

Rapidly Reconfigurable Inextensible Inflatables

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Abstract—Inextensible material inflatables, such as those made from heat-sealable fabric or Mylar, are lightweight, robust, and easily packed into compact spaces and self-deployed. They are commonly made by heat-pressing two sheets together to seal them along a desired curvilinear path. Upon inflation, the sheets wrinkle about the constraints imposed by the seals, giving rise to fascinating and functional shapes. In previous literature, once an inflatable has been manufactured, it can only attain a single inflated shape. Changing task demands of the real world motivate adaptive inflatables that are not fixed to a single deformation. Herein we present a method for creating inflatables that rapidly switch between multiple deformation modes. By patterning conductive fabric on the surface, we bestow the ability to locally form or remove seals during real-time operation. Namely, pulling a vacuum to supply compression force and locally Joule heating regions of the conductive fabric mimics the action of a heat press. Furthermore, we present a finite element model to aid in the analysis and inverse design of inextensible inflatables. Together, the reconfigurable inflatables and inverse model unlock a vast array of programmed inflated shapes with a single enclosed volume.

I. INTRODUCTION

Inflatables are transforming the field of robotics. They have been used to generate actuators mimicking the dexterity of human musculature [1], to create mechanical camouflage [2], lift objects many times their own mass [3], and replicate bio-inspired locomotion modes [4]. Elastomeric inflatables, including McKibben artificial muscles [5] and fiber reinforced actuators [6], until recently have been pre-programmed and fixed to a single deformation trajectory. The introduction of removable unidirectional laminae enabled manual reconfigurable deformation of volumetrically inflating hyperelastic bodies [7]; switchable-elasticity silicone-Fields Metal composite allowed for coarse deformation changes without manual intervention [8].

The class of inflatables made from high stretching modulus (quasi-inextensible) sheets, such as heat-sealable fabrics or Mylar, can often produce higher forces than their stretchable material counterparts. As such, they are compelling alternatives for making high force-density actuators or standalone deployable structures. Creating an inextensible inflatable generally involves heat-sealing two sheets together along a desired outer contour. Seals can be made inside of the outer contour as well. The geometry of these seals governs the equilibrium shape assumed by the inflatable. Although complex topologies may be elicited upon inflation [9], after manufacture, reported inextensible inflatables undergo only

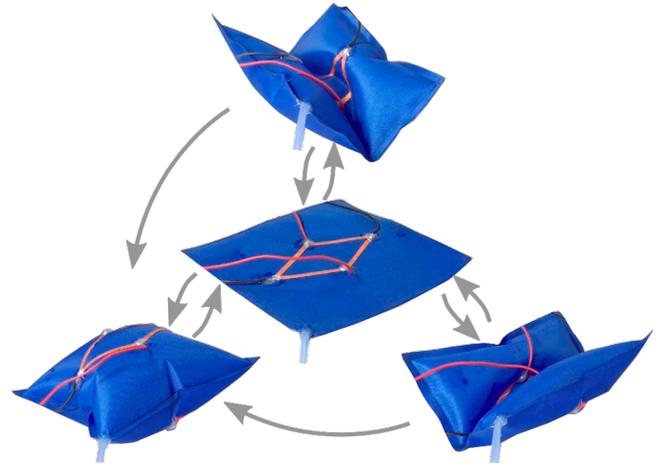


Fig. 1. Example R2I2: a single inflated volume rapidly transitions between different bending directions and out-of-plane expansion, enabled by reversible on-board seal formation. There are two diamond heaters, placed on top and bottom in opposite orientations, that form the seals needed for bi-directional bending. Arrows indicate possible state transitions.

a single deformation. Ou. et al. stacked multiple sealed volumes on top of one another and gave each its own pressure input to switch between bending modes [10]. However, their approach does not lend itself to switching between numerous drastically different deformations. The aforementioned technologies enabling reconfigurable deformations in stretchable elastomeric systems rely on surface strain limiting, and thus are not transferable to systems composed of sheets that barely stretch at operational pressures.

In this paper, we introduce rapidly reconfigurable inextensible inflatables (R2I2), systems that can autonomously change deformation, on the order of seconds, via localized on-board heat sealing operations. R2I2s are enabled by conductive fabric, which is laser-cut into the desired pattern and adhered to an inflatable's surface. When the fabric is Joule-heated and a vacuum applied, a seal forms that causes the inflatable to deform in a programmed way. The process is also reversible: by merely heating the fabric again, the seal weakens and the force of inflation separates the sheets, restoring the inflatable to its original state. We showcase the concept of an R2I2 by modulating between two bending modes and out-of-plane inflation with a single volume (Fig. 1 and Supplementary Video part 1).

