Humidity-Sensitive Polypyrrole Films for Electro-Active Polymer Actuators

Hidenori Okuzaki, Takayoshi Kuwabara, Keiichi Funasaka, and Tomooki Saido*

Electrochemically synthesized polypyrrole (PPy) films undergo rapid and intensive bending in response to sorption of water vapor, which is applied to a new type of motor capable of transducing chemical free energy change of sorption directly into mechanical work. Furthermore, the PPy film contracts in air under an electric field, which is associated with desorption of water vapor due to Joule heating. This paper features an overview of our comprehensive study on the humidity-sensitive PPy films and applications to electro-active polymer actuators. Upon application of 3 V, the film generates contractile stress of 9.8 MPa which is 4 orders of magnitude larger than the gram-force of its own weight (7.6 mgf) and is nearly 30 times that of skeletal muscle in animals (0.35 MPa). The work capacity increases with the applied voltage and reaches 48 kJ m^{-3} at 3 V, while the degree of contraction is about 1%. The 'origami' actuator fabricated by folding the PPy film exhibits a significant expansion as large as 147% at 2 V, which lies in the electrically induced changes in the elastic modulus of the humidity-sensitive PPy film. Utilizing the origami technique, a biomorphic origami robot is fabricated, which can move with a caterpillar-like motion by repeated expansion and contraction at a velocity of 2 cm min^{-1}.

1. Introduction

An isothermal system transducing chemical energy directly into a mechanical work using polymer materials is called a "mechanochemical" or "chemomechanical" system,[1–4] which underlies the motility of all living organisms. Polymer gels have attracted considerable attention for biomimetic or bioinspired systems as replicas of biology because gels can change their size and shape in response to various environmental stimuli such as heat, pH, solvent composition, chemical substances, light, magnetic, and electric field.[1–11] On the other hand, conductive polymers, such as polypyrrole, polythiophene, and polyaniline, have been noted because dimensional changes resulting from electrochemical doping, characterized by transportation of solvated ions between inside of the polymer matrix and the surrounding electrolyte solution, electrostatic repulsion, and/or structural distortion through oxidation of π-conjugated polymers, can be applied to produce electro-active polymer (EAP) actuators or artificial muscles.[12–14] However, most of them operate in an electrolyte solution or in a swollen state, while few reports have been investigated on solid-state polymers in a redox gas atmosphere or that employ a polyelectrolyte or ionic liquid.[15–19]

We have previously discovered that electrochemically synthesized polypyrrole (PPy) films undergo rapid and intensive bending in air, the mechanism of which is based on reversible sorption and desorption of water vapor[20,21] differing from conventional doping and dedoping of conductive polymers. It is found that the cooperation between the electrical conductivity and hygroscopic nature of conductive polymers can provide an insight into the development of a new class of EAP actuators or artificial muscles working in ambient air. This paper features an overview of our comprehensive study on the humidity-sensitive PPy films and applications to EAP actuators.

2. Results and Discussion

2.1. Humidity-Sensitive Bending of PPy Film

Since the first observation of a curious phenomenon whereby an electrochemically synthesized PPy film doped with perchlorate underwent rapid bending due to water vapor sorption,[20,21] we have investigated the detailed mechanism from both phenomenological and theoretical viewpoints.[22] For example, if the PPy film (20-μm thick) is placed on a filter paper covered a Petri dish containing lukewarm water, the film starts crawling which lasts for more than an hour (**Figure 1**). A similar phenomenon was observed only for hygroscopic polymers such as cellulose and polyelectrolyte films but the motion was comparatively slow and less reproducible, while no notable bending was seen for other conventional polymer films made of polyolefin, polyester, nylon, and polystyrene.

A clear indication of the importance of humidity on dimensional change of the PPy film is demonstrated in **Figure 2**. The degree of water vapor sorption increases monotonically with increasing RH and the value attains 9.9% at 94% RH.

Prof. H. Okuzaki, T. Kuwabara, K. Funasaka,
T. Saido
Interdisciplinary Graduate School
of Medicine and Engineering
University of Yamanashi
4-4-37 Takeda, Kofu 400-8511 Japan
E-mail: okuzaki@yamanashi.ac.jp

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The isothermal sorption curve can be described using a dual-mode sorption model in which the sorption is assumed to take place by two mechanisms: one is an ordinary dissolution described by a Henry's law type sorption and another is hole-fitting described by a Langmuir type sorption.[23] The amount of water sorbed in the PPy film in the RH range from 13% RH to 90% RH is 0.53×10^{-3} g calculated using the volume (5.25×10^{-3} cm^3) and density (1.452 g cm^{-3}) of the PPy film.[24] On the other hand, the PPy film undergoes linear elongation with increasing RH. Assuming that the dimensional change of the film occurs isotropically, the volume change in the same RH range is 0.48×10^{-3} cm^3, which corresponds to 9.1% of volume of the dry film. Thus, the apparent density of water in the film is estimated as 1.1 g cm^{-3}, which is close to that of liquid water. DSC thermograms of the film, however, exhibited no apparent endothermic peak relating to the melting of freezable liquid water in the heating trace from –50 °C up to 20 °C.[23] These facts suggest that the water molecules sorbed in the film are differently structured as nonfreezing water when bound to the polymer chains and/or dopant ions.

