A Biomorphic Origami Actuator Fabricated by Folding a Conducting Paper

H Okuzaki, T Saido, H Suzuki, Y Hara and H Yan
Laboratory of Organic Robotics, Interdisciplinary Graduate School of Medicine and Engineering, University of Yamanashi, 4-4-37 Takeda, 400-8511, Japan

E-mail: okuzaki@yamanashi.ac.jp

Abstract. Cooperation between the electrical conductivity and hygroscopic nature of conducting polymers can provide an insight into the development of a new class of electroactive polymer (EAP) actuators or soft robots working in ambient air. In this paper, we describe an ‘origami’ actuator fabricated by folding a sheet of conducting ‘paper’. The principle lies in the electrically induced changes in the elastic modulus of a humidosensitive conducting polymer film through reversible sorption and desorption of water vapor molecules, which is responsible for amplifying a contraction of the film (~ 1%) to more than a 100-fold expansion (> 100%) of the origami actuator. Utilizing the origami technique, we have fabricated a biomorphic origami robot by folding an electrochemically synthesized polypyrrole film into the figure of an accordion shape, which can move with a caterpillar-like motion by repeated expansion and contraction at a velocity of 2 cm min⁻¹.

1. Introduction

Origami, the art of paper folding, is not only an art form for children of all ages but also a powerful tool for solving problems in the area of science and technology, such as mathematics [1], biomechanics [2], and space engineering [3]. Barbastathis et al. fabricated nanostructures with a nanostructured origami process consisting of patterning a two-dimensional (2D) silicon nitride membrane and then folding it into a three-dimensional (3D) configuration using stressed metal hinges [4]. Vaccaro and colleagues used semiconductor films to construct an array of micromirrors by the micro-origami technique, which allowed the fabrication of hinges for movable parts.[5] However, they did not really ‘fold’ the origami but processed hinges by using a technology from photolithography or micro electro-mechanical systems (MEMS) so as to fold into the 3D structures. This paper deals with the first study on real origami actuators fabricated by folding a sheet of conducting ‘paper’. The principle lies in the electrically induced changes in the elastic modulus of a humido-sensitive conducting polymer film through reversible sorption and desorption of water vapor molecules, which is responsible for amplifying a contraction of the film (~ 1%) to more than a 100-fold expansion (> 100%) of the origami actuator.

2. Experimental

Polypyrrole films doped with tetrafluoroborate were electrochemically synthesized by anodic oxidation of pyrrole (0.06 M) in the presence of tetraethylammonium tetrafluoroborate (0.05 M) in propylene carbonate containing 1wt% of water. A constant current (0.125 mA cm⁻²) was applied through a platinum plate (100 mm long, 50 mm wide, 100 μm thick) acting as the anode and an
aluminum foil (300 mm long, 100 mm wide, 50 µm thick) as the cathode with a potentiostat (HA-301, Hokutodenko) for 10 h at -20°C. After polymerization, the polypyrrole film was peeled from the platinum electrode, soaked in a large amount of propylene carbonate, and dried overnight in a vacuum. Thickness, electrical conductivity, and doping ratio of the resulting film were 20 µm, 140 S cm⁻¹, and 0.33, respectively. The actuation of the origami actuators was evaluated with a digital video camera (DCR-PC1000, Sony) and laser displacement meter (LB-80, Keyence). The isothermal sorption of water vapor to the polypyrrole film was measured at 25°C with a Belsorp aqua3 (Bel Japan). Prior to the measurement, the film was dried at 140°C for 1 h in air followed by at 100°C for 6 h in a nitrogen stream. The degree of sorption, defined as the weight percent of sorbed water to dry polymer, was measured at each water vapor pressure after reaching the equilibrium. The dimensional change of the film (20 mm long, 2 mm wide, 20 µm thick) was measured with a TMA/SS6200 (SII NanoTechnology) at 25°C in a RH range from 25% to 90% at a rate of 0.5%RH min⁻¹ in a nitrogen stream under a constant tension of 49 mN mm⁻². These were the minimal values to tense the film.