The proposed reconfigurable inflatable paradigm can generate a diverse array of shapes with a single inflatable volume. Crucially, R2I2s can reconfigure both their extrinsic bending and intrinsic curvature (metric) as a function of the applied heater pattern. As a complement to the vast design

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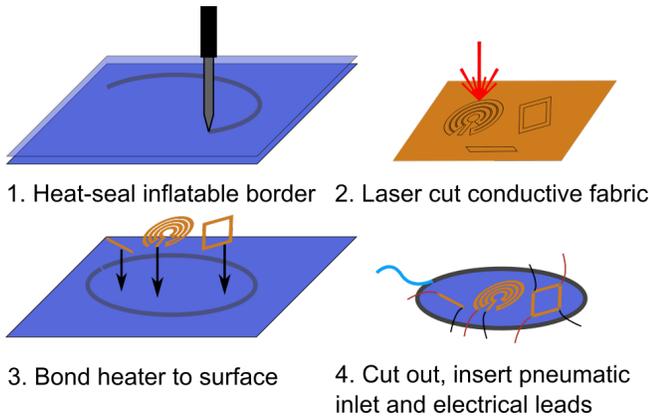


Fig. 2. Fabrication of R2I2s consists of four steps. First, two heat-sealable fabric sheets are bonded together in a curvilinear path with a heat press to define the contour of the inflatable. Second, conductive fabric is laser cut to desired pattern. Third, the heater is bonded atop the inflatable. Lastly, electrical lead wires are attached to the heater with liquid metal encapsulated in silicone, and the pneumatic inlet is inserted.

space opened up by R2I2s, we present a finite element analysis (FEA)-based inverse model that takes as input a desired array of inflated shapes and outputs conductive fabric patterns that will create seals necessary to modulate between those shapes—the first model of its kind for inflatable systems.

II. MATERIALS AND METHODS

The fabrication process for R2I2s is simple (Fig. 2). An initial inflatable geometry is created by heat sealing two pieces of fabric along a specific contour. R2I2s in this paper are made from thermoplastic-impregnated nylon fabric (200D, Seattle Fabrics). Uniaxial quasi-static tensile tests reveal the high Young’s modulus of the heat-sealable fabric at $E = 510$ MPa (Fig. 3A; this value is used later in the FEA model). Melting the thermoplastic of two sheets pressed together allows the resin to flow between the sheets, and when cooled, forms a contiguous seal (note with traditional heat sealing, melting only the thermoplastic and not the reinforcing fibers is desired for mechanical integrity). Conductive fabric (copper Taffeta from Less EMF Inc. with a sheet resistance of $0.05 \Omega/sq$) is laser-cut to a desired pattern (VLS 2.0 Lasers) and bonded to the surface of the fabric with Silpoxy (SmoothOn Inc.). Then, flexible wires or conductive threads are connected to the conductive fabric with eutectic Gallium Indium alloy encapsulated in silicone (Dragon Skin 10 Medium, SmoothOn Inc.), so that the fabric can be tethered to an external power supply for Joule heating. Lastly, a small slit is cut into the enclosed volume, and a pneumatic inlet is inserted.

Owing to its low bending modulus, the fabric heater is able to conform to the wrinkle field that occurs locally at seals, and thus has a negligible impact on the shape assumed by the inflatable. Similarly, the liquid metal and silicone interface between the wires and conductive fabric can deform in the presence of a wrinkle field without losing continuity or shearing off, as would a brittle connection.

A. Fabric heaters

Laser-cut fabric heaters are able to create a wide variety of internal seal geometries, and consequently, inflated shapes. Resistance R of a heater influences the power P it dissipates as heat by $P = I^2R$, where I is the current. Sudden changes in heater trace width augment R and thereby P , causing unwanted hot-spots. Fortunately, with laser-cutting it is easy to ensure smooth changes in width. Heaters with proximate traces, like the circular (Fig. 3B top left) and serpentine (bottom left) heaters, can entirely seal areas, though the contours of the sealed areas can be imprecise. We found that individually heating straight sections, as with the diamond shape (top right), results in more precise seals, likely due to less resistance variation across the span of the material. Heating straight sections requires a lead wire at each junction; multiple lead wires can be connected to create a quad-diamond (bottom right) network heater with dozens of possible seal patterns. Yet, adding more leads can become unwieldy as desired seal patterns scale in complexity. This fact highlights an inherent trade-off between burdening the R2I2 with additional electrical leads and desired seal precision.