Hidenori Okuzaki obtained his PhD in polymer science from the Hokkaido University in 1994. He has been working on organic and plastic electronics using conductive polymers since 1994 at the University of Yamanashi as an assistant professor. Since 2003, as an associate professor, he has been focusing on the synthesis of highly conductive polymers and applications to soft actuators, conductive micro- and nano-fibers, and organic field-effect transistors.

2.2. PPy Motor

Figure 3 shows time profiles of the bending displacement and sorption degree of the PPy film in response to opening and closing the shutter of solvent container placed at a distance of 2 mm from the film surface. The displacement represents a geometrical change of the film (20-mm long, 5-mm wide, 21-μm thick) at its free end. When the shutter of water container opens, the film bends to the opposite side and the displacement reaches 6–7 mm within 1–2 s where speeds of bending and recovery motion attain 5.6 and 4.9 mm s^{-1}, respectively. Instead of water, polar organic solvents such as alcohol, aldehyde, ketone, amine, or monohaloalkane bring about bending of the film to the same side, namely, the direction of the bending is just opposite to that observed for water. Among the polar organic solvents, iodomethane and *N,N*-dimethylformamide cause the most quick and intensive bending. On the other hand, nonvolatile, aromatic, and nonpolar organic solvents could not induce bending.

In order to clarify the mechanism in more detail, the sorption degree for various solvents is measured by the quartz crystal microbalance (QCM) technique.[21] The dry weight of the PPy polymerized on the gold electrode of the QCM is estimated to be 65.6 μg cm^{-2}. By exposing the QCM to ambient air at room temperature and around 40% RH, the sorption degree immediately increases and attains an equilibrium of 4.3% within 3 min. If the shutter of water container opens, further sorption of water vapor occurs and the sorption degree attains 7% within 5 s. By closing the shutter, the sorbed water molecules rapidly desorb and the sorption degree recovers to the original level. Since the changes in the sorption degree coincide with the deformation of the film, the mechanism of bending might be associated with the asymmetric surface expansion due to the sorption of water vapor from one side of the film.[22] In contrast, iodomethane first decreases the sorption degree and then increases, which indicates that the iodomethane molecules sorb in the PPy by replacing the water molecules sorbed in ambient air. Thus, the bending of the film to the opposite direction can be explained by contraction of the film surface. On the other hand, the sorption degree of benzene within 5 s is one order of magnitude smaller than that of water, thereby no notable bending of the film.

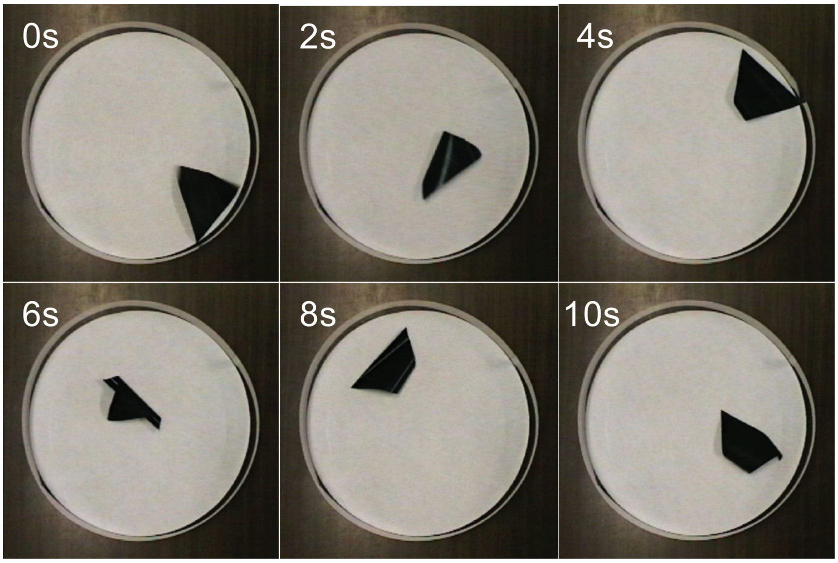


Figure 1. Time profiles of sorption-induced crawling motion of electrochemically synthesized PPy film.