3. Results and Discussion
Differing from one-dimensional (1D) fiber and 2D film actuators, the origami actuator has its 3D structure constructed by folding a conducting polymer film. Figure 1 shows successive profiles of a biomorphic origami robot fabricated by connecting two accordion-shaped origami actuators in series and a pair of plastic plates acting as pawls attached to the ends. Upon application of 3 V DC for 5 s through copper wires connected to the ends with a silver paste and then turned off for 10 s, the origami robot moved forward with a caterpillar-like motion by repeated expansion and contraction at a velocity of 2 mm min⁻¹. Unlike EAP actuators using conducting polymers [6-11], polymer gels [12-15], or carbon nanotubes [16], the origami robot walked in air without using an electrolytic solution or counter and reference electrodes.

Figure 1. Time profiles of the biomorphic origami robot in action measured at 25°C and 50% RH. Upon application of 3 V DC for 5 s through copper wires attached to the ends (A), the front pawl can slide forward due to the expansion of the accordion, but the rear hook is prevented by the teeth of the ratchet formed on the substrate from sliding backwards (B and D). When the electric field is turned off for 10 s, the rear hook can move forward due to the contraction of the accordion, but the front hook is prevented from sliding backwards (C and E). Thus, the origami robot moves forward with a caterpillar-like motion by repeated expansion and contraction at a measured velocity of 2 cm min⁻¹.
A procedure to fabricate the accordion-shaped origami actuator is schematically shown in Figure 2. An L-shape film made from electrochemically synthesized polypyrrole doped with tetrafluoroborate (36 mm long, 3 mm wide, 20 µm thick in one side) is folded into the figure of an accordion shape (A-D). The resulting origami actuator is sandwiched between two glass plates and annealed at 140°C for 1 h in air to crease properly. Figure 2E shows an optical image of the as-folded accordion-shaped origami actuator before annealing. The origami robot in Figure 1 was composed of two accordion-shaped origami actuators adhered top-to-bottom in series with a silver paste instead of attaching copper wires at both ends.

Figure 2. Schematic illustrating the fabrication process (A-D) and photograph (E) of an accordion-shaped origami actuator. A film made from electrochemically synthesized polypyrrole doped with tetrafluoroborate is cut into an L shape (36 mm long, 3 mm wide, 20 µm thick in one side) and a copper wire is attached to the corner with a silver paste (A). After turning the film over, one side is folded to the opposite side (B), and then the other side is folded in the same manner (C). By alternately folding each side of the film six times, the polypyrrole film is folded into the figure of an accordion shape and then a copper wire is attached on its top (D).

Figure 3 demonstrates a clear difference between the origami and film actuators. The current passing through the origami actuator is proportional to the applied voltage, indicative of an ohmic nature (Figure 3A). At 2V, almost the same current flows through the film actuator while the surface temperature rises from 25°C to 36°C. It is seen from Figure 3B that the origami actuator exhibits rapid and significant expansion upon application of the electric field. The maximum strain increases as the applied voltage is increased with the value reaching 147% at 2 V as shown in the inset of Figure 3B, which is two orders of magnitude larger than the film contraction (0.8%) caused by desorption of water vapor due to Joule heating (Figure 3C). A similar phenomenon is observed in the deformation of the other origami actuator prepared by folding a polypyrrole strip (40 mm long, 3 mm wide, 20 µm thick) in a spring shape (Figure 4A). It is found that the average angle of the creases \( \theta_{av} \) increases from 25° to 31° with the application of 2 V for 5 s, demonstrating that the unfolding of the creases causes significant expansion of the origami actuator. At the creases formed by folding the polypyrrole film, the force to fold balances with that to unfold, thereby exhibiting spring characteristics. This balancing of forces also determines the length of the origami actuator. The application of the electric field causes desorption of water vapor and contraction of the film, leading to an increase in the elastic modulus making the film more difficult to be deformed. Therefore, a force to unfold the creases allows the angles to be extended, thereby expanding the origami actuator. The expansive force measured under isometric conditions attained 24 kPa at 2 V, that is two orders of magnitude smaller than the film actuator contractile forces (6 MPa) [17]. Thus, the origami actuator demonstrates extremely large strains with over 100% trade-off with small stresses, ascribed to the small value of the origami actuator spring constant (14 kPa).
It can also be seen from Figure 3B that a further application of the electric field brings about contraction of the actuator. This can be explained by refolding the creases where a force to hold overcomes that to unfold due to further film contractions. On the other hand, when the electric field is turned off, resorption of water vapor lowers the modulus allowing the creases to be folded, leading to the slight contraction of the origami actuator. Finally, further sorption of water vapor restores the actuator to its original size. Here, the annealing temperature critically influences the initial length and strain of the origami actuator. As shown in Figure 5, the accordion-shaped origami actuator becomes flatter with increasing in the annealing temperature. It is noted that the maximum strain at 2 V attains 150% for the actuator annealed at 140°C because the initial length becomes shorter. On the other hand, the annealing at higher temperatures decreases the strain as well as the electric current, which is provably due to the degradation of polypyrrole chains.