B. Heat-sealable fabric

To determine the strength of seals formed along joined sheets of heat-sealable fabric, we conducted t-peel tests on an Instron Materials Tester at both room temperature (23°) and 180° (above the melting temperature of the thermoplastic resin). A peel test subjects the material to a shear condition highly similar to that which seals experience under inflation. The 23° average peel force from 20 mm to 80 mm is seven times that of the heated specimens’ (Fig. 3C).

An optical micrograph juxtaposes delaminated sections of a representative t-peel specimen at each temperature. Notable texturing on the room temperature specimen confirms that the mode of delamination fractures thermoplastic resin bond sites between the two sheets, consistent with the elevated force required to peel them. On the other hand, the heated specimen has a relatively unmarred surface, suggesting that bonds may be formed and removed without compromising the sealing mechanism. We take advantage of the ability to cleanly remove seals when transitioning between different shapes with a single inflatable.

We quantified how many times a seal could be formed and removed before it failed to create a stable bond under inflation (Fig. 3D). A test was conducted on a square inflatable of side length 60 mm with a single heater in the center of dimension 30×4 mm, consisting of cycles of 1) Joule heating while applying a -80 kPa vacuum to apply a local seal, 2) inflating to 7 kPa, and 3) Joule heating again to remove the seal. For more than 50 cycles no seal failed, testifying to the fact that the seal formation mechanism is highly reversible. Granted, the exact geometry of the inflatable, operating pressure, and heater pattern will influence results. This study was merely intended to provide an estimate of R2I2 reconfiguration repeatability with a representative seal geometry.