Figure 4 shows the schematic illustration and photograph of a 'PPy motor' in which a closed belt made of PPy film is wound around two pulleys. When a water container is placed below the small pulley, the curvature of the belt increases by surface expansion of the film due to the sorption of water vapor. On the other hand, when the other container of

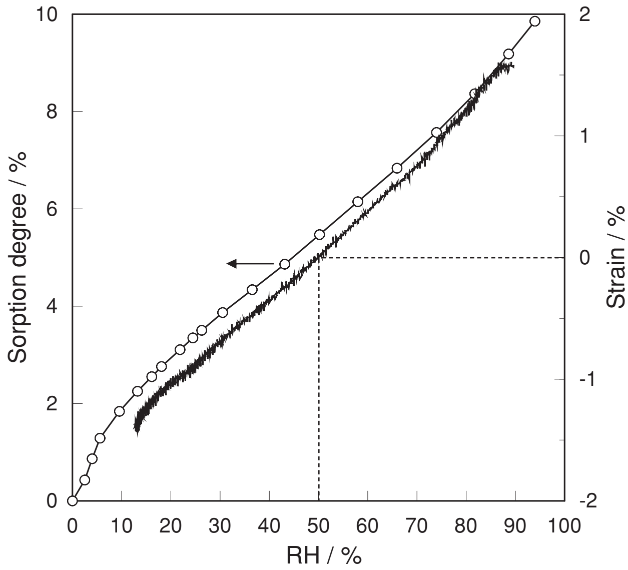


Figure 2. Isothermal sorption of water vapor (open circles) and sorption-induced strain of PPy film (35-mm long, 5-mm wide, 30-µm thick) calculated with respect to the length at 50% RH measured at 25 °C. Reproduced with permission.[24] Copyright 2000, Elsevier.

polar organic solvent is placed above the same pulley, the curvature of the belt decreases by the surface contraction due to the desorption of water vapor. Thus, a net rotary moment acts on the pulleys and causes a clockwise rotation. The rotation brings a new surface of the belt to at the containers, thereby enabling a continuous rotation, where speed of the rotation would depend on the dimensions of the film, the ratio of the radius of the pulleys, and the sorbate used. We have found that when water and iodomethane are used, the belt of PPy film rotates at a speed of 22 cm min^{-1}, corresponding to the rotation of the small pulley at 6–7 rpm.[20,21] The rotation continues until the solvents completely vaporize. Replacing the water container by a finger caused similar rotation of the motor, in which the belt deformed by sorption of water vapor evaporated from the finger surface (Figure 3). Moreover, the PPy motor can rotate by using only water without polar organic solvent,[25] which can provide a clean and silent power source as a new type of molecular engine capable of transducing the chemical free energy change of sorption directly into the mechanical work.

2.3. EAP Actuator

Figure 5 shows time profiles of strain, electric current, surface temperature of the PPy film doped with tetrafluorobotate (35-mm long, 5-mm wide, 30-µm thick), and of humidity in the vicinity of the film surface measured under the thermostatic conditions (25 °C, 50% RH). When DC 2 V is applied, the film exhibits contraction by 1.2% in air and it recovers to the original length after removal of the electric field. The current passing through the film is about 50 mA, and raises the surface temperature from 25 to 32 °C due to Joule heating. The heat produced by Joule heating can be estimated as 60 J using the values of $E = 2$ V, $I = 50 \times 10^{-3}$ A, and $t = 600$ s. It should be noted here, the humidity in the vicinity of the film surface abruptly rises by application of the electric field, demonstrating that water vapor molecules sorbed in the film desorb and scatter into ambient air. A gradual decrease of relative humidity (RH) will be due to the temperature rise near the film because saturated water vapor pressure increases with the temperature, while absolute humidity (AH) reaches an almost constant level. On the other hand, a drop of the humidity when the electric field is turned off can be explained as the resorption of water vapor from the air surrounding the film. Because the humidity change coincides with the dimensional change of the film, the contraction of the film will be associated with desorption of water vapor induced by Joule heating. Here, the electric field is capable of controlling the sorption equilibrium, and the film can sorb and desorb water vapor reversibly in response to changes in the magnitude of the electric fields.[24,26]

