

Sensitively Humidity-Driven Actuator Based on Photopolymerizable PEG-DA Films

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Hydrogels such as poly(ethylene glycol) diacrylate are a class of cross-linked polymers that have the ability to absorb water and change volume. Here, a humidity responsiveness of hydrogel film fabricated by the photopolymerization of poly(ethylene glycol) diacrylate (PEG-DA) monomer is reported. This kind of film could be driven by a small humidity gradient, thus spontaneous deformation and motion can be achieved by placing the film onto a moist filter paper. The influence factors of humidity response sensitivity such as the exposure time and number-average molecular weight of PEG-DA monomer used during the fabrication are investigated. Under different relative humidity, the film displays different degrees of pink color and the intensities of fluorescence under 365 nm illumination are also different, which renders the film to be used as a “humidity test strip.” A humidity-driven walking device is fabricated and the walking velocity is about 3 mm min⁻¹ under the actuating water vapor. Vapor of volatile polar solvents could also drive the actuator to achieve fast deformation and shows the variety of the film responsivity.

In nature, plants and muscular organs are capable of converting physical or chemical signals into macroscopic movements with changing shape, size, or properties in response to specific external stimuli in the environment.^[1–4] For example, sea cucumbers can alter the stiffness of their dermis within seconds to obtain survival advantages;^[5] the Venus flytrap can close their leaves in a second for efficient prey capture.^[6] Recently, inspired by these biological examples, a variety of artificial smart actuators that convert a change in environmental conditions, for example, electric,^[7,8] thermal,^[9–11] light,^[12,13]

and chemicals^[14–17] into a mechanical response, has attracted much interests.

Humidity-responsive behaviors are widely spread in nature, such as humidity-responsive events of plants and fungi during the dispersion of seeds and spores,^[18] the opening of pine cones,^[19] and twisting and bending of wheat awns (*Triticum turgidum*).^[20] During the alteration of the environmental moisture content, that is, relative humidity, a particular part of the biological systems reversibly absorbs or releases the moisture. During this process, a mechanical deformation takes place, with the goal to perform a desired function such as directed complex motions. Inspired by nature, moisture-responsive materials have awoken great interest. Due to the environment-friendly nature of water vapor and its easy accessibility, moisture-responsive materials

are promising candidates for broad applications, for example, sensors,^[21] actuators,^[22–27] or construction of soft robots.^[28–31] A universal strategy is the use of polymer films, which can undergo reversible water-sorption-induced swelling for actuation. For example, Ma et al. reported that composite polymer film combining polypyrrole and polyol-borate rapidly performed moisture-driven locomotion and is of particular interest for generation of piezoelectric energy.^[32] Very recently, Zhang et al. fabricated a polymer film actuator composed of hygroscopic agarose and azobenzene-containing photoactive poly(ethylene glycol), which is capable of self-actuation in response to humidity gradient and of photogated control by light.^[33] Cellulose-type film showed significant moisture response, especially the film of cellulose stearoyl esters shows reversibly moisture-responsive shape transition,^[34] and nanofibrillated cellulose film is responsive to small humidity of human hand.^[35] In addition, scaling up bilayer film of *Bacillus subtilis* spores and polyimide tape as engines is able to drive a miniature car and to power light-emitting diodes (LEDs).^[36]

There is ever greater interest of using hydrogels for the construction of functional materials in recent years.^[37–42] Hydrogels are 3D hydrophilic polymeric networks, which swell but do not dissolve when contact with water, allowing one to achieve the desired degrees of polymer hydration and volume changes in response to a change in surrounding moisture.^[43–45] For example, Aizenberg and co-workers reported hydrogel film based on polyglycidyl methacrylate can actuate

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to reorientation of embedded silicon nanocolumns depending on the humidity level.^[46] Schenning and co-workers reported polymer liquid crystalline hydrogel containing poly(acrylic acid) is able to lead color change due to humidity response.^[47] The moisture-responsive devices from hydrogel materials with large deformation are expected to promote the application of hydrogel. Poly(ethylene glycol) (PEG)-based materials are widely investigated for biomedical applications due to their biocompatibility, low immunogenicity, and ease of use. PEG-based materials showed a high humidity sorption due to the presence of hydrogen bond between the PEG chains and water molecule. A photopolymerizable-curing technique assures an easy preparation of the films based on the PEG-based materials, such as poly(ethylene glycol) diacrylate (PEG-DA). As the conversion of the double bonds is complete, the hydrogel networks formed. In PEG-DA, the crosslinking density was homogeneous because the molecular weight of PEG chain segments between the acrylate groups is the same.^[48] The homogeneous PEG chain segments in PEG-DA enable it to be used as an ideal material for studying humidity sorption properties of gel. Under the influence of ultraviolet rays, the acrylate double bond breaks and connects with other PEG-DA molecule to form the crosslinking point and network. When moisture is applied, diffusion of water molecule and small solutes in network results in an intermolecular hydrogen bonding. The polymer network would be responsive to water sorption and desorption. Therefore, PEG-DA hydrogel film exhibits dramatic, fast, and reversible deformation and recovery in response to environmental moisture, visually reminiscent of “fast twitch” muscle activity. In this report, we fabricated PEG-DA hydrogel humidity-responsive films, which could be actuated by small humidity gradient such as physiological moisture, and a continuous flipping motion was achieved by placing the film onto a moist filter paper substrate.