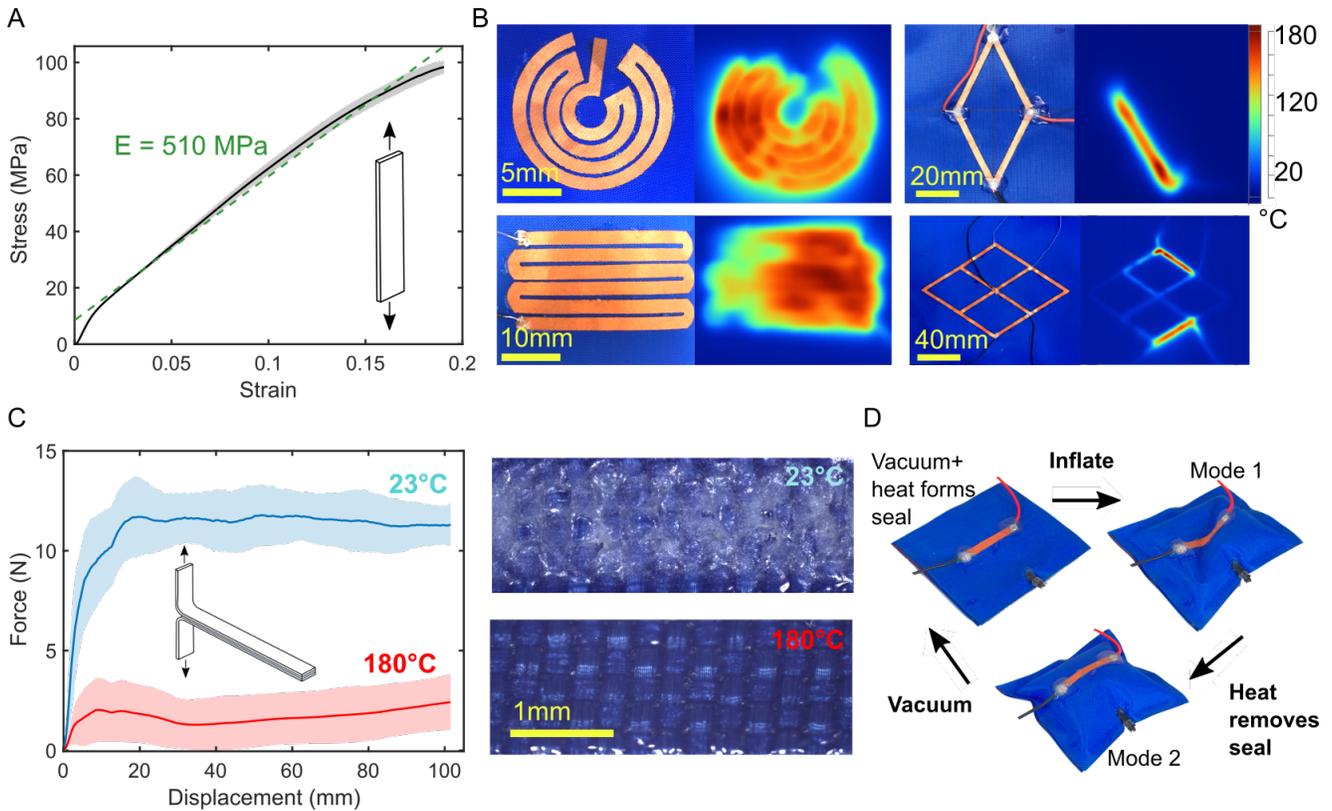


Fig. 3. A. Uniaxial tensile test of heat-sealable fabric. A solid mean line, calculated from five rectangular samples, is surrounded by a cloud indicating one standard deviation from the mean. The dashed line is a linear fit. B. Laser cut heater patterns at different scales. More precise or complex seals require additional electrical leads. C. Standard t-peel test at room temperature and elevated past the melting point of the thermoplastic resin in the fabric, revealing a seven-fold difference in peel strength. Clouds indicate one standard deviation from the mean for five samples. Inset is an optical micrograph of a representative specimen exhibiting delamination. D. Snapshots from the cyclic reconfigurability test, wherein a single volume modulated between two different deformation modes more than 50 times.

III. APPLICATIONS

A salient application of R2I2s is to make dexterous and reconfigurable actuators. We made an R2I2 capable of rapidly twisting in one of two directions or expanding out-of-plane (Fig. 4A). A long, continuous fabric heater adhered to the R2I2's surface exploits varying trace width to proportionally dissipate power in areas of higher resistance (thinner sections). The same heater pattern is mirrored on the backside, enabling on-demand seals at $\pm 45^\circ$ distributed along the length of the R2I2. Transitions from out-of-plane inflation to handed twisting took 20 sec, limited mainly by the cool-down period required for a seal to set. Transitions from handed twisting to out-of-plane expansion took only 5 sec (when 7 W was applied).

Some seal patterns can be difficult to form with the proposed fabric heaters and vacuum mechanism, particularly if the pattern requires many electrodes, or extremely high-resolution and close-proximity features within 1 mm. Yet such patterns are desirable because they distort the inflatables' metric and can doubly-curve its midplane section. Fortunately, there is a way to realize R2I2s that modulate between said deformations: fabric heaters can be utilized like valves that selectively open or close sections of a volume, directing air flow between regions containing pre-

patterned (with a heat press) seals. For instance, we made a single volume capable of morphing between a saddle and an twisted anti-cone (Fig. 4B). The pre-patterned geometries were adapted from work by Siéffert et al. [9]. The R2I2 was fabricated by stacking the two pre-patterned inflatables, gluing them along their edges, and connecting them with a tube to render a single volume. Rectangular fabric heaters adhered on either side served as valves to selectively direct air flow. Transitions between shapes took 35 sec (at 1 W) because we needed to seal one valve when pulling a vacuum and then open the other valve when inflating.

Combining valving, pre-patterned seals, and on-demand seal formation further expands the R2I2 design space. As a demonstration, we fabricated an artificial flower (Fig. 4C). The flower has two diamond heaters circumferentially about its "pedals", one central spiral heater, and a valve at its "stalk" regulating air flow into a pre-patterned seal section. The flower can morph between several distinct states. Shown are two bloom phases, the left in which the stalk is straight and a portion of the pedals are unfurled. This is the default state, without making seals or releasing the valve. The right bloom is toggled by sealing the central spiral heater and peripheral diamond heaters, and opening the valve to the pre-patterned geometry within the stalk. The result is a non-