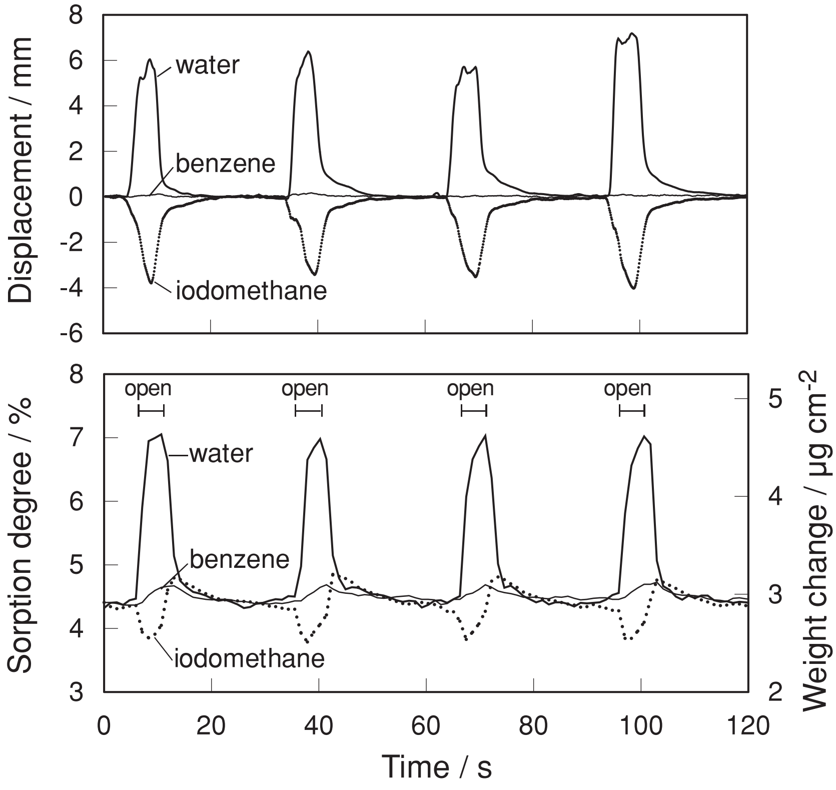


Figure 3. Changes in bending displacement and sorption degree of PPy film in response to opening and closing the shutter of solvent container placed at a distance of 2 mm from the film surface for water (thick solid line), benzene (thin solid line), and iodomethane (broken line) measured by laser displacementometer and QCM technique, respectively. Reproduced with permission.[21]

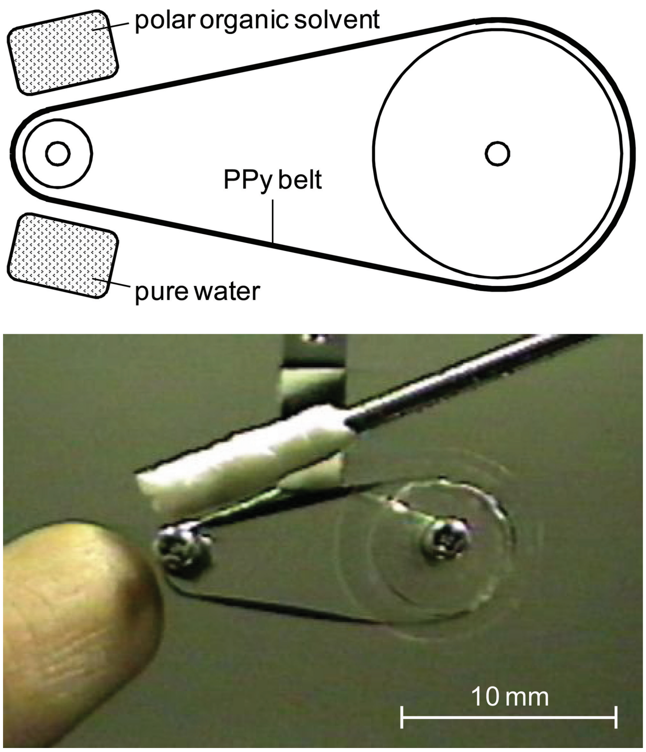


Figure 4. Schematic illustration and photograph of PPy motor. Reproduced with permission.[20,21]

To evaluate the electromechanical properties of the PPy film, the equilibrated stress at each strain is measured under various applied voltages, and the results are shown in **Figure 6**, where the strain (γ) is calculated using the initial length of the film (35 mm) at 25 °C under 50% RH at 0 V. First, a voltage is applied to the film at zero tension under isometric conditions, leading to generate the contractile stress (σ). The stress reaches 9.8 MPa at 3 V, corresponding to an equivalent contractile force of 150 gf, which is 4 orders of magnitude larger than the gram-force of its own weight (7.6 mgf) and is nearly 30 times that of skeletal muscle in animals (0.35 MPa).[27] Furthermore, the stress is proportional to the strain at each applied voltage, indicating that the deformation of the film occurs elastically, where the increase in the applied voltage not only shifts the strain at which the stress begins to rise toward the negative strain side but also increases the slope of the plot. The Young's modulus of the film (E) can be calculated from the slope of the stress-strain relationship by assuming the cross sectional area of the film is nearly constant because the γ values are very small, less than 1%. The parameters obtained are summarized in the inset of Figure 6. The values of σ and γ increase as the applied voltage becomes higher, demonstrating that the stress generation-contraction process is reversible. On the other hand, the Young's modulus of the film at 0 V is 0.6 GPa, which is 4 orders of magnitude higher than that of the swollen gels.[4] It should be noted that the E rises with the applied voltage and achieves 1 GPa under 2 V, indicative of an increase of the value by 67%. This can be interpreted as the removal of the plasticizing effect of water which may enhance the motion of polymer chains to lower the elastic modulus.[28]