The freestanding PEG-DA hydrogel film was prepared by photopolymerization (Figure 1a), and the hydrogel was dyed by rhodamine B, which has a strong water adsorption property (Figure 1b). Optical graphs of the film just released from the mold and at equilibrium swelling state in water at room temperature are shown in Figure 1c,d, respectively. The PEG-DA hydrogel film was almost colorless in the air (Figure 1c). When we put it into water, it absorbed water and became pink (Figure 1d). The exposure of PEG-DA hydrogel film to water resulted in slight swelling and the size of the sample, the length, width, and thickness of the sample increased by 17%, 15%, and 15%, respectively. The total volume increase was ~56%. Atomic force microscopy images of the surface showed that the adsorption of water altered the surface morphology by formation of holes and increased roughness from 0.453 to 1.02 nm (Figure 1e,f). The formation of these holes increased the surface-area-to-volume ration and a high surface-area-to-volume ration facilitates the water desorption. The adsorption or desorption of water continued until a dynamic equilibrium between the film and the surrounding environment was established. The rate of water adsorption and desorption could be reflected by the change of the film's weight. The PEG-DA hydrogel film could absorb water molecules up to 13% of self-weight from humid air with relative humidity (RH) of 100% in 3 min and restored to the initial weight after desorbing water

in the dry environment of 20% RH at approximately the same rate (Figure 1g). Similar to composite of pentaerythritol ethoxylate-polypyrrole^[32] and composite of agarose-poly(ethylene glycol)-conjugated azobenzene derivative,^[33] the larger than 10% strong water absorbing capacity of PEG-DA hydrogel was induced by the formation of hydrogen bonds between the hydrogel molecules and the water molecules. From the Fourier transform infrared (FT-IR) spectra, when the PEG-DA film is exposed to different humidity condition, the $\nu(\text{OH})$ band at $\approx 3500\text{ cm}^{-1}$ increased with broadening and red-shifting of the absorption band as the relative humidity increased (Figure 1h), demonstrated the water adsorption of the PEG-DA hydrogel film. The mechanical property of the PEG-DA hydrogel films was also studied by measuring their stress–strain curves. The average tensile strength of the PEG-DA film before adsorption of water was about 2.80 MPa and after adsorption of water, the average tensile strength was about 2.42 MPa. The decrease of the tensile strength was about 13.6% (Figure S1, Supporting Information). And after adsorption of water, the elongation of the film increased from 8.82% to 10.82%.

Due to the adsorption and desorption of water, the PEG-DA film spontaneously and continuously flipped over a moist filter paper substrate. As a PEG-DA film contacted the moist substrate, the bottom surface absorbed more water than the top surface, which resulted in asymmetric swelling and film curling away from the substrate. A water gradient from high humidity at the face of filter substrate and low humidity in air environment above filter paper caused water adsorption under high humidity and desorption under low humidity.

At temperature of 50 °C from a moist filter paper on an electric hot plate and an environment temperature of 20 °C, relative humidity of 25%, a PEG-DA film with rectangular shape 2.5 cm × 1 cm on the moist filter paper presented a series of flipping motion (Figure 2a, Movie S1, Supporting Information). First, the film was just put on the moist filter paper, both long sides immediately curled up to form a long tube due to water adsorption at bottom surface film and its more stable tubular structure (I). Then the long tube structure had a long and narrow decreased film/substrate contact area and risen film gravity center, so a rocking motion from side to side appeared many times due to the mechanical instability occurred and it finally slanted to one side (II). The initial bottom surface containing water tended to dehydrate, divorced from moist substrate, and initial top surface was just opposite. Considering the tubular shape a horizontally spreading movement generated, and finally, most of the contact area spread on substrate with a new face down (III) to start a new cycle. The similar flipping motion maintained for several cycles. During the flipping motion process of PEG-DA film, both faces of the film were equilibrating with water in the substrate and the low-humidity air above. Thus, the water gradient between the substrate and the air was reflected in the asymmetric film deformation that drove the film locomotion. To investigate the effect of water gradient between substrate and air we changed the temperature of moist substrate to 25 °C and environment relative humidity to 60% (Figure 2b, Movie S2, Supporting Information). During the first cycle the similar process was performed, including formation of tubular structure, horizontal spreading, and opposite face down. But the film deformation and motion speed were

