the total energy must consider the mechanical energy applied to the system as well as the interfacial energy itself: $U_{total} = U_{mechanical} + U_{interface}$. We can then consider how the motion of a small "virtual" crack which would create some small amount of interface, ∂A, changes the overall energy. In particular,

$$\partial U_{total} = (G - w)\partial A, \tag{S2}$$

where we have defined the systems energy release rate by $G \equiv \partial U_{mechanical}/\partial A$. If $G < w$ the system reduces energy by closing the virtual crack and if $G > w$ opening the crack decreases free energy. G represents the measured part of the system and so includes any losses which occur during crack propagation [1].

An adhesion experiment typically involves an instrument which measures forces and displacements as stress is increased along the interface. Often experiments report a critical force, F_c, measured at the point where the interface completely separates. Note that defining the critical point requires a stability analysis of the free energy because a crack will open for a range of G values. It is not enough for $G \geq w$, but in fact the system must also have $\partial G/\partial A \leq 0$ for the crack to drive through the interface. The critical energy release rate (defining the critical force through $U_{mechanical}$) occurs at the last point of stability. In other words, G_c occurs uniquely at the point where $\partial G/\partial A = 0$.

III. SOME ADDITIONAL EXAMPLES OF KUTTSUKIGAMI DESIGN

Fig. S2. Photograph of several kuttsukigami designs.

IV. ADHESION OF A RACQUET

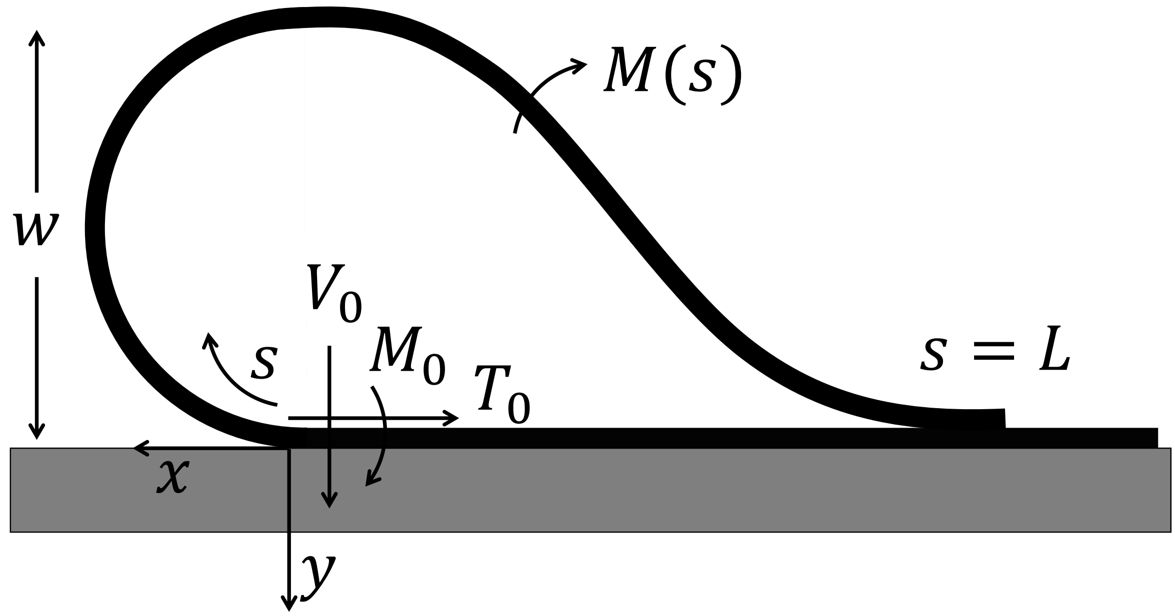

Fig. S3. Schematic racquet shape bend defining several quantities discussed in the supplementary text.