These characteristics of the PPy film are capable of producing mechanical work in air by subjecting the film to the ideal stress generation-contraction cyclic process in Figure 6.[29] Under isometric condition, the film charges contractile force as elastic energy. The tensed film then releases the elastic energy as work by contracting reversibly along the stress-strain line under the applied voltage from maximum tension to near zero. After removing the applied voltage, the cycle is then closed by equilibrating the contracted film on exposure to air, where the film expands to the initial length owing to the resorption of water vapor. Thus, the area below the stress-strain line represents the volumetric work capacity (W) in Equation (1)

$$W = \frac{1}{2}\sigma \times \gamma \tag{1}$$

The results are shown in **Table 1**. A marked rise in the W is observed as the applied voltage becomes higher and the value reaches 48.2 kJ m^{-3} under 3 V, which is much higher than in muscles (0.8 kJ m^{-3}) and comparable to that generated during the electrochemical doping-dedoping cycle for various conductive polymers.[30,31] It is obvious that the W increases by stretching the film, for example, the value under 3 V at strain of 1% attains 381 kJ m^{-3}. If we define the energy efficiency (η) as the ratio of work capacity to the electric energy, we obtain the η using the volume of the film ($V = 5.25 \times 10^{-9}$ m^3) with Equation (2)

$$\eta = \frac{W \times V}{E \times I \times t} \times 100 \tag{2}$$

where E, I, and t are applied voltage, electric current, and typical response time, about 30 s, respectively. It is seen from Table 1 that the values of η are on the order of 10^{-3}% regardless of the applied voltage, which is much smaller compared with electrical motors or mechanical engines. This will arise from the fact that almost of the electric energy is dissipated as the heat through Joule heating and/or the long response time. We should emphasize here that the PPy film exhibits a similar performance and work capacity as that driven by the electrochemical or chemical doping. Unlike in the conductive polymers based on the electrochemical doping, this system can serve as a new type of electrically driven muscle that works in air without using an electrolyte solution or counter and reference electrodes.

2.4. Origami Actuator

Differing from 1D fiber and 2D film actuators, the origami actuator has its 3D structure constructed by folding a conductive polymer film. A procedure to fabricate the accordion-shaped origami actuator is schematically shown in **Figure 7**. A film made from electrochemically synthesized PPy 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 (Figure 7A). After turning the film over, one side is folded to the opposite side (Figure 7B), and then the other side is folded in the same manner (Figure 7C). By alternately folding each side of the