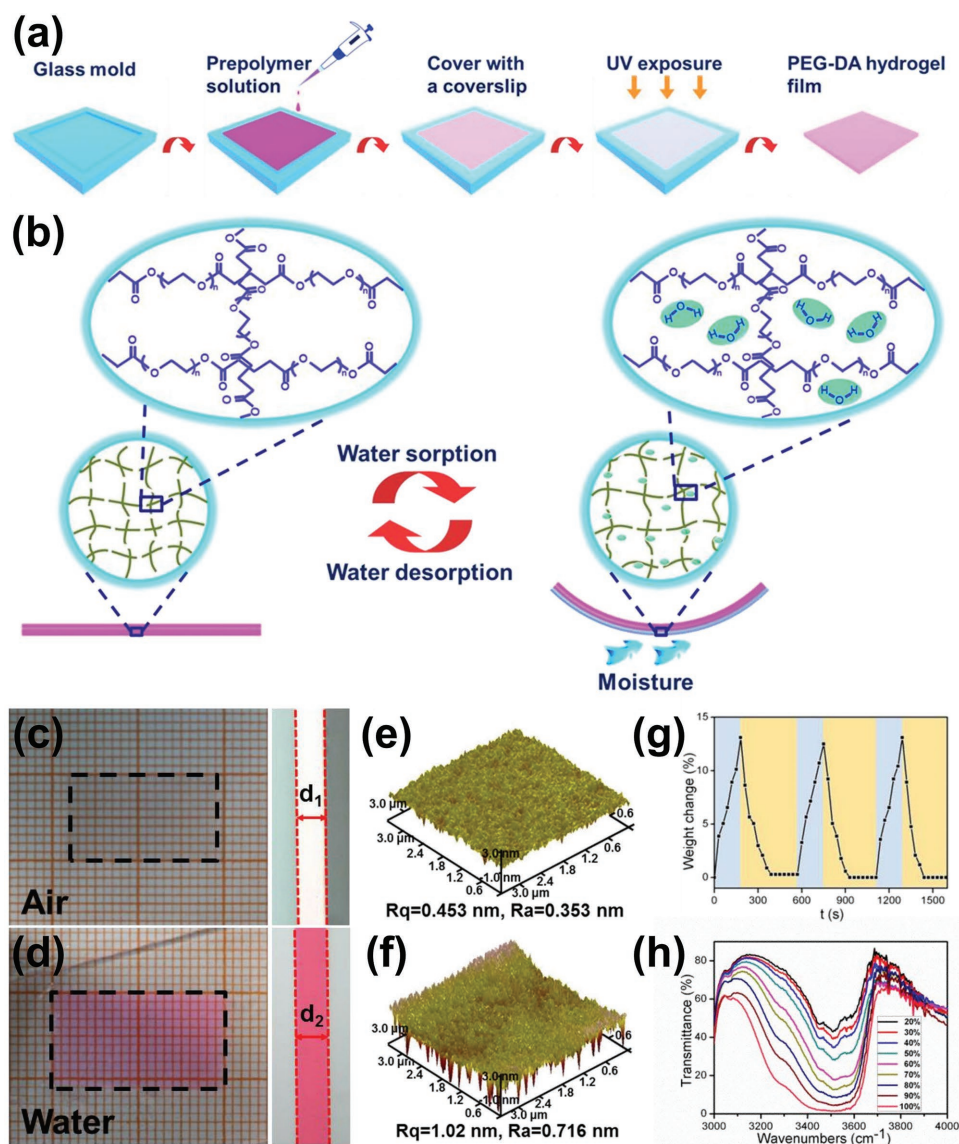


Figure 1. a) Schematic illustration of the fabrication process of the humidity-driven PEG-DA hydrogel films. b) Schematic of the swelling process, under humidity gradient, the side of the film which was faced to moisture swelled by absorbing water. Optical graphs of the c) freestanding PEG-DA hydrogel film with area of $1.7 \times 1 \text{ cm}^2$ and $d_1 \approx 39 \mu\text{m}$ in the air and d) equilibrium swelling state with area of $2 \times 1.15 \text{ cm}^2$ and $d_2 \approx 45 \mu\text{m}$ in water. d_1 and d_2 stood for the thicknesses of the film in the air and in water. AFM images of the e) surface morphology of the PEG-DA hydrogel films in the air with RH = 20% and f) exposed to the humid air with RH = 100%. g) The weight change of the hydrogel film with the change of air relative humidity. The blue and yellow background regions, respectively, represented that the film absorbed water vapor in the humid environment (RH = 100%) and desorbed water vapor in the dry environment (RH = 20%). h) FT-IR spectra of the hydrogel film placed in different relative humidity showed the $\nu(\text{OH})$ band at $\approx 3500 \text{ cm}^{-1}$ increased with broadening and red-shifting of absorption band as the relative humidity increased.

significantly slower than substrate temperature of $50 \text{ }^\circ\text{C}$ and humidity of 25%. And since the second cycle no two sides of film curled up at the same time, usually a corner preferred to curl up and drove the overturning, the original top face down, a complete reversal was performed. After four cycles, nearly a short side, no corner, preferred to curl up to start the flipping process. It is obvious that with increasing cycle number the deformation speed was slower and cycle time was longer. Because there is no heat-induced bending from the PEG-DA hydrogel film (Figure S2, Supporting Information), the difference of the flipping motion under the two conditions was

due to the effect of water gradient between substrate and air. Also from the movie, we can see that if the film was just put on wet filter paper the film showed a pale pink color. After 1 min of flipping process, the color of the film became deeper, indicating that after several cycles the moisture content of the film was greater than the original film. It means that during flipping process of the thin film, the adsorption speed was larger than desorption, and there was residual water in the film. The residual water can lead to a smaller volume difference between the upper surface exposed to air and the bottom surface in contact with moist substrate, so the turnover