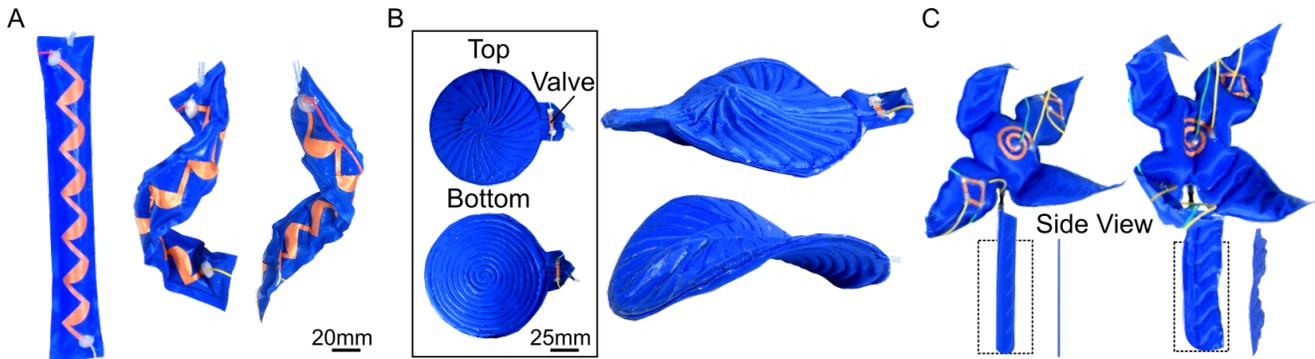


Fig. 4. A. Long R2I2 rapidly changing twisting direction, made possible by a single heater with only two electrodes on either side. B. Fabric heaters used as switches to modulate between complex pre-patterned topographies of drastically different Gauss curvatures. Design adapted from [9]. C. Combining fabric heater valves to switch between pre-patterned deformations, and creating on-demand seal geometries with more complex fabric heaters, we made an artificial flower capable of distinct “blooms.”

Euclidean undulation along the stem reminiscent of frustrated elastic sheets [11] and all pedals curling inward about a sunken center. Due to the multiple heaters and distinct pressurization steps, transitions between blooms can take about 1 min.

IV. PROGRAMMING R2I2 DEFORMATION

With hardware that enables rapid reconfiguration between multiple deformations, naturally the question arises whether we can specify a set of shapes and use a model to generate the heater pattern(s) required to realize those shapes. The equilibrium shape of a certain class of simpler, often axisymmetric inextensible inflatables is well-studied, and can be described with analytical solutions [12]. Numerical inverse design has also been demonstrated for more complex 3D inflatables, but has two significant limitations: it does not provide a user with the option to inspect physically meaningful quantities post-simulation, and it has trouble processing planar surfaces and sharper edges of deformed geometries [13].

Seeking to predict the shape of R2I2s, we conducted non-linear quasi-static analysis in ABAQUS/Explicit. Established FEA software is attractive because it gives us: 1) a high resolution map of surface wrinkles for qualitative comparison with hardware, 2) insight into the governing physics of the inflatable system, and 3) the ability to simulate highly complex designs (which we will touch on in a later section) that existing models cannot accommodate.

A. FEA model

The FEA model was formulated with material stiffness data from Fig. 2A, using a linear elastic continuum model with Young’s modulus of 510 MPa and Poisson’s ratio of 0.38. The R2I2 in particular was modeled as an enclosed thin shell with its seals constrained using *TIE constraints. Four-node doubly curved shell elements (S4R) with an hourglass control were used to define the R2I2 contours. To improve the computational efficiency and limit numerical oscillations during dynamic simulations, mass scaling and Rayleigh damping were performed. To ensure quasi-static conditions

in the simulations, we verified that the kinetic energy of the model was low in comparison to the strain energy. Moreover, because dynamic simulations in ABAQUS are conditionally stable, we verified that the kinetic energy was very small relative to the internal energy at all steps.

To verify the FEA model in a simplified design space, we simulated three rectangular bending R2I2s—inspired by Ou et al. [10]—of dimensions ($l \times w$) 120x60 mm. Each actuator has a diamond-shaped fabric heater located at its geometric midpoint, with spanning segments axb of 14x28 mm, 28x28 mm, and 42x28 mm, respectively. The diamond shape makes a seal that induces extrinsic bending, upon inflation, through a buckling instability. The magnitude of bending is contingent on the diamond’s aspect ratio (a/b). All actuators were inflated to 15 kPa in experiment; an equal magnitude distributed pressure load was applied in simulation.