In contrast, another spring-shaped actuator prepared by electrochemical polymerization of pyrrole on a spring-shaped Ti electrode used as a template (B). The 2 V was applied for 5 s through copper wires attached to the ends at 25°C and 50% RH.
This clearly indicates that the unfolding the creases is not due to the kinetics of the desorption between inside and outside of the crease, and folding the polypyrrole film is critical in the deformation of the origami actuator. Thus, the results allow us to conclude that the elastic modulus of the film plays a predominant role in controlling the force balance between folding and unfolding at the creases. Indeed, Young’s modulus and tensile strength of the film respectively increase from 0.68 ± 0.14 GPa and 41 ± 10 MPa to 0.96 ± 0.31 GPa and 56 ± 5 MPa with the application of 2 V, while elongation at the break point slightly decreases from 25 ± 14% to 21 ± 11% (Figure 6). This indicates that the film becomes stiffer and less deformative under the electric field. This can be explained in terms of the plasticizing effect of water vapor molecules sorbed in the film that may enhance the micro-Brownian motion of polypyrrole chains thus lowering the elastic modulus [18].

Figure 5. Dependence of annealing temperature on initial length (A) and strain (B) of the accordion-shaped origami actuator under various voltages measured at 25°C and 50%RH. The as-folded origami actuators were sandwiched between two glass plates and annealed at different temperatures in air for 1 h.

Figure 6. Typical stress-strain curves of polypyrrole film measured under 0 V and 2 V at 25°C and 50%RH. Chuck distance: 40 mm, Strain rate: 10% min⁻¹.

A clear indication of the importance of humidity on dimensional changes of the origami actuator is demonstrated in Figure 7A. At 80%RH, the maximum length of the origami actuator increases with the applied voltage with strain attaining 110% at 2 V. On the other hand, no notable expansion is seen
at 20%RH because the film is almost completely dry and has shrunk even without the electric field. In fact, a change in the relative water vapor pressure from 0.77 to 0.22 decreases sorption degree from 8.2% to 2.7% under a linear contraction of the film (Figure 7B). In this instance, the degree of contraction from 50%RH to 25%RH is 0.9%, being consistent with the contraction under 2 V (Figure 3B), demonstrating the electric field is critical in the effective desorption of water vapor molecules [19]. We should emphasize at this point, that the electrically induced changes in the elastic modulus of the humidity-sensitive conducting polymer is responsible for the film contraction amplification to more than a 100-fold expansion of the origami actuator. Here, the electric field is capable of controlling the sorption equilibrium and mechanical properties of the film.