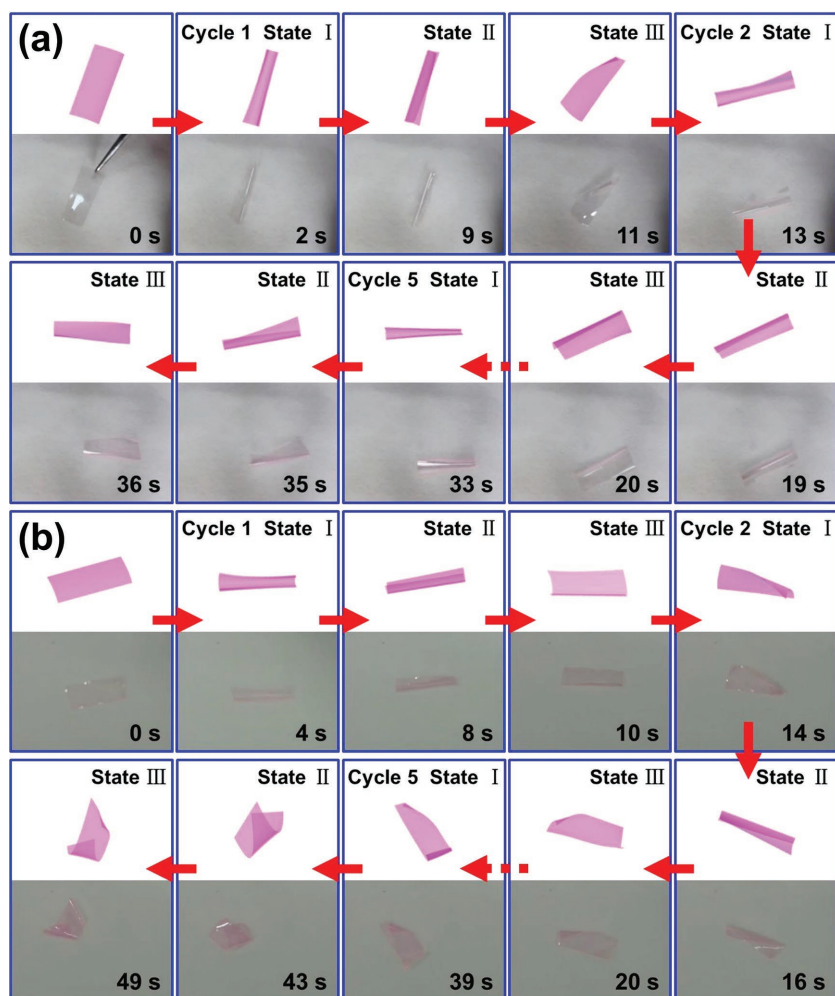


Figure 2. a) PEG-DA film spontaneously and continuously flipped over a filter paper with the paper temperature of 50 °C and RH of 25% and b) the paper temperature of 25 °C and RH of 60%. Both cartoons and snapshots of the real film are shown.

period of the film could be prolonged. A square film with size of 1.5 cm × 1.5 cm was also used to observe the continuous flipping motion (Figure S3, Supporting Information). Until complete evaporation of water in the filter paper the flipping motion sustained. The surrounding conditions affecting the flipping motion, high temperature, and large humidity difference between the moist substrate and the air were beneficial to a fast, continuous, and steady motion. The thickness of the PEG-DA hydrogel film can also affect the motion, the turnover frequency of the flipping motion increased as the film became thinner (Figures S4 and S5, Supporting Information). But when the film was too thin, the film was easy to collapse and adhere, the flipping motion stopped (Figure S6, Supporting Information). So we chose the PEG-DA film with thickness of 40 μm to investigate the continuous flipping motion.

Besides the spontaneous and continuous flipping over a moist filter paper substrate, the PEG-DA hydrogel was able to absorb moisture even from very dilute water vapor environment. Even putting a thin PEG-DA film on a bare palm, it could respond to the physiological moisture. The side

contact to the skin absorbed moisture in the palm and the film started to bend upward (Figure 3a, Movie S3, Supporting Information). The bending reached a steady state in a few seconds and after reaching the steady state, the film could maintain this state and would not relax back even if holding the film for prolonged times. If the bent hydrogel film is turned upside down, the film would become flat first and then bend upward again, that is, a reverse bending process. It shows that the response process was reversible and bidirectional and the film could recover to its original state after moving away from the palm. As a comparison, the PEG-DA hydrogel film was put on a glove-covered palm, no deformation occurred and the film remained flat (Figure S7, Supporting Information). And if we fixed a PEG-DA strip, when a finger is coming close to the strip from right side and without direct touching, the strip will bend to left side because the moisture close to the finger increased and right local area of the film swelled, after the finger moved away, it would come back to the initial position. If the finger came from the left side, the strip would bend to the right side (Figure 3b, Movie S4, Supporting Information). It means that even a very small gradient of humidity close to the PEG-DA film can create local asymmetric swelling of the film and induce local bending.