Qualitatively, there is good agreement between wrinkles in simulation and experiment, testifying to the high fidelity of the simulation (Fig. 5A). The percentage error between actual (θ) and simulated (θ_s) bending angles for designs 1, 2 and 3 is 3%, 10% and 8%, respectively, with simulation consistently overestimating the angle. We suspect that a seal formed by the conductive fabric heater might be slightly thinner than the geometry of the heater due to convective dissipation of heat around the edges, and that this decreased thickness was manifested systemically through smaller bend angles in hardware. A simulated parametric study in which we held a/b of a design constant and swept over a range of seal thicknesses t , recording θ_s (Fig. 5B), elucidates how θ_s is highly sensitive to t . Even a 1 mm change in t can result in an 8° change in θ_s (per design 2, going from 2 to 3 mm). Based on the findings of this study, we verified that subtleties in seal geometry can radically change apparent deformation of an R2I2. Some level of discrepancy between projected and actual deformations must therefore be expected due to the variability of a formed seal’s thickness.

B. Inverse design for reconfigurable deformations

Having validated the forward FEA model, we proceeded to conduct inverse design. We reiterate that the scope of this

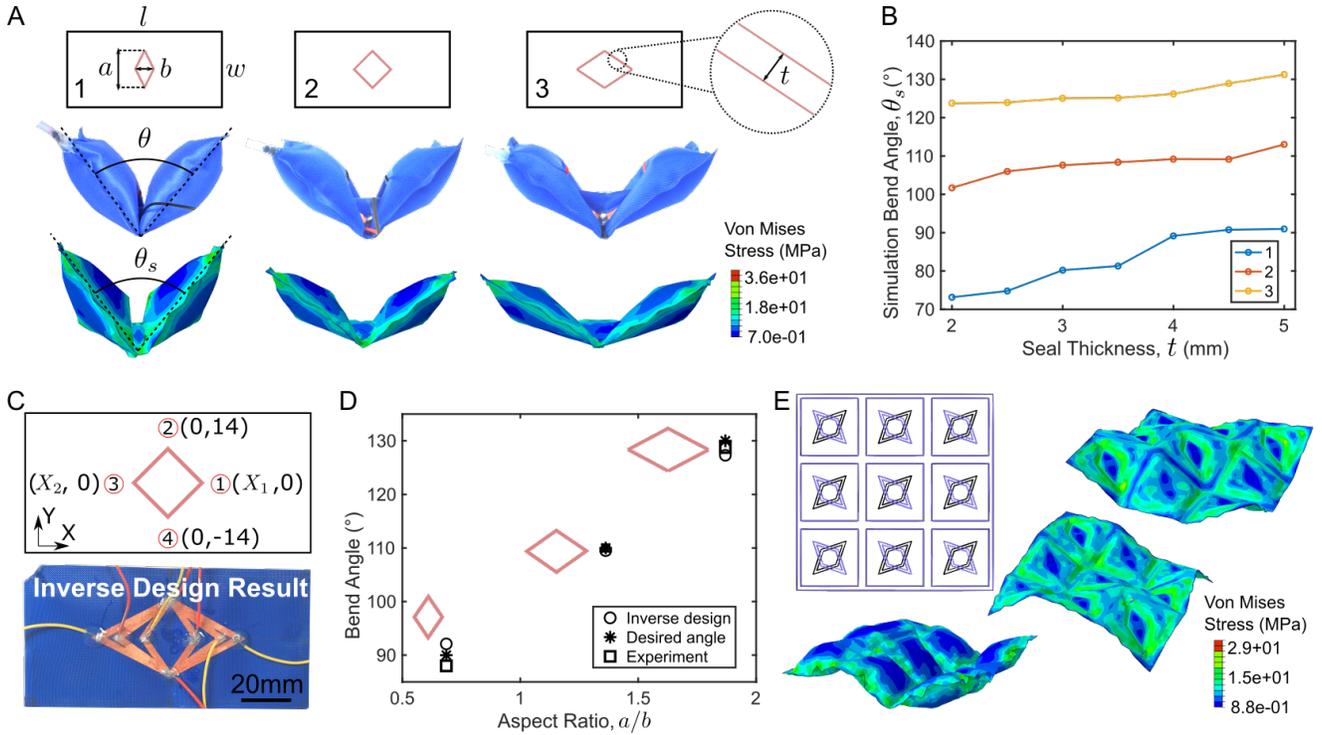


Fig. 5. A. Top row: schematics of bending R2I2s with three different aspect ratio diamond heaters, showing relevant dimensions. Bottom rows: validation of ABAQUS simulation with experiment, with different bending angles as a result of seal geometry. B. Results of the parametric study. A plot of bending angle versus seal thickness, holding a design aspect ratio constant. Legend refers to designs labeled in A. C. Top: schematic showing optimization variables for the inverse design experiment in the context of the pre-defined R2I2 contours. Bottom: model-generated heater pattern transferred to an R2I2. D. Bend angle versus heater aspect ratio. Points are plotted for the desired bending angle, the angle attained by inverse design for the pattern it generated, and the experiment. Schematic insets illustrate which of the three diamond heaters was used to reach that angle in the experiment. E. Simulation provides insight into designs that may be difficult to prototype, such as this 3×3 grid. The schematic in purple and black shows possible seal geometry. Images from FEA show three distinct topographies achieved by sealing all patterns along a unique direction.

