Bistable Electroactive Polymers for Refreshable Tactile Displays

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ABSTRACT: Bistable electroactive polymers (BSEP) combine shape memory with large-strain actuation at the rubbery state to achieve rigid-to-rigid actuation. The stiffness of the BSEP is tunable via glass transition or phase changing. The reversible melting-crystallization of the polymer chains in the phase changing BSEP contributes to the stiffness change within a narrow temperature range. A modulus change of more than 1000 folds can be achieved within 3 °C. Additionally, large actuation strains rivaling those of VHB acrylic elastomers can be obtained at the rubbery state. Explorations regarding potential applications of this material have been focused on tactile displays. In one design, Joule heating of a serpentine-shaped compliant electrode coated on a BSEP film, coupled with a pneumatic pressure source has been employed to raise diaphragm dots with 1.5 mm base diameter to heights up to 0.7 mm. The resulting Braille electronic readers could thus be actuated with low voltages.

Keywords: Bistable electroactive polymer, phase-changing, dielectric elastomer, rigid-to-rubbery transition, shape memory, rigid-to-rigid actuation.

INTRODUCTION: Among all the electroactive polymers (EAPs) investigated, dielectric elastomers (DEs) stands out by exhibiting a unique combination of properties, including large strains, fast response, high energy densities, mechanical compliancy, and low cost. For example, when sandwiched with compliant electrodes, the acrylic elastomer, 3M VHB 4910, can reach 100% or greater area expansion with a maximum elastic energy density of 3.4 J/g under high driving voltages. However, the extraordinarily large actuation strain is obtained at the sacrifice of materials' mechanical stiffness which in turn hinders the applications of the DE actuators in adaptive structures. In addition, the high voltages are required to maintain the large deformation, leading to significant energy consumption, material fatigue, and a reduced lifetime.

Bistable electroactive polymers (BSEPs) can amalgamate the shape memory property with dielectric elastomers to obtain rigid-to-rigid actuation⁵⁻⁶ owing to their temperature dependent stiffness. The BSEP is rigid below its transition temperature, while above this temperature, it behaves like a dielectric elastomer and exhibits large electrically-induced actuation strain and high dielectric field strength. The deformation can be locked by cooling the BSEP below its transition temperature when the polymer turns stiff again. The transition temperature of BSEP is also tunable.

Fast response and low energy consumption can be achieved with narrowed transition temperature range. These features render the BSEP a promising smart material candidate for the fabrication of Braille electronic readers.

Braille has been the media to educate blind children literacy. There are 1.3 million legally blind individuals in the Unites States and among them 55,000 are children.^{7,8}. Despite the advancements

in smartphones and tablets, a tactile version of comparable compactness and low cost serving the vision impaired population is not available on the market. The technical challenge has been the creation of a suitable actuation mechanism that produces larger deformation with a sufficiently high blocking force.⁹

Here, we report a refreshable tactile display with Braille standard resolution that combines the specific actuation feature of BSEP with the Joule heating of a serpentine-patterned carbon nanotube (CNT) electrode. The system can be heated up to 70 °C under 30 V voltage supply in less than 1 s. The blocking force is 51 grams after the BSEP film is cooled down. The out-of-plane displacement induced by pneumatic pump is 0.5 mm. The demonstrated 3 x 2 pneumatic tactile device can be operated for over 100,000 cycles.

Mechanical Properties of the BSEP Polymer.

Utilizing techniques derived in our previous research, ^{10,11} modified BSEP films comprising stearyl acrylate (SA) and a urethane diacrylate oligomer (UDA) were synthesized. The resulting SA-UDA copolymers were characterized using a dynamic mechanical analyzer (DMA) at a temperature ramping rate of 2 °C/min from 25 to 55 °C at the mechanical loading frequency of 1 Hz. Fig. 1A shows that below the transition temperature, the storage modulus is on the order of 50-200 MPa. This is due to the crystalline aggregates of stearyl acrylate (SA) acting as hard segments. Increasing the temperature above the transition temperature melts the crystalline aggregates and causes a dramatic decline in modulus. The rigid-to-rubbery transition can be completed within 3 °C. Once reaching the rubbery state, further increasing temperature does not affect the storage modulus, which is beneficial for obtaining stable electrical or pneumatic actuation with large deformation. Varying the ratios of SA: UDA could tune the transition temperature of the BSEP; higher SA: UDA ratios resulted in larger storage moduli in the rigid state as the percent crystallinity increased. A modulus change of more than 1000 folds can be achieved within 3 °C. The stressstrain curves of the BSEPs were obtained at 60 °C to maintain their rubbery states. Samples with dimensions of 3 mm width, 6 mm length and 170 µm thickness were tested at a stretch rate of 3.33 mm/s. The tensile strengths are 0.26 MPa and 2.64 MPa for BS80 and BS60 along with increasing tear strain from 191% to 234%, respectively. Results in Fig. 2B and Table 1 show that a larger amount of UDA contributes to higher toughness, but diminishes the modulus at the rigid state. Further, the copolymers softens at lower temperature as the UDA content is increased.