In order to investigate the humidity response sensitivity and its influence factors of PEG-DA hydrogel film, we measured the bending curvature of the film under different relative humidity difference (ΔRH). The sample was placed outside the window of a home-made humidity chamber, the window

was covered by a coverslip, and the relative humidity difference inside and outside the chamber could be controlled. When the coverslip was removed, the window was open and the side of the film which was faced to the higher relative humidity absorbed water and swelled, the film would bend to the lower relative humidity side (Figure 4a, Movie S5, Supporting Information). The bending angles were measured and the curvature k (cm⁻¹) was calculated using the following equation^[35]

$$k = \frac{\pi}{180^\circ} \frac{\theta}{L}$$

where θ is the bending angle and L is the film's free bending length, here in the experiment L is about 1.65 cm.

We first investigated the effects of the exposure time on the humidity response sensitivity of the PEG-DA hydrogel films. PEG-DA films with different exposure time of 30 s, 1 min, and 2 min were used and Figure 4b showed that all the bending curvature of the three films increased gradually as ΔRH increased. And under the same ΔRH, the film with longer

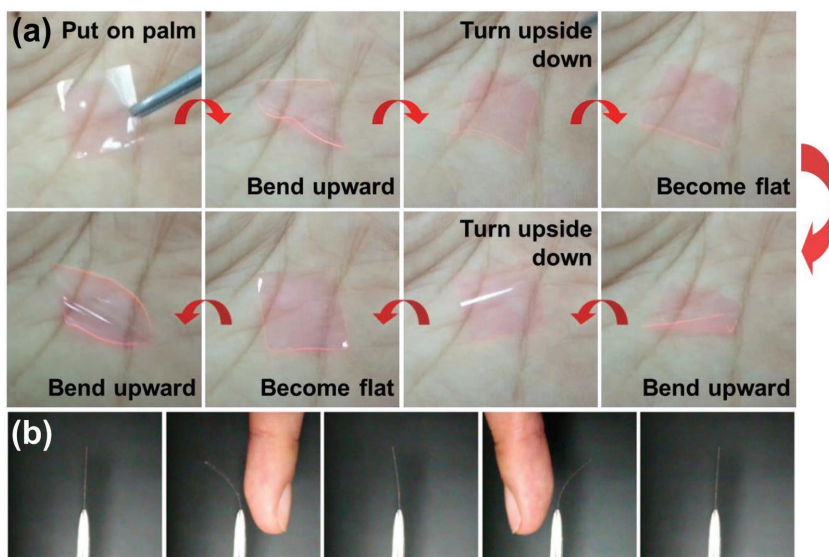


Figure 3. a) The PEG-DA hydrogel film could absorb moisture in the bare palm and bend upward. After turning it upside down, the film became flat and bent upward again. Then again turning the film upside down, the film again became flat and bent upward. b) The fixed PEG-DA strip could bend to the opposite side when a finger came close to it.

exposure time had a smaller bending curvature, which attribute to a high crosslinking density of the polymer networks with the longer exposure time. And the high crosslinking density would decrease the water diffusion^[48] and reduce the humidity response sensitivity of the PEG-DA films.

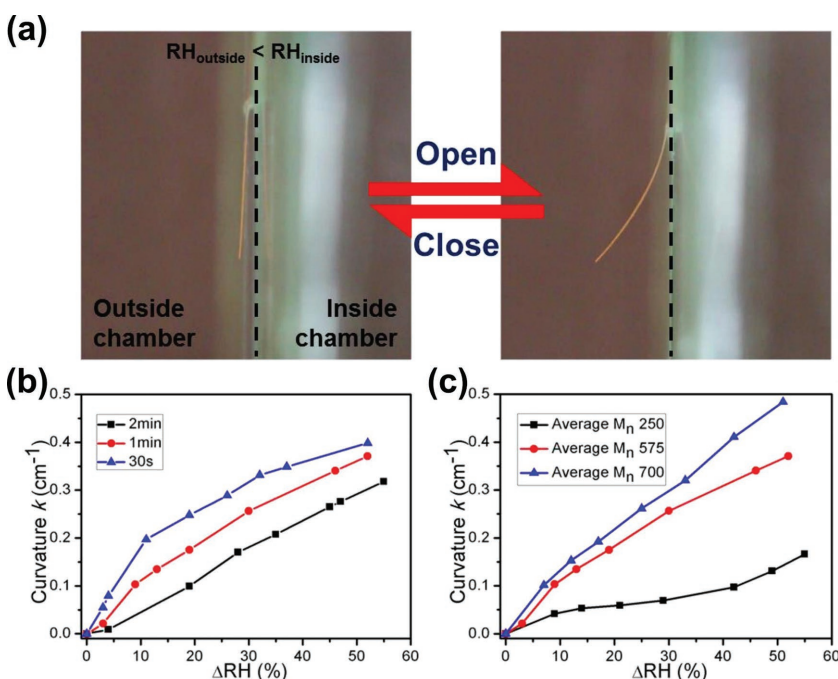


Figure 4. a) PEG-DA hydrogel film was fixed outside the window of a humidity chamber, it remained straight when the window was closed (left) and bent when the window was open (right). b) Dependent curve of bending curvature on ΔRH for hydrogel films with different exposure time. c) Dependent curve of bending curvature on ΔRH for hydrogel films fabricated by using the PEG-DA monomer with average M_n of 700, 575, and 250 (exposure time was 1 min).