portion of the present paper focuses on a simplified subset of the design space—namely bending actuators via a buckling instability—to exhibit the R2I2 concept. For the inverse design problem, one seeks to find the required geometry of a heater that elicits several desired bend angles from an R2I2. Posed as a shape optimization problem, this objective is:

$$\begin{aligned} \min_{X_i} : & (\theta_s - \theta_s^*)^2 \\ \text{s.t.} : & L_{min,i} \leq X_i \leq U_{max,i} \end{aligned} \quad (1)$$

Here, θ_s is the bend angle from the FEA model and θ_s^* is the desired bend angle. The design variables X_i 's are the X-coordinates of the points (Fig. 5C top). $L_{min,i}$ and $U_{max,i}$ are the lower and upper bounds (enforcing realistic heater dimensions), respectively. t is kept constant at 3 mm. To ensure symmetry of a heater, we imposed an equality constraint on the design variables as well.

The optimization was performed in a Python IDE with ABAQUS scripting for the FEA model, post-processing, and result validation. Due to the stationary nature of the bend angle with small variations in the design variables, standard gradient descent algorithms are ill-suited to solving this inverse design problem. We thus employed basin-hopping [14], a stochastic global optimization algorithm.

We implemented our inverse model to generate the heater

pattern for an R2I2 to achieve three bend angles: 90° , 110° and 130° . The model output three diamond shapes that when united become a tri-diamond (Fig. 5C bottom). We proceeded to Joule-heat and thereby seal each diamond individually and then inflate the R2I2 to measure the bending angle. Despite the overlap in heater trace edges for the $a/b = 1.5$ and 2 cases, the experimental bend angles are very similar to those projected by the inverse design and the original desired input, boasting less than 4% error at each angle (Fig. 5D). Strong agreement between the output heater pattern from the inverse model and the physically realized R2I2 attest to its efficacy in this simplified design space; furthermore, the accurate results bode well for inverse design of more sophisticated R2I2s.

C. Exploring novel R2I2s in simulation

To explore the design space for R2I2s even further, we simulated a 3×3 square grid R2I2 (Fig. 5E, Supplementary Video part 2). We specifically show the R2I2 taking on three configurations, by activating all of the constraints in one orientation (Fig. 5E schematic shows possible configurations in purple and black). The high-fidelity FEA model can capture the wrinkling and buckling of the system, even in the context of an intricate energy landscape arising from stretching and bending energy competitions between neighboring unit cells. Rifts and valleys along the simulated R2I2's surface exhibit

a spectrum of curvatures. For R2I2s of size and complexity rivaling this simulated 3x3 grid, it is pragmatic to simulate such cases to validate feasibility before prototyping. As R2I2 manufacturing methods and hardware improve, we expect to be able to realize these large-scale systems: reconfigurable topography with unprecedented control of the metric.

V. CONCLUSION

With a simple concept we have presented herein—leveraging low-profile conductive fabric to locally create or remove seals between heat-sealable sheets—we have shown a glimpse of the rich spectrum of inflated shapes attainable with a single volume. We demonstrated how R2I2s can independently modulate both extrinsic bending and intrinsic curvature along their surface. While the R2I2 concept holds significant promise in the robotics space, it is transferable to other domains: in surgery, R2I2s with a lower sealing point could be used for shape-morphing stents, in rehabilitation, they could be applied to adaptive exosuits, and in architecture, R2I2s could be the foundation of smart structures.

However, R2I2s need further characterization and improvements before they are widely adopted. For the same types of deformations, R2I2s may surpass the speed of reconfigurations in existing elastomeric systems [7], [8], but a primary bottleneck for R2I2 reconfiguration speed is cool-down time while seals form. Though the demonstrations herein span a range of cool-down times based on heater complexity—from 5 sec to minutes—implementing a heat transfer system or thermally isolating sections of heater from the inflatables’ surface would certainly improve their response times.