Table 1. Mechanical properties of SA-UDA Copolymers at 60 °C.

Sample	Elongation at break (%)	Tensile strength (MPa)
BS50	205	2.80
BS60	234	2.64
BS70	232	1.87
BS80	191	0.26

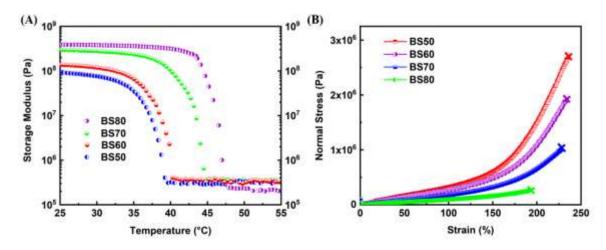


Fig. 2. Mechanical properties of SA-UDA copolymers. (A) Evolution of storage modulus measured by DMA with temperature ramping from 25 °C to 55 °C at 2 °C/min. (B) Stress-strain curves at 60 °C.

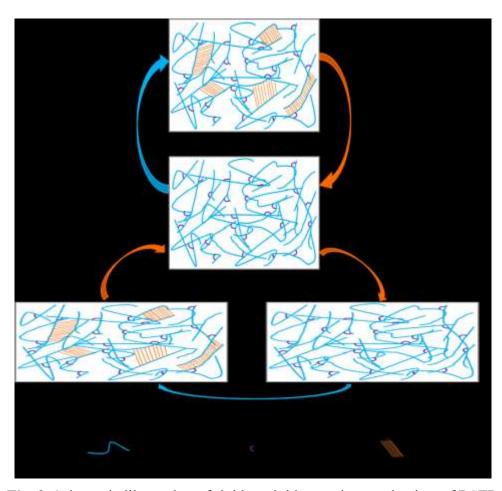


Fig. 3. Schematic illustration of rigid-to-rigid actuation mechanism of BSEP.

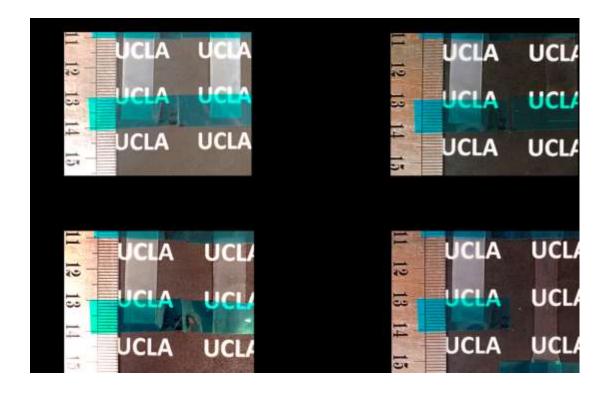


Fig. 4. Demonstration of shape memory effect of BSEP. (*left*: reference sample without any treatment; *right*: testing sample going through heating-stretching-cooling cycle)

Shape Memory Property. The variable stiffness and actuation of BSEP are illustrated in Fig. 3. The BSEP comprises crystalline aggregates of the long alkyl side chains in a crosslinked polymer matrix, which makes the polymer film translucent. By melting the crystalline aggregates, the BSEP turns clears and rubbery. This deformation can be preserved by cooling down the material due to re-crystallization of the long alkyl chains. Reheating the polymer above T_m recovers its original shape. Fig. 4 shows the shape memory property of BS60. Two BS60 strips of 1 cm width was marked with two parallel blue tapes gapping 2 cm (Fig. 4A). The left strip serves as reference, while the right one is heated to 70 °C when it turned from translucent to transparent (Fig. 4B). The heated strip was stretched to a 100% strain, and then cooled down to ambient temperature. The deformation was locked. The strain fixity rate was close to 100% after removing the load (Fig. 4C). Full recovery of the original shape and translucency was obtained by a reheating-cooling cycle (Fig. 4D).

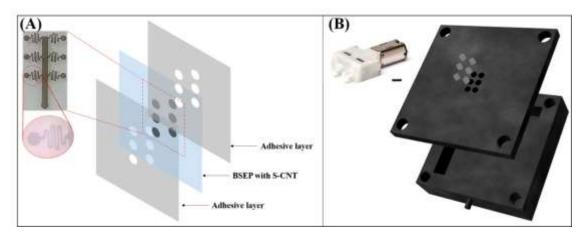


Fig. 5. Details of a Braille cell architecture. (A) BSEP active film sandwiched with two adhesive layers. (B) Compact tactile display with 3 X 2 pixels array in the pneumatic system.