In conclusion, the origami actuator exhibits strains two orders of magnitude larger (> 100%) than that induced by the electrochemical or chemical doping of conducting polymers [7-11], driven at voltages two orders of magnitude lower (< 3 V) than that of piezoelectric [20] and electrostatic actuators [21] or dielectric elastomers [22,23]. Moreover, the polypyrrole film can be folded into various figures and shapes, such as a paper crane and the Miura-ori. Endo et al. demonstrated that double-walled carbon nanotube ‘buckypaper’ was tough and flexible enough to fold into an origami plane [24]. Thus, a variety of origami papers undergoing mechanical property changes in response to environmental stimuli can be employed using these same principles. This will open up a new field of tailor-made EAP actuators or soft and flexible robots using origami technology as the foundation.

Acknowledgments
The authors gratefully acknowledge Tokyo Electron Ltd. and Takano Co., Ltd. for their financial support. This work was partly supported by a Grant for Practical Application of University R&D Results under the Matching Fund Method from New Energy and Industrial Technology Development Organization (NEDO), Japan.

References
[1] Hull T C 2002 Proceedings of the 3rd International Meeting of Origami Mathematics, Science, and Education (Origami3) ed A K Peters
[2] Mahadevan L and Rica S 2005 Science 307 1740
[3] Miura K 1980 *Proceedings of the 31st Congress of the International Astronautical Federation* (New York: American Institute of Aeronautics and Astronautics) pp 1-10

[4] Arora W J, Nichol A J, Smith H I and Barbastathis G 2006 *Appl. Phys. Lett.* 88 53108

[5] Ocampo J M Z, Vaccaro P O, Fleishmann T, Wang T- S, Kubota K, Aida T, Ohnishi T, Sugimura A, Izumoto R, Hosoda M and Nashima S 2003 *Appl. Phys. Lett.* 83 3647

[6] Bar-Cohen Y 2001 *Electroactive Polymer Actuators (EAP) as Artificial Muscles: Potential and Challenge* (USA: SPIE)

[7] Baughman R H, Shacklelette L W, Eisenbaumer R L, Plichta E J and Becht C 1991 Molecular Electronics (Dordrecht, The Netherlands: Kluwer Academic Pub.)

[8] Smela E, Inganäs O and Lundström I. 1995 *Science* 268 1735

[9] Otero T F and Rodriguez J 1993 Intrinsically Conducting Polymers (Dordrecht, The Netherlands: Kluwer Academic Pub.)

[10] Pei Q and Inganäs O 1992 *J. Phys. Chem.* 96 10507

[11] Winokur M J, Wamsley P, Moulton J, Smith P and Heeger A J 1991 *Macromolecules* 24 3812

[12] Osada Y, Okuzaki H and Hori H 1992 *Nature* 355 242

[13] Kakugo A, Shin S, Gong J P and Osada Y 2002 *Adv. Mater.* 14 1124

[14] Xi J, Schmidt J J and Montemagno C D 2005 *Nature Mater.* 4 180

[15] Yeghiazarian L, Mahajan S, Montemagno C, Cohen C and Wiesner U 2005 *Adv. Mater.* 17 1869

[16] Baughman R H, Cui C, Zakhidov A Z, Iqbal Z, Barisci J N, Spinks G M, Wallace G G, Mazzoldi A, De Rossi D, Rinzler A G, Jaschinski O, Roth S and Kertesz M 1999 *Science* 284 1340

[17] Okuzaki H and Funasaka K 2000 *Macromolecules* 33 8307

[18] Okuzaki H, Kondo T and Kunugi T 1999 *Polymer* 40 995

[19] Okuzaki H and Kunugi T 1998 *J. Polym. Sci., Polym. Phys.* 36 1591

[20] Basedow R W and Cocks T D 1980 *J. Phys. E: Sci. Instrum.* 13 840

[21] Egawa S, Niino T and Higuchi T 1991 *Proc. 1991 IEEE Workshop on Micro Electro Mechanical Systems* pp 9-14

[22] Pelrine R, Kornbluh R, Pei Q, Joseph J 2000 *Science* 287 836

[23] Pelrine R, Kornbluh R and Kofod G 2000 *Adv. Mater.* 12 1223

[24] Endo M, Muramatsu H, Hayashi T, Kim Y A, Terrones M, Dresselhaus M S 2005 *Nature* 433 476