Here we describe the racquet shape following the formalism of Glassmaker and Hui in [2]. Given a sheet of length L bent in a racquet like shape as shown in fig. S3, moments and tensions must be balanced in order for the structure to remain in equilibrium. The moment of the sheet at any point can be given by $Bd\theta(s)/ds$, where $\theta(s)$ is the angle the sheet makes with the horizontal at any point, s, along its length. B is the bending modulus. Explicitly, the equilibrium is given by,

$$M - M_0 - T_0 y - V_0 x = 0. \tag{S3}$$

where the 0 subscript indicates values of moment, tension and shear at the end of the plate. Differentiating Eqn. S3 while noting that $dx/ds = \cos\theta$ and $dy/ds = \sin\theta$, yields:

$$B\frac{d^2\theta}{ds^2} - T_0 \sin\theta - V_0 \cos\theta = 0. \tag{S4}$$

Adhesion with of the strip with itself or a substrate enters through the adhesive boundary conditions (e.g. $\theta'(L) = \sqrt{2G_c/B} = \sqrt{2}\ell_{ea}$). Again we follow Hui and denote the second adhesive boundary as a proportionality to the first ($\theta'(L) = \sqrt{2\mu}\ell_{ea}$, where μ is the ratio of critical energy release rate of the substrate/film interface to that of the film/self interface). Coupled with the more obvious boundary conditions ($x(0) = 0, y(0) = 0, \theta(0) = 0, y(L) = 0$ and $\theta(L) = -\pi$) one arrives at the elastica:

$$\frac{d^2\theta}{ds^2} - \frac{1}{2\ell_{ea}^2}(1-\mu)\sin\theta - \frac{V_0}{B}\cos\theta = 0. \tag{S5}$$

Equation S5 can be solved numerically and fit to measured racquet contours to identify precise values of the adhesive parameters in an experiment. Glassmaker also noted the usefulness of identifying how the structure depends on the intrinsic length ℓ_{ea} and how this length relates to more simply measured quantities such as the racquet width. In particular, $W = \eta(\mu)\ell_{ea}$ where η is a numerically determined function of μ and is of order 1 when the film/substrate adhesion is low (as in our experiments).

V. CYLINDRICAL CLOSURE

We note that a detailed treatment of an adhesive loop would use boundary conditions at the point of overlap to derive an elastica-like equation for the position of the sheet. This is necessary because there are no forces holding the sheet in a circular shape, as would be the case of a film rolled tightly upon itself (an Archimedean spiral). This said, a simple argument for the necessary conditions for a crack to propagate along the overlap region can be made if the loop is assumed to be sufficiently large, and the overlap sufficiently small, such that the loop can be approximated as a cylinder. At the crack tip, an incremental length of the crack $d\ell_{overlap}$ might open, freeing only the local bending energy in the opened part of the crack $\sim \frac{BL}{2R^2} d\ell_{overlap}$ and reducing the adhesion by $G_c L d\ell_{overlap}$. This local balance would conclude with the prediction that the closure would be spontaneously unstable for:

$$R < \sqrt{B/2Gc} \tag{S6}$$

VI. ADHESION OF A MÖBIUS STRIP

A Möbius strip is also easily constructed by adhering a small portion of a twisted strip to itself, forming another adheso-elastic problem. In this case a simple argument for crack propagation is not so clear due to the addition of tortion to the bending energy. We can, however, take a step in the right direction with a broad argument: energy stored mechanically in the structure must be less than the energy stord by adhesion for the loop to remain stable. The elastic energy stored in a developable Möbius strip is not easily determined, though recent calculations show promising results [3, 4]. For this work we are interested in the mechanical energy stored in the structure, which both works find to scale as $U_{tot} \sim (w/L)B$, where B is the bending modulus of the strip, L is its length, and w is its width. The structure is fabricated by overlapping a small length, $L_{overlap}$, and we assume this small double thickness region does not appreciably alter the bending energy of the structure (foci will naturally avoid this region). The adhesion energy required to hold the structure closed is given by $U_a = \gamma w L_{overlap}$. Equating the mechanical and adhesion energies then allows the scaling of the minimum overlap length to be estimated:

$$L_{overlap} \sim B/\gamma L. \tag{S7}$$

We note that this total energy comparison will only yield a broad boundary for what structures are physically impossible. This broad veiwpoint can be corrected by a more detailed approach that considers the energy released in each step of infinitesimal crack propagation. The argument used for the cylinder and spiral is not directly applicable here because the curvature at the point of film overlap is not easily related to the overall size of the loop. Further, the more difficult to estimate release of tortion will also contribute to the detailed balance at the crack tip. We leave this detailed calculation for a future work.