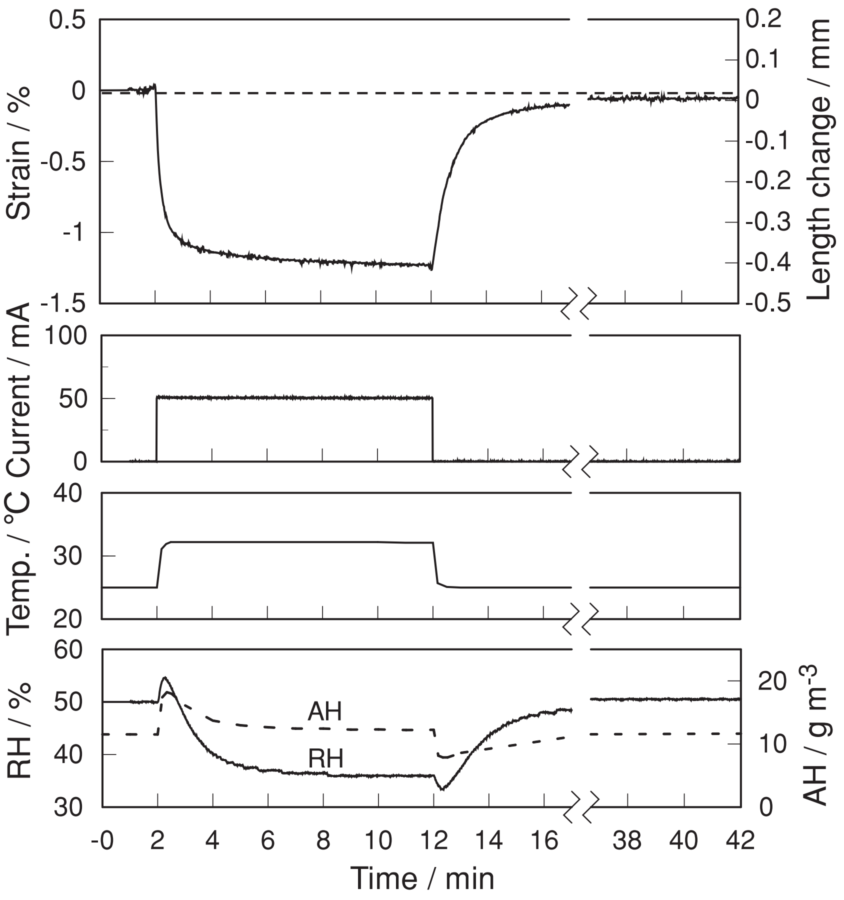
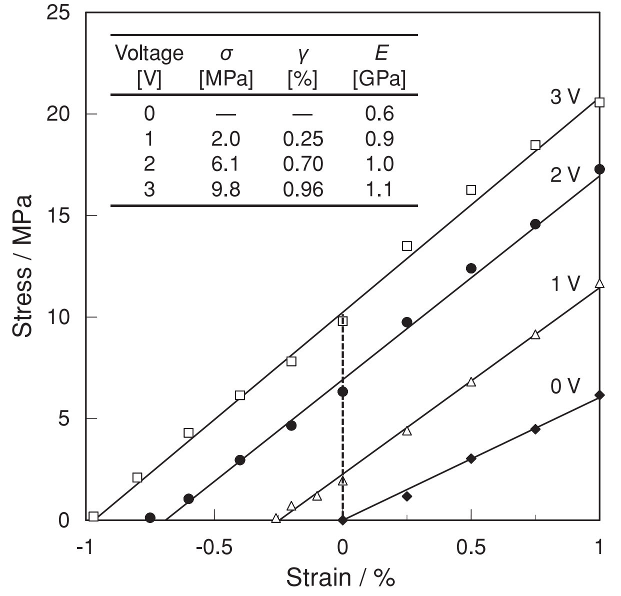


Figure 5. Time profiles of strain, electric current, and surface temperature of PPy film and of RH and AH in the vicinity of the film surface under 2 V measured at 25 °C and 50% RH. Reproduced with permission.[24] Copyright 2000, Elsevier.

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 (Figure 7D). Figure 7E shows a photograph of the as-folded accordion-shaped origami actuator before annealing. The resulting origami actuator is sandwiched between two glass plates and annealed at 140 °C for 1 h in air to crease properly (Figure 7F). It is seen that the origami actuator exhibits rapid and significant expansion upon application of the electric field (Figure 7G). The maximum strain attains 147% at 2 V, which is two orders of magnitude larger than the film contraction caused by desorption of water vapor due to Joule heating (Figure 5). The mechanism is based on unfolding of the creases that results in the significant expansion of the origami actuator.[32] At the creases formed by folding the PPy 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 an 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 (Figure 6). 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 (6.1 MPa) as shown in Figure 6. Thus, the origami actuator demonstrates extremely large strains with over 100% trade-off with small stresses, ascribed to the small value of spring constant of the origami actuator (14 kPa).

On the basis of origami technology, a biomorphic origami robot is fabricated by connecting two accordion-shaped origami actuators in series and a pair of plastic plates acting as pawls attached to the ends (**Figure 8**). Upon application of DC 3 V for 5 s through copper wires attached to the ends (Figure 8A), 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 (Figure 8B,D). When the electric field is turned off for 10 s, the rear hook can move forward due to the contraction



Voltage [V]	σ [MPa]	γ [%]	E [GPa]
0	—	—	0.6
1	2.0	0.25	0.9
2	6.1	0.70	1.0
3	9.8	0.96	1.1

Figure 6. Stress-strain relationship of PPy film under various applied voltages measured at 25 °C and 50% RH. The inserted table represents contractile stress (σ), contractile strain (γ), and Young's modulus (E). Reproduced with permission.[29] Copyright 2000, American Chemical Society.

Table 1. Volumetric work capacity (W), electric current (I), and energy efficiency (η) for PPy film under various voltages. Reproduced with permission.[29] Copyright 2000, American Chemical Society.

Voltage [V]	W [kJ/m^3]	I[mA]	$\eta^{a)}$ [%]
1	2.4	27.8	1.5×10^{-3}
2	21.4	51.5	3.6×10^{-3}
3	48.2	77.8	3.6×10^{-3}

[a)] Calculated using a typical response time of 30 s.