PEG-DA monomer with different number-average molecular weights (average M_n) were also used to fabricate hydrogel film. Figure 4c showed that the film fabricated by using the PEG-DA monomer with average M_n of 700 was more sensitive to ΔRH compared to the fabricated film with average M_n of 250 and 575. For the spontaneous and continuous flipping motion, the motion of PEG-DA films fabricated by using the PEG-DA monomer with average M_n of 700 was also more faster than the motion of other films fabricated by using the PEG-DA monomer with average M_n of 250 and 575 (Figure S8, Supporting Information). In polymerizable PEG-DA, the hydrophilic properties increase as a function of the length of the poly(ethylene oxide) chain between both acrylated groups.^[49] And in the curing process, increasing the average molecular weight of the oligomers, the crosslinking density of the network decreased, which was beneficial to improve the humidity response sensitivity.^[50]

The rhodamine B added to the PEG-DA hydrogel film not only made the film much easier to be observed, but also can be used as the environmental humidity indicator. Two PEG-DA hydrogel films were fixed (Figure 5a,b), one was in the home-made humidity chamber (left film in Figure 5a,b) and the other was outside the chamber (right film in Figure 5a,b). When the inside chamber humidity was low, the film was colorless, and under 365 nm illumination, no fluorescence could be observed. As the humidity in the chamber increased, the left film displayed different degrees of pink color and the fluorescence became brighter. The right film outside the chamber did not have any change because the surrounding humidity was constant (RH of 20%). These humidity responsive color changes of the PEG-DA films are reversible (Figure 5b), when the humidity in the chamber decreased, the color of the left film change from pink to colorless and the fluorescence of the film disappeared gradually under 365 nm illumination. We also tested that the pink color of the film did not disappear after immersing into water to absorb water and then dried in the air for 20 cycles (Figure S9, Supporting Information). This phenomenon was due to the rhodamine B concentration of 10^{-4} M in the hydrogel film was high, rhodamine B aggregation occurred by stacking up on its three-ring xanthylium plane and this hindered fluorescence yield and diffusion.^[48] By increasing the relative humidity, these effects gradually diminished. This could also be detected by the steady-state absorption spectra and the emission spectra (Figure 5c,d). The corresponding