VII. CLOSURE OF A RACQUET

To form the simplest type of packet, a sheet can be first formed into a racquet shape and then can be closed by squeezing the open end together. As the racquet shape is minimal, additional energy storage is needed to maintain this new, closed, geometry. Note that the closure will change the Gaussian curvature from zero to non-zero which means in-plane stretching has been created. The basic geometry is sketched in figure 4, which we note is very similar to the geometry of the ridge formed between two d-cones [5].

The basic geometry allows us to make a scaling estimate of the length of sheet distorted by the racquet closure, x_0 in fig. S4. The strain created along the apex of the racquet can be estimated by comparing the length of the hypotenuse of a triangle with sides x_0 and δ

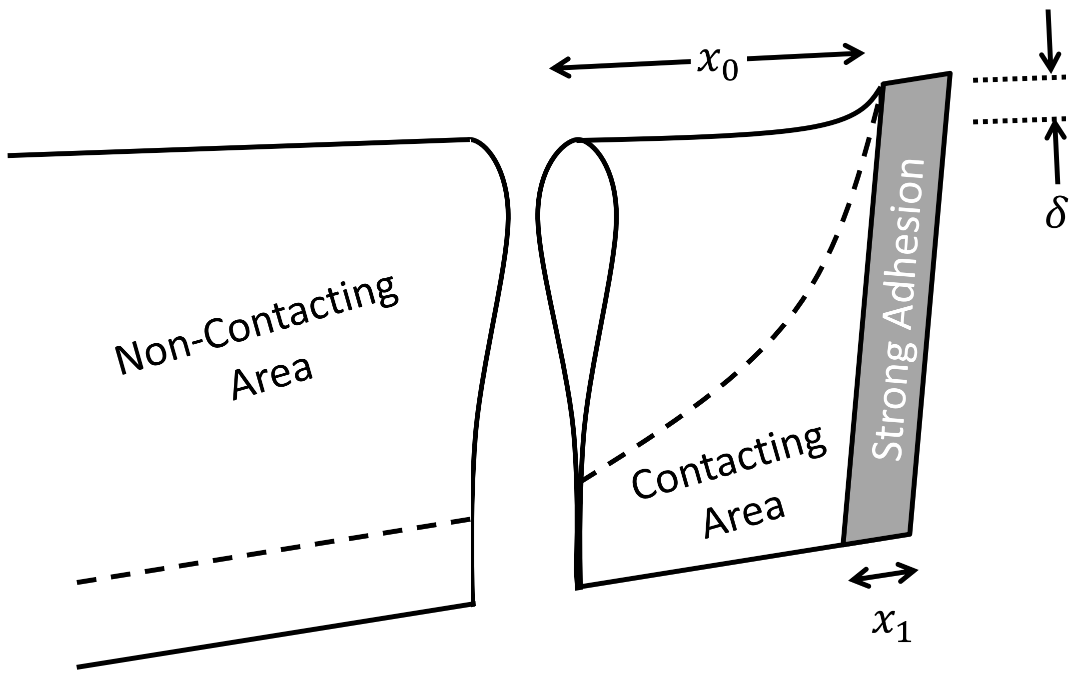

Fig. S4. The geometry of a racquet closure. The racquet is shown with an imaginary cut for clarity. The high adhesion zone forces the sheet to come into complete contact, which creates stretching in the sheet over a distance of x_0. The area below the dashed line is in contact and could therefore be influenced by sheet-sheet adhesion.