Device Fabrication and Actuation Performance. The one-cell tactile display was comprised of two major parts: BSEP active film and pneumatic system. Carbon nanotube solution was prepared following steps in our previous research.^{9, 15-16} The Joule heating serpentine CNT (S-CNT) electrode was created by spraying coating on glass substrate and then laser engraving. The functional BSEP film was sandwiched by two double-sided KaptonTM tape. The pneumatic system consists of a pneumatic chamber and a miniature pump. The pneumatic chamber and chamber cover were made by 3D printing and were sealed to ensure an airtight environment. The design can be seen in Fig. 5B. The chamber size is 40 mm x 40 mm x 10 mm. The tactile cell contains 6 tactile pixels (taxels) with layout following the Braille standard; each taxel has 1.5 mm diameter with 2.5 mm center-to-center distance between adjacent taxels. The BSEP film can be locally heated within 1 second to 70°C with a 30V voltage supply applied to the serpentine-shaped CNT electrode heater. Data supporting this heating rate can be seen in Fig. 6A. Infrared images of the S-CNT Joule heating electrodes in Fig. 6B showed "U" "C" "L" "A" in Braille characters. The Joule heating film generated uniform heat across the surface without cross talk among dots. After the BSEP film is softened, a low pneumatic pressure 160 mmHg was applied which induce a large out of plane deformation of the BSEP film. The height of the diaphragm dot was raised by? mm. Removing thermal stimuli, the BSEP film became rigid, and the out-of-plane displacement was preserved. The raised height matches the required displacement for Braille displays¹⁰.

To examine the electrical continuity of the S-CNT electrode, the resistance of the electrode was measured as a function of biaxial deformation, and the results are shown in Fig. 6A, there shows an increase of resistance by ?% when the area expansion reaches 200%. Thus the electrode can effectively heat the BSEP film at its maximum deformation. The resistance and Joule heating characteristics remained stable for over 100,000 cycles at a frequency of 0.8 Hz as shown in Fig. 6B.

The blocking force of the taxels was tuned by using BSEP films of different thicknesses. The films were all actuated to a raised height of 0.5 mm. An incrementally increased force was applied to press on the tip of the raised dots until the dots became flattened. As Fig. 7 shows, the measured blocking forces, the force required to press a raised dot of 0.5 mm height to flat, are 10, 51, and 95 grams for the BSEP films with thicknesses of 40 μ m, 90 μ m and 170 μ m, respectively. Thicker

BSEP films provide higher blocking force.

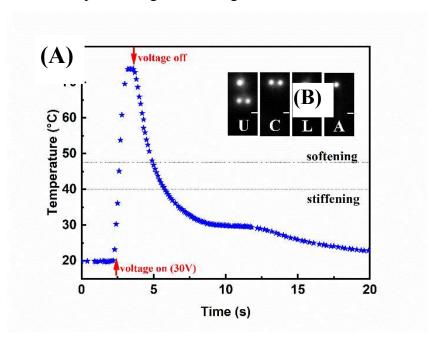


Fig. 6. Temperature profiles of S-CNT electrode under 30V (A). The "Softening" line indicate the temperature above which the polymer is soft, and the "Stiffening" line the temperature below which the polymer is stiff. (B) Demonstration by infrared images of the corresponding S-CNT Joule heating electrode showed "U" "C" "L" "A" in Braille characters. The scale bars are 2 mm.

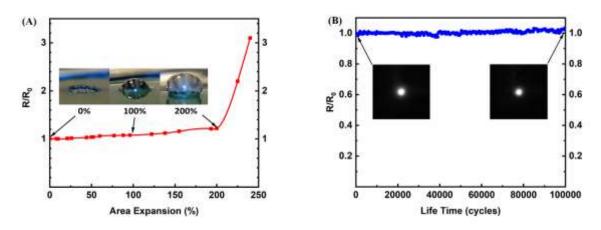


Fig. 7. (A) Normalized resistance of one S-CNT electrode under different area expansions. (B) Lifetime test on an S-CNT Joule heating electrode with a 100% area expansion deforming and releasing cycle at a frequency of 0.8 Hz for over 100,000 cycles.

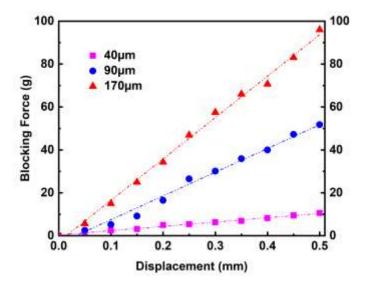


Fig. 7. Forces applied to Braille dots originally raised by 0.5 mm and the displacements of the dot from the original raised height. Thickness of the BSEP films are specified.