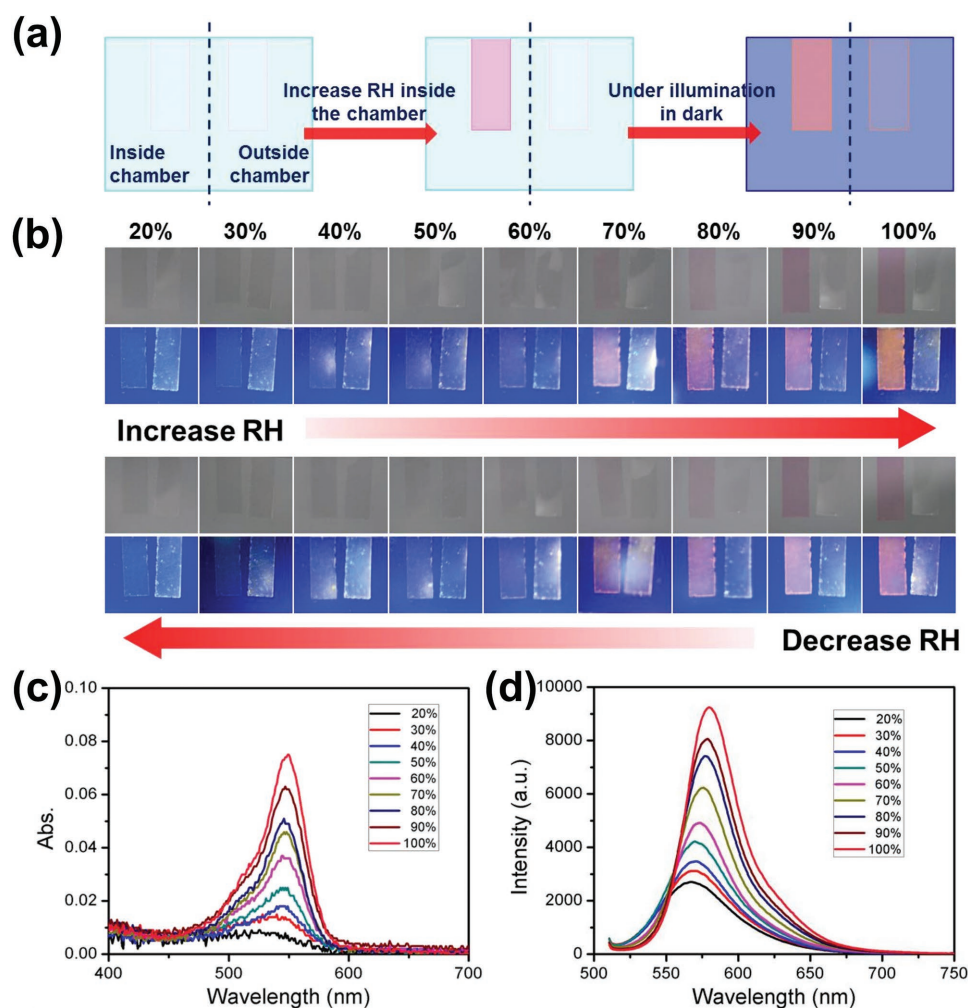


Figure 5. a) Schematic of the PEG-DA hydrogel films was placed inside (left) and outside (right) the humidity chamber. When the RH inside the chamber increased, the color of the left film changed, and the fluorescence of the left film can be observed under illumination in the dark. b) From the optical pictures (the upper line), the left film became pinker as the relative humidity increased, and under 365 nm illumination (the lower line), the fluorescence of the left film became brighter. This color change of the film was reversible, when the relative humidity decreased, the left film became colorless again and the fluorescence of the left film disappeared. The right film maintained its initial state because the relative humidity outside the chamber was constant (RH of 20%). c) Steady-state absorption spectra and d) emission spectra of the hydrogel film under different relative humidity.

absorption and fluorescence peaks increased with increasing the relative humidity. By comparing the color of the film or the fluorescence intensity, we can probably know the relative humidity of the surrounding, so it may work as a “humidity test strip.”

By using the humidity responsive property of the PEG-DA films, a humidity-driven walking device as actuator was fabricated. **Figure 6a,b** showed the actuator walked from a rough ratchet substrate to a smooth slid by applying water vapor. PEG-DA hydrogel strip with length of 2 cm and thickness of 200 μm was placed on the substrate. When the water vapor was on, the upper side absorbed the water vapor and swelled, the strip bent into an arch. And when the water vapor was off, the strip gradually stretched so the walking device moved forward. Because of the rough ratchet substrate, the direction of the motion could be controlled, the walking device could achieve a unidirectional crawling, and the walking velocity of the PEG-DA hydrogel actuator was $\approx 3 \text{ mm min}^{-1}$. This humidity

response walking device mimics the motion of muscles and the crawling of worms, which may find many applications such as soft robots.

The prepared PEG-DA films could be driven not only by humidity stimulus, but also by the vapor of some volatile polar solvents. A piece of PEG-DA film was fixed on a small bottle which contained 5 mL solvent, the distance between the surface of the solvent and the film was about 1 cm. For acetone, chloroform, ethanol, and other volatile polar solvents, the side near the solvent of the PEG-DA film absorbed the solvent vapor and swelled, the film bent upside immediately (**Figure 7a–g**). But if we place the film upon nonpolar solvents such as *n*-hexane (**Figure 7h**), or nonvolatile solvents such as ethylene glycol (**Figure 7i**), the film would remain in its initial state. This property changed the hydrogel actuator from responding to single stimulus such as humidity to responding to a variety of stimuli such as volatile polar solvents, and is beneficial to the wide variety of applications.

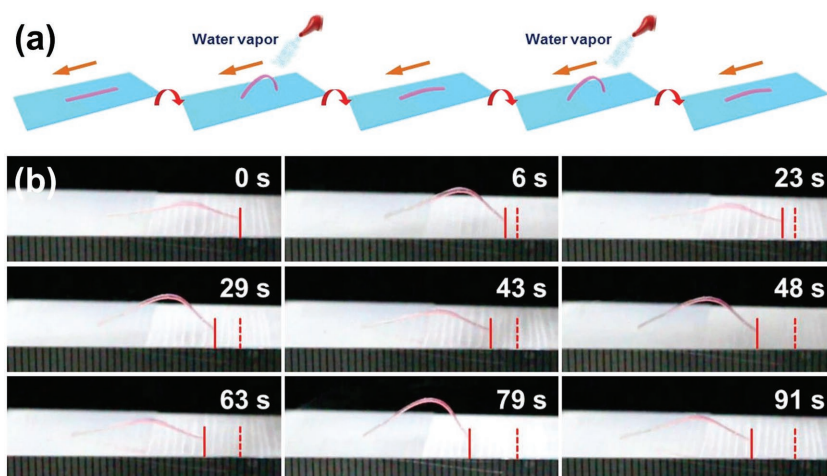


Figure 6. a) Schematic of the motion of humidity-driven walking device, the orange arrow indicated the direction of the motion. b) The walking device walked from a ratchet substrate to a smooth slid by applying water vapor. The red solid lines stood for the positions of the walking device at the corresponding moments and the red dashed lines stood for the initial position of the walking device. The division value of the ruler at the bottom of the images was 1 mm.

In summary, a kind of humidity response PEG-DA hydrogel actuator was successfully fabricated by photopolymerization. The humidity response sensitivity of the hydrogel actuator was high, thus spontaneous deformation and motion occurred under actuating by humidity difference between two sides of the film. The exposure time and the average M_n of PEG-DA monomer used during the preparing process have great influence on humidity-responsive properties. Due to the addition of rhodamine B, the hydrogel films displayed different degrees of

pink color under different relative humidity, and can be used as the environmental humidity indicator. A humidity-driven walking device was fabricated and volatile polar solvent could also drive the actuators, which have great importance for wide variety of applications.

Experimental Section

Materials: PEG-DA (average M_n of 700, 575, and 250) and rhodamine B were purchased from Sigma-Aldrich. Irg. 1173 was purchased from TCI. The ethylene glycol (analytically pure) used to dissolve rhodamine B and other solvents to actuate the PEG-DA hydrogel film were purchased from Beijing Chemical Works.