to the unstretched length x_0. In this case the strain scales as $\epsilon \sim \delta^2/x_0^2$ when assuming $\delta \ll x_0$. The strain energy is given by $U_s \sim (Et/2)\epsilon^2 A$, where A is the area over which the strain is spread. The strain is localized to the ridge, so $A \sim \ell_{ea} x_0$ as we assume the width of the racquet to scale as the elasto-adhesive length. δ can also be shown to be related to the racquet width under the assumption that there is no stretching orthogonally to the ridge (e.g. $\delta = (\pi/2 - 1)\ell_{ea}$). Dropping the $(\pi/2 - 1)$ and other constants for simplicity, $U_s \sim Et\ell_{ea}^5/x_0^3$.

There is also increased bending caused by the closure, which will scale as $U_B \sim (Et^3/2)(\kappa^2)A$, where κ is the curvature along the ridge. Assuming the curvature is dominated by the sharp curvature where the packet is held closed, $\kappa \sim 1/t$ and we find $U_B \sim Bx_0\ell_{ea}/t^2$. Some new contact is made when the packet is closed, meaning the adhesion energy has also changed ($U_{adh} \; - G_c x_0 \ell_{ea}$) and should also influence the problems lengthscale.

Minimizing the total energy determines the unknown length,

$$x_0 \sim \left(\frac{3Et\ell_{ea}^4}{B/t^2 - G_c}\right)^{1/4}. \tag{S8}$$

As adhesion becomes small with respect to Et, the result can be simplified

$$x_0 \sim \ell_{ea}. \tag{S9}$$

VIII. USER EXPERIENCE WITH ENCAPSULATED FOODS

The main text refers to several packaged food types which demonstrate kuttsukigami principles. In this section we detail the construction of edible gel packets and pasta and the response of a panel of 3 testers to the encapsulated foods. Our basic hypothesis is that the gelatin packets, constructed to encapsulate chocolate, will open easily and release the chocolate in response to the moisture of the mouth, whereas the pasta has a higher, non-responsive inter-sheet adhesion and will only open upon mechanical agitation (chewing).

Tests were conducted with thin gel sheets prepared from “Lime Jell-O” which was used as received from Horbacher Foods Inc. The gelatin was prepared by adding two 170 g packets to 0.591 L water at a temperature just below 100 °C with constant stirring. After the powder was completely dissolved in the water, the liquid was cast in various glassware. Occasionally glassware was lined with waxed paper to facilitate the peeling of thin films. Once cast, materials were put into a refrigerator and cooled for 24 h.

Once cooled, gelatin sheets were cut into various shapes for encapsulation experiments. Typically a sheet was cut to approximately 3×6 cm. Semi-sweet chocolate was added to the center of a sheet, which was then folded by hand around the encapsulate. Sheets which did not seal were not used, however, films of approximately 1 mm in thickness typically sealed completely. The packets were then given to the test panel who’s comments were recorded.

Highlights of the panel responses are given below:

Interviewer: “Describe how it was to eat.”

Tester #1: “Chocolate and jell-o does not taste good together”

Interviewer: “Did you first taste Jell-o?”

Tester #1: “Yes”

Interviewer: “Was the change in texture a surprise?”

Tester #1: "uh huh"

Interviewer: "Describe how it was to eat."
Tester #2: "That doesn't taste that bad."
Interviewer: "What flavours did you taste, and when?"
Tester #2: "The Jell-o came first, then the chocolate chips. Then a little more Jell-o after that."

Interviewer: "Describe how it was to eat."
Tester #3: "I think that it started out kind of sweet and then once I got into the chocolate the flavour changed because I was biting into the chocolate."

While the pandemic limited our ability to form a larger unbiased test panel, we feel that this preliminary study is reasonable verification of the basic stimuli responsiveness of this food based encapsulation system.

Additional tests were conducted with "Buitoni four cheese ravioli" which used as received from Hornbacher Foods Inc. The ravioli was prepared for consumption in boiling water for 5-10 minutes before being drained and allowed to cool.