CONCLUSIONS

BSEP's sharp transition temperature range allows rapid transition between its rigid and rubbery states by Joule heating. The transition temperature ranges are tunable by the polymer's composition. Above the transition temperature, the BSEP is in the rubbery state and exhibits large-strain deformation ability. This deformation is locked when it is cooled below the transition temperature. These unique properties make BSEP a suitable material for tactile display applications, such as Braille electronic readers. The tactile device we have developed can operate at a voltage supply as low as 30V. The serpentine-patterned carbon nanotube electrode promises uniform and high heating rates up to 200% strain. The Braille dots can be operated repeatedly for over 100,000 cycles.

Acknowledgements:

The work was supported by the National Science Foundation (award number 1700829).

References

- 1. Pelrine, R.; Kornbluh, R.; Pei, Q.; Joseph, J., High-speed electrically actuated elastomers with strain greater than 100%. *Science*, **2000**, *287*(5454), 836-839.
- 2. Brochu, P.; Pei, Q., Advances in dielectric elastomers for actuators and artificial muscles. *Macromol. Rapid Commun.* **2010**, *31* (1), 10-36.
- 3. Shintake, J.; Cacucciolo, V.; Shea, H.; Floreano, D., Soft biomimetic fish robot made of dielectric elastomer actuators. *Soft robot.*, **2018**, *5*(4), 466-474.

- 4. Carpi, F., Anderson, I., Bauer, S., Frediani, G., Gallone, G., Gei, M., Graaf, C., Jean-Mistral, C., Kaal, W., Kofod, G. and Kollosche, M., Standards for dielectric elastomer transducers. *Smart Mater. and Struct.*, **2015**, *24* (10), 105025.
- 5. Ren, Z.; Hu, W.; Liu, C.; Li, S.; Niu, X.; Pei, Q., Phase-Changing Bistable Electroactive Polymer Exhibiting Sharp Rigid-to-Rubbery Transition. *Macromolecules* **2015**, *49* (1), 134-140.
- 6. Yu, Z.; Yuan, W.; Brochu, P.; Chen, B.; Liu, Z.; Pei, Q., Large-strain, rigid-to-rigid deformation of bistable electroactive polymers. *Appl. Phys. Lett.* **2009**, *95* (19), 192904.
- 7. Besse, N.; Rosset, S.; Zarate, J. J.; Shea, H., Flexible active skin: large reconfigurable arrays of individually addressed shape memory polymer actuators. *Adv. Mater. Technol.* **2017**, *2*(10), 1700102.
- 8. National Federation of the Blind: Programs and Policy Research, "Statistical Facts about Blindness in the United States (2011)", https://nfb.org/factsaboutblindnessintheus, retrieved 2013-10-08.
- 9. National Eye Institute, "Blindness, Statistics and Data [NEI]." Accessed April 12, 2013. http://www.nei.nih.gov/eyedata/blind.asp. Retrieved 2013-10-08.
- 10. Qiu, Y.; Lu, Z.; Pei, Q., Refreshable Tactile Display Based on a Bistable Electroactive Polymer and a Stretchable Serpentine Joule Heating Electrode. *ACS Appl. Mater. Inter.* **2018**, *10* (29), 24807-24815.
- 11. Qiu, Y.; Ren, Z.; Hu, W.; Liu, C.; Pei, Q., Bistable electroactive polymer with sharp rigid-to-rubbery phase transition. *In Electroactive Polymer Actuators and Devices (EAPAD)* **2016,** *9798*, 97981U.
- 12. Kagami, Y.; Gong, J.P.; Osada, Y., "Shape Memory Behaviors of Crosslinked Copolymers Containing Stearyl Acrylate." *Macromol. Rapid Commun.* **1996,** 17(8), 539-543.
- 13. Matsuda, A., Sato, J.I., Yasunaga, H. and Osada, Y., Order-disorder transition of a hydrogel containing an n-alkyl acrylate. *Macromolecules* **1994**, *27*(26), 7695-7698.
- 14. Plate, N.A.; Shibaev, V.P.; Petrukhin, B.S.; Zubov, Y.A. and Kargin, V.A., Structure of crystalline polymers with unbranched long side chains. *J. Polym. Sci. Pol. Chem.* **1971**, *9*(8), 2291-2298.
- 15. Yuan, W.; Hu, L.B.; Yu, Z.B.; Lam, T.; Biggs, J.; Ha, S.M.; Xi, D.J.; Chen, B.; Senesky, M.K.; Grüner, G. and Pei, Q., Fault-tolerant dielectric elastomer actuators using single-walled carbon nanotube electrodes. *Adv. Mater.* **2008**, *20*(3), 621-625.
- 16. Yuan, W.; Brochu, P.; Zhang, H.; Jan, A.; Pei, Q., Long lifetime dielectric elastomer actuators under continuous high strain actuation. *In Electroactive Polymer Actuators and Devices (EAPAD)* **2009**, 7287, 72870O.