Additionally, exploring options for a heating material that produces precise seals would help to seal distinct features <1 mm apart with fewer electrodes. Furthermore, it would be prudent to electrically insulate the exposed heaters on the surface of the actuator such that R2I2s could more readily interact with an environment without the possibility of shorting.

On the modeling side, our inverse pipeline finds fabric heater designs to realize desired bending angles; in the future the objective could be augmented to more generic measures like mid-plane Gauss curvature or surface normals. Overall, with this work we have taken a step toward the next generation of context-sensitive robotic systems by introducing a reconfigurable actuator paradigm.

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REFERENCES

- [1] F. Connolly, C. J. Walsh, and K. Bertoldi, “Automatic design of fiber-reinforced soft actuators for trajectory matching,” *Proceedings of the National Academy of Sciences*, vol. 114, no. 1, pp. 51–56, 2017.
- [2] J. Pikul, S. Li, H. Bai, R. Hanlon, I. Cohen, and R. Shepherd, “Stretchable surfaces with programmable 3d texture morphing for synthetic camouflaging skins,” *Science*, vol. 358, no. 6360, pp. 210–214, 2017.
- [3] E. Siéfert, E. Reyssat, J. Bico, and B. Roman, “Programming curvilinear paths of flat inflatables,” *Proceedings of the National Academy of Sciences*, vol. 116, no. 34, pp. 16 692—16 696, 2019.
- [4] G. Singh and G. Krishnan, “Designing Fiber-Reinforced Soft Actuators for Planar Curvilinear Shape Matching,” *Soft Robotics*, vol. 7, no. 1, pp. 109–121, 2020.
- [5] C. P. Chou and B. Hannaford, “Measurement and modeling of McKibben pneumatic artificial muscles,” *IEEE Transactions on Robotics and Automation*, vol. 12, no. 1, pp. 90–102, 1996.
- [6] P. Polygerinos, Z. Wang, J. T. B. Overvelde, K. C. Galloway, R. J. Wood, K. Bertoldi, and C. J. Walsh, “Modeling of Soft Fiber-Reinforced Bending Actuators,” *IEEE Transactions on Robotics*, vol. 31, no. 3, pp. 778–789, 2015.
- [7] S. Y. Kim, R. Baines, J. Booth, N. Vasios, K. Bertoldi, and R. Kramer-Bottiglio, “Reconfigurable soft body trajectories using unidirectionally stretchable composite laminae,” *Nature communications*, vol. 10, no. 1, pp. 1–8, 2019.
- [8] T. Buckner, M. Yuen, S. Y. Kim, and R. Kramer-Bottiglio, “Enhanced variable stiffness and variable stretchability enabled by phase-changing particulate additives,” *Advanced Functional Materials*, vol. 29, 2019.
- [9] E. Siéfert and M. Warner, “Inflationary routes to Gaussian curved topography,” *Proceedings of the Royal Society A: Mathematical, Physical and Engineering Sciences*, vol. 476, no. 2240, p. 20200047, 2020.
- [10] J. Ou, M. Skouras, N. Vlavianos, F. Heibeck, C.-Y. Cheng, J. Peters, and H. Ishii, “aeroMorph - Heat-sealing Inflatable Shape-change Materials for Interaction Design,” in *Proceedings of the 29th Annual Symposium on User Interface Software and Technology - UIST '16*. Tokyo, Japan: ACM Press, 2016, pp. 121–132.
- [11] E. Efratia, E. Sharonam, and R. v. Kupferman, “Elastic theory of unconstrained non-euclidean plates,” *Journal of the Mechanics and Physics of Solids*, vol. 57, pp. 762–775, 2009.
- [12] E. Siéfert, J. Bico, E. Reyssat, and B. Roman, “Geometry and mechanics of inextensible curvilinear balloons,” *Journal of the Mechanics and Physics of Solids*, vol. 143, p. 104068, 2020.
- [13] M. Skouras, B. Thomaszewski, P. Kaufmann, A. Garg, B. Bickel, E. Grinspun, and M. Gross, “Designing inflatable structures,” *ACM Transactions on Graphics*, vol. 33, no. 4, pp. 1–10, 2014.
- [14] D. J. Wales and J. P. Doye, “Global optimization by basin-hopping and the lowest energy structures of lennard-jones clusters containing up to 110 atoms,” *The Journal of Physical Chemistry A*, vol. 101, no. 28, pp. 5111–5116, 1997.