Test panel response for the four cheese ravioli:

Tester #1: "I like this"
Tester #2: "This is ok, I guess"
Tester #3: "This isn't the same as spaghetti"

Upon additional probing from the interviewer, test subjects agreed that the flavour changed as the cheese was released from the noodle via chewing, verifying again the basic concept of food based encapsulation.

IX. CREASED VS RACQUET BENDS

To illustrate the value of racquet bends with respect to value added layers we created simple thin circuits by depositing copper traces on polyimide sheets. The sheets were 0.05

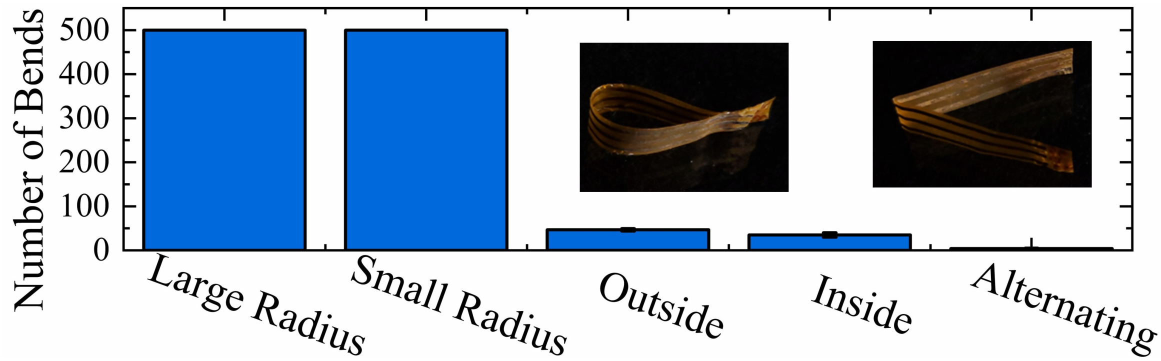

Fig. S5. Number of repeated bends until circuit failure for large or small radius racquet bends, creases with the wire on the outside or inside of the crease, creasing that alternated the side on which the wire was printed. Insets show creased and racquet bent circuits.

mm thick and were cut into strips 50 mm long. The trace was approximately with a 0.04 mm thick and 1 mm wide. Resistance could then be monitored as current is passed from one side of the sheet to the other. We then created a racquet loop or a fold orthogonally to the trace (insets in Fig. S5). In the bent or folded state resistance was measured, then the sheet was flattened. This process was repeated until resistivity was noted to be infinite.

Folding with the trace on the outside surface performed slightly better than folding with the trace on the inside of the circuit. Alternating folding (first inside, then outside) barely survived a few cycles. On the other hand both large (9.6 mm) and small (2.7 mm) radius racquet bends lasted 500 cycles without significant changes in resistivity.