Fabrication of PEG-DA Actuators: Irg. 1173 as photoinitiator was added into the PEG-DA monomer with average M_n of 575 in a mass ratio of 1:15. 200 μL ethylene glycol solution of rhodamine B (10 mg mL^{-1}) g^{-1} of PEG-DA monomer was added to the mixture of monomer and photoinitiator. The above mixture was kept in the ultrasonic cleaner in the dark for 10 min to ensure all the chemicals were mixed uniformly. The mixture was injected into glass molds with different depths and then irradiated under UV light. After UV exposure, the PEG-DA hydrogel thin films were peeled off from the glass mold. PEG-DA hydrogel films with different thicknesses were fabricated by adjusting the mold depth. In the experiment, the PEG-DA hydrogel films with thickness of 200 μm were used to fabricate the humidity-driven walking device, the PEG-DA hydrogel films with thicknesses of 20, 40, 60, 100, and 200 μm were used to investigate the effect of film's thickness on the turnover frequency of the flipping motion. The mechanical property of the PEG-DA hydrogel films was studied by using films with thickness of 100 μm . Besides these, all the films used in the experiment were 40 μm thick.

For the PEG-DA films with different exposure times, the prepolymer solution consisting of PEG-DA monomer, photoinitiator, and ethylene glycol solution of rhodamine B as above was injected into glass molds with depths of 40 μm and irradiated under UV light (Philips, QVF135), the exposure times were 30 s, 1 min, and 2 min, respectively. Then the PEG-DA hydrogel thin films with different exposure times were fabricated. For the PEG-DA films fabricated using PEG-DA monomer with different average M_n , Irg. 1173 was added into the PEG-DA monomer with average M_n of 250, 575, and 700 in a mass ratio of 1:15, respectively. 20 μL ethylene glycol solution of rhodamine B was added into the mixture, then the prepolymer solution was injected into glass molds with depths of 40 μm and irradiated under UV light for 1 min. Then the PEG-DA hydrogel thin films were peeled off from the glass mold and used for testing.

Investigate the Humidity Response Sensitivity: A home-made humidity chamber was used to measure the effect of exposure time and average M_n on bending curvature of films. The relative humidity inside the chamber was controlled and the relative humidity outside the chamber was fixed at about $25 \pm 5\%$.^[35] The sample was placed outside the window of the chamber and the window was

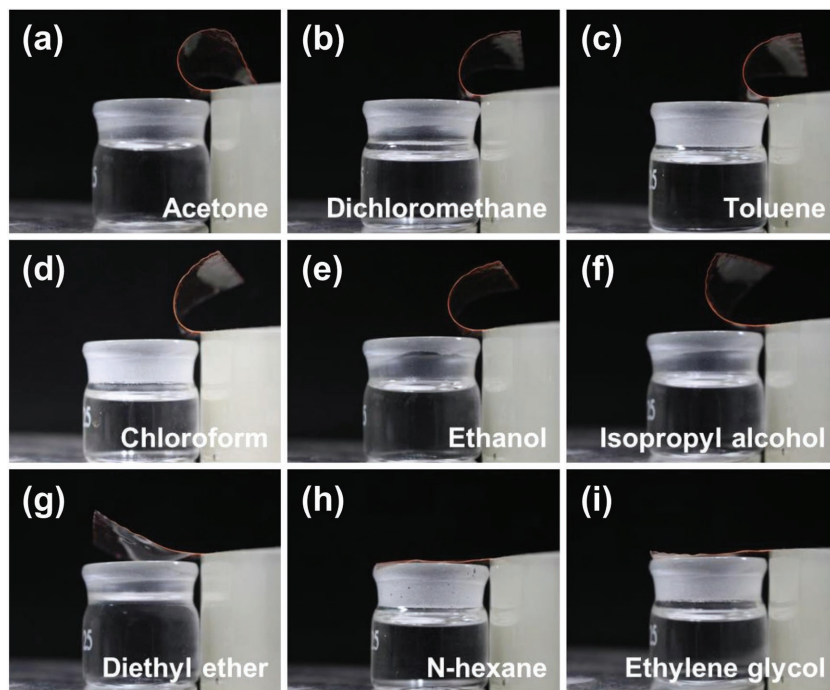


Figure 7. a–g) The PEG-DA hydrogel films could be driven by the vapors of volatile polar solvents. h) Nonpolar solvents and i) nonvolatile solvent could not drive the film.

covered by a coverslip; when the coverslip was removed, the window was open and the film was driven to bend by the relative humidity difference inside and outside the chamber.

Characterization: The surface morphologies of the PEG-DA films were characterized by using atomic force microscopy (AFM, Dimension Icon, Bruker Corporation) in the tapping mode. The FT-IR spectra were recorded by using an FTIR NICOLET 6700 spectrometer. The steady-state absorption and emission spectra of the rhodamine B:PEG-DA hydrogel films under different relative humidity were measured by using UV-vis spectrophotometer (UV-2550, SHIMADZU) and fluorescence spectrophotometer (F-4600, HITACHI).

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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