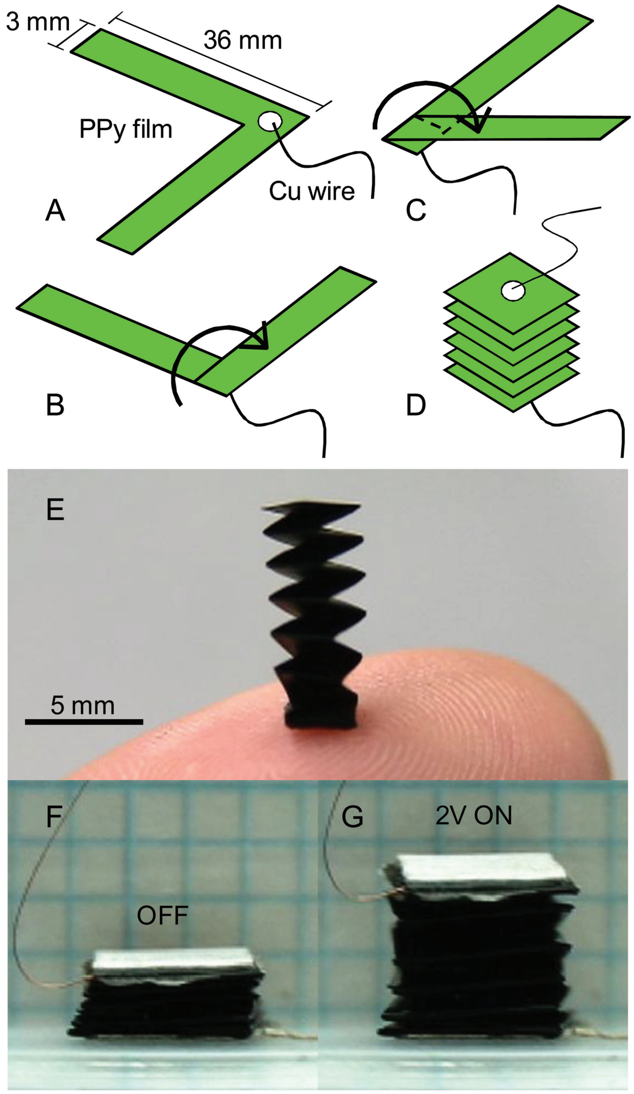


Figure 7. A–D) Schematic illustrating the fabrication process of accordion-shaped origami actuator and photographs E) before annealing and F,G) after annealing under 0 V (F) and 2 V (G). Reproduced with permission.[32] Copyright 2008, Institute of Physics.

of the accordion, but the front hook is prevented from sliding backwards (Figure 8C,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}.[32] Unlike EAP actuators using conductive polymers,[12–19] polymer gels,[1–11] or carbon nanotubes,[33] the origami robot walked in air without using an electrolytic solution or counter and reference electrodes. Furthermore, this system can work at voltages 1 or 2 orders of magnitude lower than piezoelectric actuators[34] or dielectric elastomers[35] because the dimensional change is caused by the electric current but the electric field. Moreover, various conductive polymers or their derivatives that undergo dimensional changes due to the sorption of water vapor can be employed on the same principle.[36]