X. RECONFIGURABLE, FOLDABLE LOGIC THROUGH KUTTSUKIGAMI

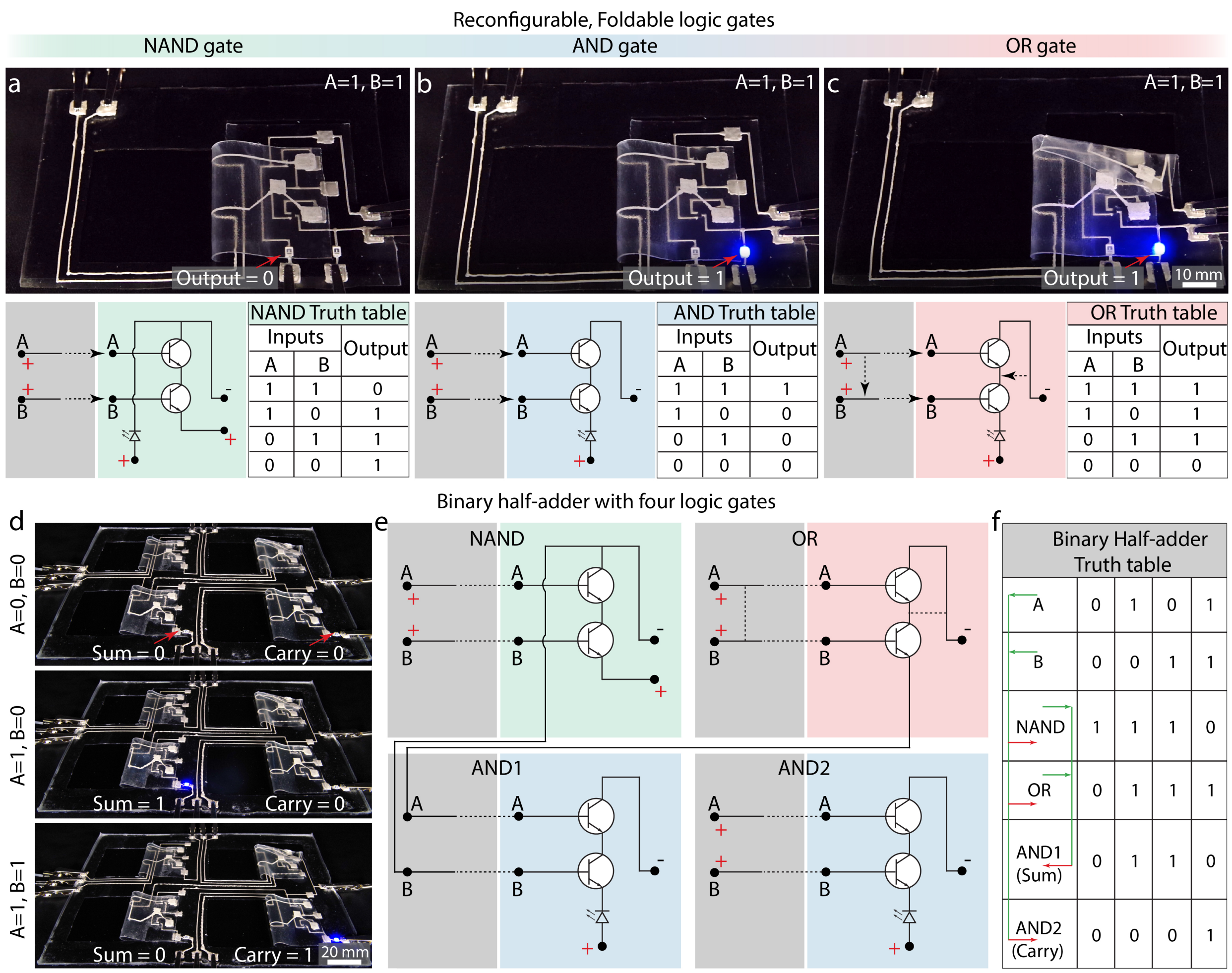

Fig. S6. Images of the reconfigurable circuit with electronic circuit layout and truth tables. Folds in the images show the physical step and dashed arrows in layouts indicate the electronic connections made for a. NAND gate b. AND gate, and c. OR gate. d. Implementation of binary half-adder circuit using four individual reconfigurable circuits – the LEDs at the bottom left and the bottom right circuits indicate the sum and carry outputs for the half-adder for different input A,B combinations. e. Electronic circuit equivalent of the half-adder. f. Half-adder truth table. Green and red arrows indicate inputs and outputs respectively.

[1] D. Maugis and M. Barquins, Fracture mechanics and the adherence of viscoelastic bodies, J. Phys. D: Appl. Phys. **11**, 1989 (1978).

[2] N. Glassmaker and C. Hui, Elastica solution for a nanotube formed by self-adhesion of a folded thin film, Journal of applied physics **96**, 3429 (2004).

[3] L. Mahadevan and J. B. Keller, The shape of a möbius band, Proc. R. Soc. Lond. A **440**, 149 (1993).

[4] E. Starostin and G. van der Heijden, The shape of a möbius strip, Nature Mater. **6**, 563 (2007).

[5] A. Lobkovski, S. Gentges, H. Li, D. Morse, and T. Witten, Scaling properties of stretching ridges in a crumpled elastic sheet, Science **270**, 1482 (1995).