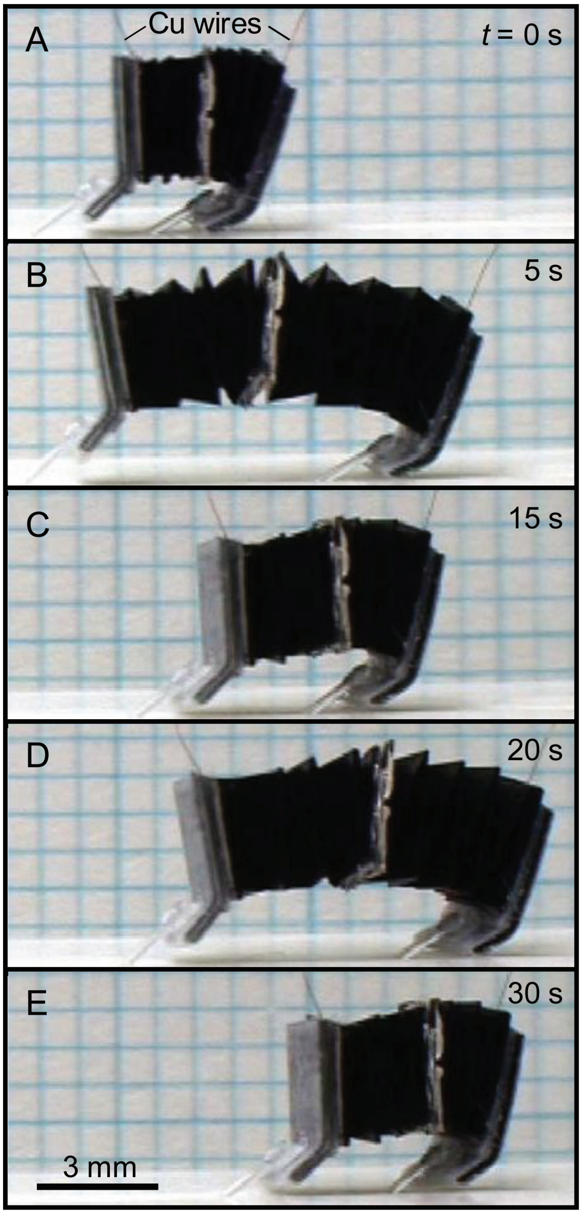


Figure 8. Time profiles of biomorphic origami robot in action measured at 25 °C and 50% RH. Reproduced with permission.[32] Copyright 2008, Institute of Physics.

3. Conclusions

The electrochemically synthesized PPy films undergo rapid and significant bending in air, which is associated with reversible sorption and desorption of water or polar organic solvent. Based on this phenomenon, we have devised the 'PPy motor' driven by sorption of water vapor producing a continuous rotation, which can provide a clean and silent power source as a new type of molecular engine capable of transducing the chemical free energy change directly into the mechanical work. On the other hand, the application of an electric field brings about reversible contraction of the PPy film in air, the mechanism of which is associated with desorption of water vapor due to Joule heating. Furthermore, we have developed an 'origami' actuator fabricated by folding the PPy film. The principle lies in the electrically induced changes in the elastic modulus of the humidity-sensitive PPy 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%). Thus, the cooperation between the electrical conductivity and hygroscopic nature of conductive polymers can provide an insight into the development of a new class of EAP actuators or artificial muscles working in ambient air.

4. Experimental Section

Pyrrole as a monomer was purified by distillation before use. Tetraethylammonium salts of perchlorate and tetrafluoroborate as electrolytes were commercially available and used without further purification. Polypyrrole (PPy) films doped with perchlorate and tetrafluoroborate were electrochemically synthesized by the anodic oxidation of the pyrrole in the presence of different electrolytes. The pyrrole monomer (0.06 mol L^{-1}) and electrolyte (0.05 mol L^{-1}) were dissolved in propylene carbonate containing water (1%). A constant current (0.125 mA cm^{-2}) was applied through a platinum plate (50-mm long, 20-mm wide, 0.1-mm thick) as an anode and aluminum foil 10 times in area as a cathode with a potentiostat (HA-301, Hokutodenko). Polymerization was carried out at –20 °C for 12 h. After polymerization, the PPy films were peeled from the platinum electrode, soaked in a large amount of propylene carbonate, and then dried overnight in vacuum.

The electrical conductivity was measured at 25 °C by a normal four-probe method with a digital multimeter (VOAC-7512, Iwatsu). The isothermal sorption of water vapor to the PPy film doped with perchlorate 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 10% RH to 90% RH at a rate of 0.5% RH min^{-1} in a nitrogen stream under a constant tension (49 mN mm^{-2}), the minimal value to tense the film.

The bending of the PPy films doped with perchlorate (20 mm long, 5 mm wide, and 20 μm thick) suspended from a metal chuck was evaluated by measuring the displacement, representing a geometrical change of the film at its free end, with a laser displacementometer (LB-080, Keyence). An electric shutter-equipped glass tube (50 mm long and 5 mm in the inside diameter) containing various solvents was located at a distance of 2 mm from one side of the film below the chuck. The changes in the sorption degree were measured by the use of a quartz crystal microbalance (QCM) technique. The PPy was polymerized on a gold electrode of the AT-cut QCM for 30 min and then dried overnight in a vacuum. The frequency change was monitored with a universal counter (SC-7202, Iwatsu) and the sorption degree was calculated using the resonance frequency change.

The contraction of the PPy film doped with tetrafluoroborate (35-mm long, 5-mm wide, 30-μm thick), magnified 10 times by the principles of the lever and fulcrum, was measured using a laser displacementometer (LB-080, Keyence) where the strain of film at 50% RH was adjusted to zero. The electric field was applied to the PPy film with a potentiostat (HA-301, Hokutodenko) through copper wires attached to both ends of the film with silver paste. The temperature at the film surface was measured with an infrared thermometer (THI-500S, Tasco) and RH in the vicinity of the film was measured with a hygrometer (MC-P, Panametrics) the probe of which was located 1 mm from the film surface. The electromechanical properties of the PPy film doped with tetrafluoroborate under the electric field were measured with a tensile tester (Tensilon, Orientec), where voltage was applied through copper wires attached to both ends of the film (35-mm long, 5 mm wide, 30-μm thick) with silver paste. Young's modulus, tensile strength, and elongation at break of the PPy film were calculated from the stress-strain curves measured at 25 °C under 50% RH.

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